Implications of gauge freedom for nonrelativistic quantum electrodynamics

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Gauge freedom in quantum electrodynamics (QED) outside of textbook regimes is reviewed. It is emphasized that QED subsystems are defined relative to a choice of gauge. Each definition uses different gauge-invariant observables. This relativity is eliminated only if a sufficient number of Markovian and weak-coupling approximations are employed. All physical predictions are gauge invariant, including subsystem properties such as photon number and entanglement. However, subsystem properties naturally differ for different physical subsystems. Gauge ambiguities arise not because it is unclear how to obtain gauge-invariant predictions, but because it is not always clear which physical observables are the most operationally relevant. The gauge invariance of a prediction is necessary but not sufficient to ensure its operational relevance. It is shown that, in controlling which gauge-invariant observables are used to define a material system, the choice of gauge affects the balance between the material system's localization and its electromagnetic dressing. Various implications of subsystem gauge relativity for deriving effective models, for describing time-dependent interactions, for photodetection theory, and for describing matter within a cavity are reviewed.

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I. INTRODUCTION

Traditional regimes of light-matter physics involve relatively small values of a ratio r that compares, in a qualitative sense, the interaction strength to the energies characterizing the bare light and matter subsystems (Devoret, Girvin, and Schoelkopf, 2007). Over the past two decades, however, much more extreme light-matter interaction regimes have become an important topic in both applied and fundamental physics. In the simplest case of a two-level emitter coupled to a single photonic mode, the so-called ultrastrong-coupling regime ($r \gtrsim 0.1$) is typically taken as the point at which the rotating-wave approximation certainly breaks down, incurring a departure from Jaynes-Cummings (Jaynes and Cummings, 1963) physics. This regime has now been realized in a relatively large range of experimental platforms; recent reviews were given by Forn-Díaz *et al.* (2019) and Kockum *et al.* (2019). Even values $r \gtrsim 1$, which define the so-called deep-strong coupling regime, have now been realized both in superconducting circuits (Yoshihara *et al.*, 2017) and via plasmonic nanoparticle crystals (Mueller *et al.*, 2020).

Beyond those systems in which only a few photonic modes dominate, there are now diverse multimode photonic systems in which non-Markovian effects may become significant. These platforms include materials within dielectric and metallic environments, which may be uniform or nanostructured (Ma *et al.*, 2021), superconducting circuits coupled to transmission lines (Forn-Díaz *et al.*, 2017), solid-state systems (Nazir and McCutcheon, 2016; de Vega and Alonso, 2017), and cavity-molecule systems that offer a promising means by which to control chemical processes (Hertzog *et al.*, 2019). Experimental progress in ultrafast light-matter interactions is also continuing steadily. Femtosecond laser pulses offer the potential to control bare charges on ultrafast timescales (Ciappina *et al.*, 2017), while subcycle ultrastrong light-matter interaction switching was achieved some time ago (Günter *et al.*, 2009).

Recent reviews (Forn-Díaz *et al.*, 2019; Kockum *et al.*, 2019; Boité, 2020) of light-matter physics outside of weak-coupling regimes have focused on effective models and new theoretical methods, which are required because standard weak-coupling quantum optics cannot be applied. Despite new methods, our understanding continues to be based on processes involving real and virtual bare quanta, which can vary significantly with the form of the model considered. Nonstandard regimes where weak-coupling theory breaks down are precisely where effective models that are only superficially motivated are liable to fall short. This necessitates an appraisal of the fundamental physics from first principles, as is the focus of this review. We focus specifically on the implications of QED's gauge-theoretic aspects.

Gauge freedom in ultrastrong-coupling and deep-strong-coupling QED has recently been investigated in a number of contexts, including the truncation of a material subsystem to a finite number of energy levels (De Bernardis, Jaako, and Rabl, 2018; De Bernardis *et al.*, 2018; Roth, Hassler, and DiVincenzo, 2019; Stefano *et al.*, 2019; Stokes and Nazir, 2019, 2020a, 2020b, 2021b; Garziano *et al.*, 2020; Taylor *et al.*, 2020; Ashida, İmamoğlu, and Demler, 2021; Settineri *et al.*, 2021), time-dependent interactions (Stefano *et al.*, 2019; Settineri *et al.*, 2021; Stokes and Nazir, 2021b), Dicke model superradiance (De Bernardis, Jaako, and Rabl, 2018; Garziano *et al.*, 2020; Stokes and Nazir, 2020b), and photodetection theory (Settineri *et al.*, 2021).

Gauge freedom in QED implies a relativity in the assignment of physical meaning to the vectors and operators that represent states and observables. This is akin to the relativity encountered in theories of space and time. For example, the time interval Δt_X between two events x and y as measured by a clock at rest in frame X does not predict the outcome Δt_Y of measuring the time between x and y in a comoving frame Y. We have $\Delta t_X \approx \Delta t_Y$ only if the relativistic mixing incurred by the Lorentz transformation from X to Y can be ignored. Otherwise, we must recognize that we have two different predictions Δt_X and Δt_Y for two different experiments, one in frame X and one in frame Y. We do know, however, which prediction corresponds to which experiment, that is, we always know which prediction is relevant. This is determined by the rest frame of the clock; i.e., it is determined by the apparatus.

In the same way that intervals in space and time can be defined only relative to an inertial frame in Minkowski spacetime, light and matter quantum subsystems can be defined only relative to a gauge frame in Hilbert space. Unlike in special relativity, where it is straightforward to identify which predictions of space and time intervals are relevant in which situations, in QED there are a number of conceptual subtleties regarding the identification of the most relevant theoretical subsystems. The problem is closely related to the interpretation of virtual processes and particles, an aspect of light-matter physics that already possesses a long history of theoretical studies predominantly confined thus far to the weak-coupling regime. Such studies possess significant overlap with the quantum theory of measurement (Dalibard, Dupont-Roc, and Cohen-Tannoudji, 1982; Passante, Compagno, and Persico, 1985; Drummond, 1987; Compagno, Passante, and Persico, 1988a, 1988b, 1990, 1991, 1995; Stokes, 2012) as well as with the identification of local fields and causal signal propagation (Fermi, 1932; Cohen-Tannoudji, Dupont-Roc, and

Grynberg, 1989; Biswas *et al.*, 1990; Buchholz and Yngvason, 1994; Milonni, James, and Fearn, 1995; Power and Thirunamachandran, 1997, 1999a).

The primary purpose of this review is to identify what gauge ambiguities occur beyond the regimes traditionally considered in quantum optics and to clarify how they arise. In Sec. II we begin with a pedagogical introduction to gauge freedom. We then provide a rigorous derivation of arbitrary-gauge (nonrelativistic) QED using the principles of modern gauge-field theory, showing that the implications of gauge freedom discussed in Sec. II.F onward are a fundamental feature. They are not in any way an artifact of approximations or simplifications. In particular, we emphasize that gauge ambiguities arise not because it is unclear how to obtain gauge-invariant predictions, but because it is not always clear which gauge-invariant subsystems are *operationally relevant*. In Sec. II.F we address a number of common pitfalls related to gauge freedom in QED.

In Sec. III we introduce the notion of *subsystem gauge relativity*. We explain its relation to gauge invariance, identify the regimes within which it is important, and discuss its implications. In Sec. IV we review the theoretical background for the implementation of material level truncations (De Bernardis *et al.*, 2018; Roth, Hassler, and DiVincenzo, 2019; Stefano *et al.*, 2019; Stokes and Nazir, 2019, 2020a, 2020b; Ashida, İmamoğlu, and Demler, 2021), noting that the resulting gauge noninvariance is prosaic because it can always be avoided by avoiding the truncation. We review various proposals for obtaining two-level models, along with their varying degrees of accuracy in different regimes, as well as their significance for understanding gauge ambiguities.

In Sec. V we discuss time-dependent interactions. We first review the QED S-matrix formalism. Here subsystem gauge relativity does not occur, due to the condition of adiabatic interaction switching, which implies strict conservation of the bare energy h, where $H = h + V$ is the full Hamiltonian and V is the interaction Hamiltonian. We directly show that conventional weak-coupling and Markovian approximations mimic the S matrix, enforcing the conservation of h and thereby eliminating subsystem gauge relativity. In this sense, these traditional regimes are *gauge nonrelativistic*. In contrast, it is shown that when describing non-Markovian and strong-coupling effects subsystem gauge relativity cannot be ignored.

In Sec. VI we consider photodetection theory. We emphasize that gauge ambiguities arise because it is not always clear that any one definition of photon is always the most operationally relevant. For example, the Coulomb-gauge definition has recently been preferred in the ultrastrong-coupling light-matter physics literature (Stefano *et al.*, 2019; Settineri *et al.*, 2021). However, as has been known for some time, certain predictions, such as the natural line shape of spontaneous emission, have been found to be closer to experiment if photons are defined relative to the multipolar gauge (Power and Zienau, 1959; Fried, 1973; Davidovich and Nussenzveig, 1980; Milonni, Cook, and Ackerhalt, 1989; Woolley, 2000; Stokes, 2013).

We identify how the definitions of the subsystems, as controlled by the choice of gauge, are related to photodetection divergences (Drummond, 1987; Stokes *et al.*, 2012). We determine the relation between subsystem gauge relativity and electromagnetic dressing. We extend these considerations to cavity QED beyond standard regimes and discuss how subsystem gauge relativity relates to weak measurements of intracavity subsystems and to ground-state superradiance. We mention our outlook for predictions regarding extracavity fields. Finally, we summarize our work in Sec. VIII.

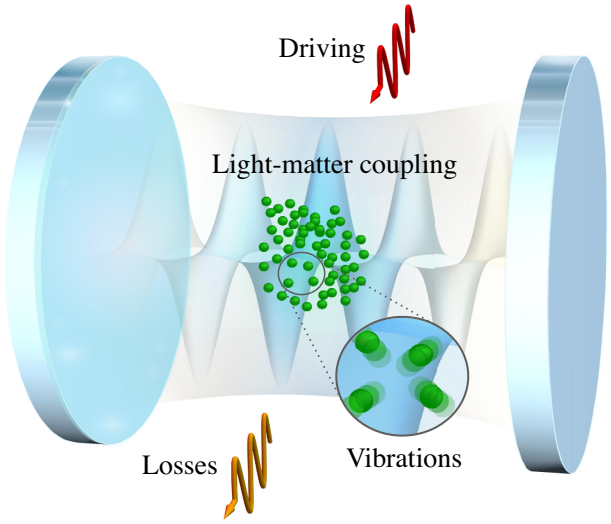

FIG. 1. Material systems, such as atoms or molecules, confined within an electromagnetic cavity that enhances the light-matter coupling. Internal vibrational interactions may also be strong and non-Markovian. Driving via laser light may take many forms, including the use of ultrafast and strong pulses. Losses within such systems may be complex including direct emission to external modes, as well as leakage through the cavity mirrors.

Throughout this review we use natural units such that $\hbar = c = \epsilon_0 = \mu_0 = 1$. The elementary electric charge is $e = \sqrt{4\pi\alpha_{\rm fs}}$, where $\alpha_{\rm fs}$ is the fine-structure constant. Unless otherwise stated, latin characters $(i, j, k, \ldots)$ denote Cartesian components of vectors whenever they appear as subscripts, and we adopt the summation convention for repeated Cartesian indices. The imaginary unit is also denoted as i (not a subscript). We use the notation $\dot{f}(t)$ as shorthand for the total derivative $df(t)/dt$.

II. GAUGE FREEDOM AND GAUGE FIXING

Quantum electrodynamics is the underpinning theory that describes all physical interactions occurring from the atomic scale upward, until gravitation becomes significant. Modern light-matter physics encompasses an extremely broad and diverse range of natural and artificial systems with numerous interactions that span a large parameter space. Dividing composite systems into quantum subsystems that emit, absorb, and exchange photons remains the basic conceptual framework used to understand light-matter physics, but beyond traditional regimes new challenges arise, both conceptual and technical. QED's gauge freedom becomes important because the choice of gauge controls the physical nature of the adopted theoretical quantum subsystem decomposition.

Weak-coupling theory will typically break down when one deals with complex or artificial systems of the type depicted in

Fig. 1. However, to identify and understand the challenges faced in as simple a setting as possible, we begin by revisiting the case of elementary charged particles in free space. Although sound treatments can be found in various textbooks (Cohen-Tannoudji, Dupont-Roc, and Grynberg, 1989; Craig and Thirunamachandran, 1998), the role and significance of gauge freedom is less widely understood and has even recently been debated (Rousseau and Felbacq, 2017, 2018; Andrews *et al.*, 2018; Vukics, Kónya, and Domokos, 2021). This motivates a collation of present understanding and the provision of a coherent overview. Section II.C summarizes the results of a rigorous derivation of arbitrary-gauge nonrelativistic QED that uses the principles of modern gauge-field theory, with further details given in note II of the Supplemental Material (245). We define the gauge principle, gauge freedom, gauge-symmetry transformations, gauge-fixing transformations, and gauge invariance. We address conceptual issues and common pitfalls.

A. A single stationary atom in standard gauges

Consider a single charge q with position $\mathbf{r}$ bound to a fixed charge $-q$ at the origin $\mathbf{0}$ of our chosen inertial frame. The charge and current densities are

$$\rho(\mathbf{x}) = -q\delta(\mathbf{x}) + q\delta(\mathbf{x}-\mathbf{r}), \tag{1}$$

$$\mathbf{J}(\mathbf{x}) = \frac{q}{2}[\dot{\mathbf{r}}\delta(\mathbf{x}-\mathbf{r}) + \delta(\mathbf{x}-\mathbf{r})\dot{\mathbf{r}}], \tag{2}$$

such that $\partial_t\rho = -\boldsymbol{\nabla}\cdot\mathbf{J}$. Note that in quantum theory $[r_i, \dot{r}_j] \neq 0$, so the expression for the current must be symmetrized. The material fields ρ and $\mathbf{J}$ together with the electric and magnetic fields $\mathbf{E}$ and $\mathbf{B}$ exhaustively assign material and electromagnetic properties to each event $x = (t, \mathbf{x})$ in spacetime. Gauge freedom can be understood as a many-to-one correspondence between auxiliary mathematical objects used to express the theory and the physical observables ρ, $\mathbf{J}$, $\mathbf{E}$, and $\mathbf{B}$. It is hailed by the occurrence of nondynamical constraints $\boldsymbol{\nabla}\cdot\mathbf{B} = 0$ and $\boldsymbol{\nabla}\cdot\mathbf{E} = \rho$, which imply redundancy within the formalism. The scalar and vector potentials A_0 and $\mathbf{A}$ are defined as

$$\mathbf{E} = -\boldsymbol{\nabla}A_0 - \partial_t\mathbf{A}, \tag{3}$$

$$\mathbf{B} = \boldsymbol{\nabla}\times\mathbf{A}, \tag{4}$$

which imply that the homogeneous Maxwell equations $\boldsymbol{\nabla}\cdot\mathbf{B} = 0$ and $\partial_t\mathbf{B} = -\boldsymbol{\nabla}\times\mathbf{E}$ are automatically satisfied. The inhomogeneous constraint $\boldsymbol{\nabla}\cdot\mathbf{E} = \rho$ (Gauss's law) must be imposed within the theory, while the remaining inhomogeneous equation is dynamical ($\partial_t\mathbf{E} = \boldsymbol{\nabla}\times\mathbf{B} - \mathbf{J}$) (Maxwell-Ampère's law). This is an equation of motion that must be produced by any satisfactory Lagrangian or Hamiltonian description. The electric and magnetic fields are invariant under the *gauge transformation*

$$\mathbf{A}' = \mathbf{A} + \boldsymbol{\nabla}\chi, \tag{5}$$

$$A_0' = A_0 - \partial_t\chi, \tag{6}$$

where χ is arbitrary.

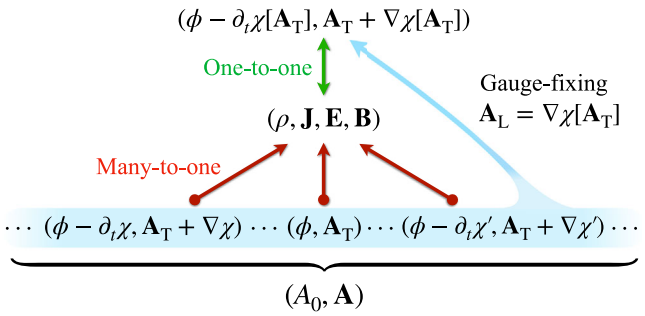

FIG. 2. Schematic representation of gauge redundancy in electrodynamics. The central potential pair is $(\phi, \mathbf{A}_\mathrm{T})$ (Coulomb gauge). The blue (shaded) band represents an uncountable infinity of potential pairs, all of which produce the same physical fields and all of which are related to each other by gauge transformation. Gauge fixing can be achieved by setting the redundancy that causes this many-to-one correspondence $\mathbf{A}_\mathrm{L}$ equal to a known functional χ of the fixed and gauge-invariant object $\mathbf{A}_\mathrm{T} = (\boldsymbol{\nabla}\times)^{-1}\mathbf{B}$. Afterward, the map between the chosen fixed potential pair and the physical fields is invertible (one to one). The fixed potentials can be written as known functions of $(\rho, \mathbf{J}, \mathbf{E}, \mathbf{B})$, while $\mathbf{E}$ and $\mathbf{B}$ are also known functions of the fixed potentials [Eqs. (3) and (4)].

An unconstrained Hamiltonian description in terms of the potentials A_0 and $\mathbf{A}$ requires elimination of gauge redundancy. Recall that the Helmholtz decomposition of a square-integrable vector field $\mathbf{V}$ into transverse and longitudinal fields ($\mathbf{V} = \mathbf{V}_\mathrm{T} + \mathbf{V}_\mathrm{L}$) is unique. The transverse and longitudinal components satisfy $\boldsymbol{\nabla}\cdot\mathbf{V}_\mathrm{T} = 0$ and $\boldsymbol{\nabla}\times\mathbf{V}_\mathrm{L} = \mathbf{0}$. Transverse and longitudinal delta functions (dyadics) are defined by the nonlocal conditions

$$\mathbf{V}_{\mathrm{L,T}}(\mathbf{x}) = \int d^3x'\,\delta^{\mathrm{L,T}}(\mathbf{x}-\mathbf{x}')\cdot\mathbf{V}(\mathbf{x}'). \tag{7}$$

The process of *gauge fixing* eliminates the mathematical redundancy within the formalism by specifying all freely choosable objects as known functions of objects that cannot be freely chosen (Fig. 2). Since the curl of the gradient is identically zero, the transverse vector potential $\mathbf{A}_\mathrm{T}$ is gauge invariant; that is, if $\mathbf{A}' = \mathbf{A} + \boldsymbol{\nabla}\chi$, then $\mathbf{A}_\mathrm{T}' \equiv \mathbf{A}_\mathrm{T}$, which cannot be freely chosen. Gauge freedom is therefore the freedom to choose the longitudinal vector potential $\mathbf{A}_\mathrm{L} = \boldsymbol{\nabla}\chi$, where $\mathbf{A} = \mathbf{A}_\mathrm{T} + \boldsymbol{\nabla}\chi$. In note I of the Supplemental Material (245), this gauge freedom is related to the U(1) phase of material wave functions and electromagnetic wave functionals.

One of the most commonly chosen gauges is the Coulomb gauge defined by the choice $\mathbf{A}_\mathrm{L} = \mathbf{0}$, such that $\mathbf{A} = \mathbf{A}_\mathrm{T}$. From Gauss's law $\boldsymbol{\nabla}\cdot\mathbf{E} = \rho$ and Eq. (3), it follows that in the Coulomb gauge the scalar potential A_0 coincides with the Coulomb potential defined by

$$\phi(\mathbf{x}) = -\boldsymbol{\nabla}^{-2}\rho(\mathbf{x}) = \int d^3x'\,\frac{\rho(\mathbf{x}')}{4\pi|\mathbf{x}-\mathbf{x}'|}, \tag{8}$$

where the kernel $1/4\pi|\mathbf{x}|$ is the Green's function for the Laplacian; $\boldsymbol{\nabla}^2(1/4\pi|\mathbf{x}|) = -\delta(\mathbf{x})$. Specifying $\mathbf{A} = \mathbf{A}_\mathrm{T}$ and $A_0 = \phi$ is an example of gauge fixing.

The other commonly chosen gauge in nonrelativistic electrodynamics is the Poincaré (multipolar) gauge defined by $\mathbf{x}\cdot\mathbf{A}(\mathbf{x})=0$. This is the Coulomb-gauge condition applied in reciprocal space. More generally, we may define the arbitrary-gauge potential

$$\mathbf{A}_\alpha(\mathbf{x})=\mathbf{A}_{\mathrm{T}}(\mathbf{x})-\alpha\boldsymbol{\nabla}\int_0^1 d\lambda\,\mathbf{x}\cdot\mathbf{A}_{\mathrm{T}}(\lambda\mathbf{x}),\tag{9}$$

where the value of α selects the gauge by specifying $\mathbf{A}_{\mathrm{L}}$. The Coulomb and multipolar gauges are now simply special cases given by $\alpha=0$ and $\alpha=1$, respectively (Stokes and Nazir, 2019, 2020b, 2021b). Equation (9) can be written as

$$\mathbf{A}_\alpha=\mathbf{A}_{\mathrm{T}}+\nabla\chi_\alpha,\tag{10}$$

where

$$\chi_\alpha(\mathbf{x})=\int d^3x'\,\mathbf{g}_{\mathrm{T}\alpha}(\mathbf{x}',\mathbf{x})\cdot\mathbf{A}_{\mathrm{T}}(\mathbf{x}'),\tag{11}$$

$$\mathbf{g}_{\mathrm{T}\alpha}(\mathbf{x}',\mathbf{x})=-\alpha\int_0^1 d\lambda\,\mathbf{x}\cdot\delta^{\mathrm{T}}(\mathbf{x}'-\lambda\mathbf{x}).\tag{12}$$

For each value of α, all freely choosable objects are known functions of objects that cannot be freely chosen. More precisely, the theory has been expressed entirely in terms of $\mathbf{A}_{\mathrm{T}}$ (Fig. 2), which serves as an elementary dynamical coordinate. Different values of α provide different choices of $\mathbf{A}_{\mathrm{L}}$ as different fixed functionals of the coordinate $\mathbf{A}_{\mathrm{T}}$. Parametrization via α in this way does not exhaust all possible gauge choices. It does, however, allow us to provide clear definitions of *gauge invariance* and *gauge relativity*, as is done in Sec. III.B. In note II of the Supplemental Material (245) we provide a more general encoding of gauge freedom, and the results are summarized in Sec. II.C.

It is useful to define the polarization field $\mathbf{P}$ using the equation $-\boldsymbol{\nabla}\cdot\mathbf{P}=\rho$, which specifies $\mathbf{P}_{\mathrm{L}}$ uniquely but leaves $\mathbf{P}_{\mathrm{T}}$ an essentially arbitrary transverse field. We are free to define $\mathbf{P}_\alpha\coloneqq\mathbf{P}_{\mathrm{L}}+\mathbf{P}_{\mathrm{T}\alpha}$, where $\mathbf{P}_{\mathrm{T}\alpha}$ is called the α-gauge transverse polarization defined by the condition

$$\int d^3x\,\rho(\mathbf{x})\chi_\alpha(\mathbf{x})=-\int d^3x\mathbf{P}_{\mathrm{T}\alpha}(\mathbf{x})\cdot\mathbf{A}_{\mathrm{T}}(\mathbf{x}).\tag{13}$$

It follows from Eqs. (11) and (12) that we may set

$$\begin{aligned}\mathbf{P}_{\mathrm{T}\alpha}(\mathbf{x})&=-\int d^3x'\,\mathbf{g}_{\mathrm{T}\alpha}(\mathbf{x},\mathbf{x}')\rho(\mathbf{x}')\\&=\alpha q\int_0^1 d\lambda\,\mathbf{r}\cdot\delta^{\mathrm{T}}(\mathbf{x}-\lambda\mathbf{r})=\alpha\mathbf{P}_{\mathrm{T}}(\mathbf{x}),\end{aligned}\tag{14}$$

where $\mathbf{P}_{\mathrm{T}}\coloneqq\mathbf{P}_{\mathrm{T}1}$ is the multipolar transverse polarization. According to these definitions, in the Coulomb gauge we have $\mathbf{P}_{\mathrm{T}0}=\mathbf{0}$, and therefore $\mathbf{P}_0=\mathbf{P}_{\mathrm{L}}$. In the multipolar gauge we have

$$\mathbf{P}_1(\mathbf{x})\coloneqq\mathbf{P}_{\mathrm{T}1}(\mathbf{x})+\mathbf{P}_{\mathrm{L}}(\mathbf{x})=q\int_0^1 d\lambda\,\mathbf{r}\delta(\mathbf{x}-\lambda\mathbf{r}).\tag{15}$$

This field specifies a straight line of singular dipole moment density that stretches from the charge $-q$ at $\mathbf{0}$ to the dynamical charge q at $\mathbf{r}$.

We now provide a canonical (Hamiltonian) quantum description. Typically this would be derived from a suitable Lagrangian and the gauge would be fixed from the outset. However, our only requirement is that the theory produces the correct Maxwell-Lorentz system of equations, and it can therefore be obtained through a series of *Ansätze*. A rigorous and more general derivation of arbitrary-gauge QED is given using modern gauge-field theory in notes II–IV of the Supplemental Material (245).

We proceed by writing the total energy of the system as a sum of the kinetic and electromagnetic energies as follows:

$$\begin{aligned}E&=\frac{1}{2}m\dot{\mathbf{r}}^2+\frac{1}{2}\int d^3x(\mathbf{E}^2+\mathbf{B}^2)\\&=\frac{1}{2}m\dot{\mathbf{r}}^2+U(\mathbf{r})+V_{\mathrm{self}}(\mathbf{r})+\frac{1}{2}\int d^3x(\mathbf{E}_{\mathrm{T}}^2+\mathbf{B}^2),\end{aligned}\tag{16}$$

where $\mathbf{E}_{\mathrm{T}}=-\partial_t\mathbf{A}_{\mathrm{T}}$ and

$$U(\mathbf{r})+V_{\mathrm{self}}=\frac{1}{2}\int d^3x\mathbf{E}_{\mathrm{L}}^2\equiv\frac{1}{2}\int d^3x\mathbf{P}_{\mathrm{L}}^2.\tag{17}$$

In Eqs. (16) and (17) $U(\mathbf{r})=-q^2/4\pi|\mathbf{r}|$ is the Coulomb energy binding the charges q and $-q$, while V_{self} is the sum of the infinite Coulomb self-energies of each individual charge. Equation (17) is obtained by solving Gauss's law $\boldsymbol{\nabla}\cdot\mathbf{E}=\rho$, which yields $\mathbf{E}_{\mathrm{L}}=-\mathbf{P}_{\mathrm{L}}=-\boldsymbol{\nabla}\phi$, with ϕ as defined in Eq. (8).

The canonical operators $\mathbf{y}=\{\mathbf{r},\mathbf{A}_{\mathrm{T}},\mathbf{p},\boldsymbol{\Pi}\}$ in terms of which we express the theory satisfy

$$[r_i,p_j]=i\delta_{ij},\tag{18}$$

$$[A_{\mathrm{T},i}(\mathbf{x}),\Pi_{\mathrm{T},j}(\mathbf{x}')]=i\delta_{ij}^{\mathrm{T}}(\mathbf{x}-\mathbf{x}'),\tag{19}$$

while all other commutators between canonical operators vanish. Here we assume these commutators and show that they yield the correct result. A systematic derivation is given in note IX of the Supplemental Material (245). Since energy generates translations in time, the Hamiltonian that we seek must equal the total energy expressed in terms of the canonical operators $[H(\mathbf{y})=E]$. Given this constraint, we must now make suitable *Ansätze* for the velocities $\dot{\mathbf{r}}\equiv\dot{\mathbf{r}}(\mathbf{y})$ and $\partial_t\mathbf{A}_{\mathrm{T}}\equiv\partial_t\mathbf{A}_{\mathrm{T}}(\mathbf{y})$. We require that upon substitution into the right-hand side of Eq. (16) our *Ansätze* define a Hamiltonian $H(\mathbf{y})$, for which the Heisenberg equation $\partial_t O=-i[O,H]$ together with Eqs. (18) and (19) yields the correct Maxwell-Lorentz equations.

Since we want to provide a Hamiltonian description in an arbitrary gauge, we make the arbitrary-gauge minimal coupling *Ansätze*

$$m\dot{\mathbf{r}}=\mathbf{p}-q\mathbf{A}_\alpha(\mathbf{r}),\tag{20}$$

$$\partial_t\mathbf{A}_{\mathrm{T}}=\boldsymbol{\Pi}+\mathbf{P}_{\mathrm{T}\alpha}.\tag{21}$$

Note that minimal coupling is not synonymous with any one gauge and, in particular, it is not synonymous with the

Coulomb gauge despite the fact that the Coulomb-gauge Hamiltonian is often called the minimal coupling Hamiltonian. This point is discussed in more detail in Sec. II.F.6. From Eqs. (16), (20), and (21) we obtain

$$E = \frac{1}{2m}[\mathbf{p} - q\mathbf{A}_\alpha(\mathbf{r})]^2 + U(\mathbf{r}) + V_{\text{self}} + \frac{1}{2}\int d^3x[(\mathbf{\Pi} + \mathbf{P}_{\text{T}\alpha})^2 + (\boldsymbol{\nabla}\times\mathbf{A}_{\text{T}})^2] =: H_\alpha(\mathbf{y}). \quad (22)$$

Equation (22) defines the arbitrary-gauge Hamiltonian H_α, which coincides with the one derived by Stokes and Nazir (2019, 2020b, 2021b). The canonical commutation relation (CCR) algebra, Eqs. (18) and (19), yields

$$\mathbf{p} - q\mathbf{A}_\alpha(\mathbf{r}) = -im[\mathbf{r}, H_\alpha], \quad (23)$$

$$\mathbf{\Pi}(\mathbf{x}) + \mathbf{P}_{\text{T}\alpha}(\mathbf{x}) = -i[\mathbf{A}_{\text{T}}(\mathbf{x}), H_\alpha]. \quad (24)$$

Equations (23) and (24) show that the *Ansätze* in Eqs. (20) and (21) are self-consistent because they are reobtained using the Heisenberg equation. It is a straightforward exercise to verify that H_α does indeed yield the correct Maxwell-Lorentz system of equations for any choice of gauge α.

It is readily verified that Hamiltonians of different fixed gauges α and α' are unitarily equivalent,

$$H_{\alpha'} = R_{\alpha\alpha'}H_\alpha R^\dagger_{\alpha\alpha'}, \quad (25)$$

where $R_{\alpha\alpha'}$ is called a *gauge-fixing transformation* and is defined by (Lenz *et al.*, 1994; Chernyak and Mukamel, 1995; Stokes, 2012; Stokes and Nazir, 2019, 2020b)

$$R_{\alpha\alpha'} := \exp\left(i\int d^3x[\mathbf{P}_{\text{T}\alpha}(\mathbf{x}) - \mathbf{P}_{\text{T}\alpha'}(\mathbf{x})]\cdot\mathbf{A}_{\text{T}}(\mathbf{x})\right) = \exp\{-iq[\chi_\alpha(\mathbf{r}) - \chi_{\alpha'}(\mathbf{r})]\}, \quad (26)$$

in which the second equality follows from Eq. (13). We emphasize that the definition of gauge freedom continues to be the freedom to choose α, which specifies $\mathbf{A}_{\text{L}}$. It therefore constitutes the freedom to transform between distinct minimal coupling prescriptions as

$$R_{\alpha\alpha'}[\mathbf{p} - q\mathbf{A}_\alpha(\mathbf{r})]R^\dagger_{\alpha\alpha'} = \mathbf{p} - q\mathbf{A}_{\alpha'}(\mathbf{r}), \quad (27)$$

$$R_{\alpha\alpha'}(\mathbf{\Pi} + \mathbf{P}_{\text{T}\alpha})R^\dagger_{\alpha\alpha'} = \mathbf{\Pi} + \mathbf{P}_{\text{T}\alpha'}, \quad (28)$$

from which Eq. (25) follows. The effect of the transformation has been the replacement $(\mathbf{A}_\alpha, \mathbf{P}_{\text{T}\alpha}) \to (\mathbf{A}_{\alpha'}, \mathbf{P}_{\text{T}\alpha'})$, which constitutes a gauge transformation from the fixed gauge α to the fixed gauge α'. The reason that Eq. (28) occurs is that in Eq. (9) we chose to fix the gauge $\mathbf{A}_{\text{L}}$ as a functional of $\mathbf{A}_{\text{T}}$, which generates translations in $\mathbf{\Pi}$. The gauge freedom inherent in the polarization is discussed further in note I.2 of the Supplemental Material (245). Note that since $Uf(O)U^\dagger = f(UOU^\dagger)$ for any unitary transformation U, suitably well-defined function f, and operator O, Eqs. (27) and (28) are necessary and sufficient to define how arbitrary functions of the canonical operators transform under a gauge transformation.

For one to implement the gauge transformation $\mathbf{p} - q\mathbf{A}(\mathbf{r}) \to \mathbf{p} - q[\mathbf{A}(\mathbf{r}) + \boldsymbol{\nabla}\chi(\mathbf{r})]$, the canonical momentum must transform as $e^{iq\chi(\mathbf{r})}\mathbf{p}e^{-iq\chi(\mathbf{r})} = \mathbf{p} - q\boldsymbol{\nabla}\chi(\mathbf{r})$, which states that $\mathbf{r}$ generates translations in $\mathbf{p}$. This property relies upon the canonical commutation relation in Eq. (18). Equation (27), in particular, features the gauge-fixing transformation $R_{\alpha\alpha'} = e^{-iq[\chi_\alpha(\mathbf{r}) - \chi_{\alpha'}(\mathbf{r})]}$. As recognized long ago by Weyl (Weyl, 1927), the CCR algebra cannot be supported by a finite-dimensional Hilbert space. Thus, retaining only a finite number of material energy levels will ruin gauge invariance. Material truncation is discussed in detail in Sec. IV.

B. Electric-dipole approximation

The electric-dipole approximation (EDA) of the theory presented in Sec. II.A can be performed while preserving all kinematic and algebraic relations of the theory such that gauge invariance is also preserved. We define the Fourier transform of a field f by $\tilde{f}(\mathbf{k}) := \int d^3x f(\mathbf{x})e^{-i\mathbf{k}\cdot\mathbf{x}}/\sqrt{(2\pi)^3}$. Considering the charge and current densities in Eqs. (1) and (2), the EDA (also known as the long-wavelength approximation) is defined by retaining only the leading contributions after performing the expansion $e^{-i\mathbf{k}\cdot\mathbf{r}} = 1 - i\mathbf{k}\cdot\mathbf{r} + \ldots$. This gives $\tilde{\rho}(\mathbf{k}) \approx -iq\mathbf{k}\cdot\mathbf{r}/\sqrt{(2\pi)^3}$, $\rho(\mathbf{x}) \approx -q\mathbf{r}\cdot\boldsymbol{\nabla}\delta(\mathbf{x})$, $\mathbf{J}(\mathbf{x}) \approx q\dot{\mathbf{r}}\delta(\mathbf{x})$, and $\mathbf{A}_{\text{T}}(\mathbf{r}) \approx \mathbf{A}_{\text{T}}(\mathbf{0})$, and in turn

$$P_{\text{T}\alpha,i}(\mathbf{x}) \approx \alpha q r_j \delta^{\text{T}}_{ij}(\mathbf{x}), \quad (29)$$

$$\chi_\alpha(\mathbf{r}) \approx -\alpha\mathbf{r}\cdot\mathbf{A}_{\text{T}}(\mathbf{0}), \quad (30)$$

$$\mathbf{A}_\alpha(\mathbf{r}) \approx (1-\alpha)\mathbf{A}_{\text{T}}(\mathbf{0}). \quad (31)$$

When Eqs. (29)–(31) are substituted into Eq. (22), the α-gauge Hamiltonian in the EDA is obtained. Similarly, the unitary gauge-fixing transformation $R_{\alpha\alpha'}$ in Eq. (26) becomes

$$R_{\alpha\alpha'} = \exp[i(\alpha - \alpha')q\mathbf{r}\cdot\mathbf{A}_{\text{T}}(\mathbf{0})]. \quad (32)$$

Since unitarity is preserved, so too is gauge invariance; see Sec. III.B. Hamiltonians belonging to different gauges continue to be unitarily equivalent, as in Eq. (25).

Certain nonfundamental properties hold within (and only within) the EDA (Stokes and Nazir, 2020a). In particular, the gauge function χ_α in Eq. (11) becomes that of Eq. (30), which gives $\nabla\chi_1(\mathbf{r}) = -\mathbf{A}_{\text{T}}(\mathbf{0})$, such that $\mathbf{p} - q\mathbf{A}_1(\mathbf{r}) \approx \mathbf{p}$. Thus, letting $\alpha = 1$ on the left-hand side of Eq. (27), we obtain $R_{1\alpha}\mathbf{p}R^\dagger_{1\alpha} = \mathbf{p} - q\mathbf{A}_\alpha$, where $\mathbf{A}_\alpha := (1-\alpha)\mathbf{A}_{\text{T}}(\mathbf{0})$ is the EDA of $\mathbf{A}_\alpha(\mathbf{r})$. Within the full three-dimensional setting and without the EDA, this is impossible because for any differentiable function f we have $e^{-if(\mathbf{r})}\mathbf{p}e^{if(\mathbf{r})} = \mathbf{p} + \boldsymbol{\nabla}f(\mathbf{r})$. The gradient $\boldsymbol{\nabla}f$ is a longitudinal field such that we cannot have $\boldsymbol{\nabla}f(\mathbf{r}) = -q\mathbf{A}(\mathbf{r})$ for all $\mathbf{r}$, because $\mathbf{A}_{\text{T}}(\mathbf{r})$ is nonvanishing. The gauge transformation $e^{iqf(\mathbf{r})}[\mathbf{p} - q\mathbf{A}(\mathbf{r})]e^{-iqf(\mathbf{r})} = \mathbf{p} - q[\mathbf{A} + \boldsymbol{\nabla}f(\mathbf{r})]$ is fundamental and yields the result $R_{1\alpha}\mathbf{p}R^\dagger_{1\alpha} = \mathbf{p} - q\mathbf{A}_\alpha$ as an approximate special case in which we let $f = \chi_\alpha - \chi_1$ and perform the EDA.

C. Generalizations and the gauge principle

Modern gauge-field theories are understood to result from the *gauge principle* applied to a material field ψ. The principle states the following.

- The form of electromagnetic and other interactions should be invariant under the local action of a group $\mathcal{G}$ on the matter field ψ, written as $\psi'(x) = g(x) \cdot \psi(x)$. In QED $\mathcal{G} = \mathrm{U}(1)$ and $\psi'(x) = e^{iq\chi(x)}\psi(x)$, where χ is arbitrary.

In note I of the Supplemental Material (245), we review how gauge invariance can be understood as U(1)-phase invariance. In notes II–V of the Supplemental Material (245), we provide a general derivation of arbitrary-gauge nonrelativistic QED using the principles of modern gauge-field theory. The main results are summarized later. A sufficiently general expression of the theory that is suitable for our purposes results from encoding gauge freedom into the arbitrary transverse component $\mathbf{g}_{\mathrm{T}}$ of the Green's function $\mathbf{g}$ for the divergence operator,

$$\boldsymbol{\nabla} \cdot \mathbf{g}(\mathbf{x}, \mathbf{x}') \equiv \boldsymbol{\nabla} \cdot \mathbf{g}_{\mathrm{L}}(\mathbf{x}, \mathbf{x}') = \delta(\mathbf{x} - \mathbf{x}'), \tag{33}$$

$$\mathbf{g}_{\mathrm{L}}(\mathbf{x}, \mathbf{x}') = -\boldsymbol{\nabla} \frac{1}{4\pi|\mathbf{x} - \mathbf{x}'|}, \tag{34}$$

such that $\mathbf{g}_{\mathrm{T}} = \mathbf{g} - \mathbf{g}_{\mathrm{L}}$ is arbitrary.

We refer to the gauge specified by $\mathbf{g}_{\mathrm{T}}$ as the gauge g. The associated vector potential and polarization are

$$\begin{aligned} \mathbf{A}_g(\mathbf{x}) &= \mathbf{A}_{\mathrm{T}}(\mathbf{x}) + \boldsymbol{\nabla} \int d^3x' \mathbf{g}(\mathbf{x}', \mathbf{x}) \cdot \mathbf{A}_{\mathrm{T}}(\mathbf{x}') \\ &= \mathbf{A}_{\mathrm{T}}(\mathbf{x}) + \boldsymbol{\nabla}\chi_g(\mathbf{x}, [\mathbf{A}_{\mathrm{T}}]), \end{aligned} \tag{35}$$

$$\mathbf{P}_g(\mathbf{x}) = -\int d^3x' \mathbf{g}(\mathbf{x}, \mathbf{x}')\rho(\mathbf{x}'), \tag{36}$$

where

$$\chi_g(\mathbf{x}, [\mathbf{A}_{\mathrm{T}}]) = \int d^3x' \mathbf{g}(\mathbf{x}', \mathbf{x}) \cdot \mathbf{A}_{\mathrm{T}}(\mathbf{x}'). \tag{37}$$

The Hamiltonian in gauge g is

$$\begin{aligned} H_g = H(\mathbf{g}_{\mathrm{T}}) &= \frac{1}{2m}[\mathbf{p} - q\mathbf{A}_g(\mathbf{r})]^2 + U(\mathbf{r}) + V_{\mathrm{self}} \\ &\quad + \frac{1}{2}\int d^3x[(\boldsymbol{\Pi} + \mathbf{P}_{\mathrm{Tg}})^2 + (\boldsymbol{\nabla} \times \mathbf{A}_{\mathrm{T}})^2]. \end{aligned} \tag{38}$$

Hamiltonians H_g and H'_g are unitarily related by

$$H_{g'} = U_{gg'} H_g U^{\dagger}_{gg'}, \tag{39}$$

where

$$\begin{aligned} U_{gg'} &\coloneqq \exp\left(-i\int d^3x[\chi_g(\mathbf{x}, \mathbf{A}_{\mathrm{T}}) - \chi_{g'}(\mathbf{x}, \mathbf{A}_{\mathrm{T}})]\rho(\mathbf{x})\right) \\ &= \exp\left(i\int d^3x[\mathbf{P}_g(\mathbf{x}) - \mathbf{P}_{g'}(\mathbf{x})] \cdot \mathbf{A}_{\mathrm{T}}(\mathbf{x})\right) \end{aligned} \tag{40}$$

is a unitary gauge-fixing transformation from gauge g to gauge g'.

The theory is simplified by restricting $\mathbf{g}_{\mathrm{T}}$ as in Eq. (12) in terms of the gauge parameter α. The theory remains exact but reduces to the form presented in Sec. II.A; see note V of the Supplemental Material (245) for details. Gauge freedom becomes the freedom to choose the parameter α, which specifies $\mathbf{P}_{\mathrm{T}\alpha}$ and $\mathbf{A}_\alpha$ as in Eqs. (14) and (9), respectively (Stokes and Nazir, 2019, 2020b, 2021b). The Hamiltonian H_g in Eq. (38) becomes H_α as given in Eq. (22) and the gauge-fixing transformation $U_{gg'}$ in Eq. (40) becomes $R_{\alpha\alpha'}$ in Eq. (26). Hamiltonians belonging to different gauges are unitarily related as in Eq. (25).

Finally, we remark that the primary use of nonrelativistic QED lies in describing collections of charges partitioned into certain groups that we call atoms and molecules. The previously mentioned formalism describes a single hydrogen atom in which the positive charge $-q$ is assumed to be fixed (nondynamical). This is equivalent to describing the system using relative and center-of-mass coordinates instead of the charge coordinates themselves and assuming that the center of mass is fixed, with all center-of-mass couplings ignored. In note VI of the Supplemental Material (245), we provide the extension of this formalism to arbitrary charge distributions in the vicinity of fixed molecular centers (Craig and Thirunamachandran, 1998). In note VII of the Supplemental Material (245), we review the extension to linear dispersing and absorbing (macroscopic) dielectric media, which is a valuable tool in describing cavity QED systems (Knöll, Vogel, and Welsch, 1991; Gruner and Welsch, 1996; Dung, Knöll, and Welsch, 1998; Knoll, Scheel, and Welsch, 2003; Viviescas and Hackenbroich, 2003; Khanbekyan *et al.*, 2005); see also note XVII of the Supplemental Material (245). Concerning further extensions, we note that Wei, Shi-Bing, and Wei (2009) considered an anisotropic medium, and that Judge *et al.* (2013) considered a linear magneto-electric medium. Finally, we note that the use of this formalism in providing a microscopic description of electrons in crystal lattices is given in note VIII of the Supplemental Material (245).

D. Physical nature of the gauge function

We now seek to understand the ways in which different fixed-gauge formulations of QED differ. The gauge is selected by choosing χ_g. If χ_g is restricted in form as in Eq. (37), then the gauge is selected by choosing a concrete transverse function $\mathbf{g}_{\mathrm{T}}$. The gauge choice directly specifies two basic quantities $\mathbf{A}_g$ and $\mathbf{P}_{\mathrm{T}g}$. This in turn specifies the physical nature of the canonical momenta $\mathbf{p}$ and $\boldsymbol{\Pi}$, which together with $\mathbf{r}$ and $\mathbf{A}_{\mathrm{T}}$ define the quantum subsystems conventionally termed "matter" and "light." The importance of this fact is described in detail in Secs. II.E–III.C.

1. Path-dependent solution

The Green's function $\mathbf{g}$ is defined by Eq. (33). We have seen that the two most commonly chosen gauges of nonrelativistic QED can be linearly interpolated between via a parameter α, with $\alpha = 1$ specifying the multipolar gauge that possesses a

straight line of singular polarization stretching between the charges. This is a special case of the following more general path- and origin-dependent solution discussed by Woolley (1998, 2020):

$$\mathbf{g}(\mathbf{x},\mathbf{x}') = \mathbf{g}_{\mathrm{L}}(\mathbf{x},\mathbf{o}) - \int_{C(\mathbf{o},\mathbf{x}')} d\mathbf{z}\delta(\mathbf{z}-\mathbf{x}), \tag{41}$$

where $C(\mathbf{o},\mathbf{x}')$ is any curve starting at the arbitrary origin $\mathbf{o}$ and ending at $\mathbf{x}'$. Verification of the solution is most easily achieved in Fourier space, where Eq. (33) becomes $i\mathbf{k}\cdot\tilde{\mathbf{g}}(\mathbf{k},\mathbf{x}) = e^{-i\mathbf{k}\cdot\mathbf{x}}/\sqrt{(2\pi)^3}$. Using Eq. (41), we obtain independent of C and $\mathbf{o}$

$$\sqrt{(2\pi)^3}i\mathbf{k}\cdot\tilde{\mathbf{g}}(\mathbf{k},\mathbf{x}) = e^{-i\mathbf{k}\cdot\mathbf{o}} - i\int_{\mathbf{k}\cdot\mathbf{o}}^{\mathbf{k}\cdot\mathbf{x}} du\, e^{-iu} = e^{-i\mathbf{k}\cdot\mathbf{x}}, \tag{42}$$

as required.

Substituting Eq. (33) into Eq. (36), we obtain the following g-gauge polarization field:

$$\mathbf{P}_g(\mathbf{x}) = -Q\mathbf{g}_{\mathrm{L}}(\mathbf{x},\mathbf{o}) + \int d^3x' \int_{C(\mathbf{o},\mathbf{x}')} d\mathbf{z}\delta(\mathbf{z}-\mathbf{x})\rho(\mathbf{x}'), \tag{43}$$

where the first term vanishes for a globally neutral system defined by $Q = \int d^3x\rho(\mathbf{x}) = 0$. An important class of solutions is given by the straight line $C(\mathbf{o},\mathbf{x}') = \{\mathbf{z}(\sigma) = \mathbf{x}' + \sigma\hat{\mathbf{n}} : \mathbf{z}(\sigma_0) = \mathbf{o}\}$, which starts at the origin $\mathbf{o}$ specified by value σ_0, is directed along $\hat{\mathbf{n}} = [\mathbf{z}(\sigma) - \mathbf{x}']/\sigma$, and ends at $\mathbf{x}'$. If we choose the origin $\mathbf{o}$ as the coordinate origin $\mathbf{0}$, which in Eq. (1) is the position of the charge $-q$, then the associated polarization field is

$$\begin{aligned}\mathbf{P}(\mathbf{x}) &= -q\int_{\sigma_0}^{0} d\sigma \frac{\mathbf{r}}{\sigma_0}\delta\left(\mathbf{x}-\mathbf{r}\left[1-\frac{\sigma}{\sigma_0}\right]\right)\\ &= q\int_0^1 d\lambda\,\mathbf{r}\delta(\mathbf{x}-\lambda\mathbf{r}),\end{aligned} \tag{44}$$

which we recognize as the multipolar-gauge polarization. Using Eq. (40), we can also express the Power-Zienau-Woolley transformation as

$$U_{01} = \exp\left[-iq\Lambda_C\right], \tag{45}$$

where

$$\Lambda_C \coloneqq \int_{C(\mathbf{0},\mathbf{r})} d\mathbf{z}\cdot\mathbf{A}_{\mathrm{T}}(\mathbf{z}) \tag{46}$$

is a Wilson line operator (Wilson, 1974). Equation (46) provides an analogy with quark confinement (Woolley, 2020). Specifically, Woolley (2020) found that for the multipolar gauge choice of path, i.e., for the straight-line path between two charges at $\mathbf{r}_1$ and $\mathbf{r}_2$, the polarization energy

$$E_P = \frac{1}{2}\int d^3x\mathbf{P}_g(\mathbf{x})^2 = \frac{q}{2}\int_{\mathbf{r}_1}^{\mathbf{r}_2} d\mathbf{z}\cdot\mathbf{P}_g(\mathbf{z}) \tag{47}$$

possesses a contribution that increases with increasing separation. Analogously, in a state involving $e^{-iq\Lambda_C}$ as a phase factor, the electric-field energy of two oppositely charged quarks increases linearly with separation, which is interpreted as the cause of confinement. The energy E_P also includes a delta-function contribution and a term that diverges as $1/a$, where $a \to 0$ specifies the point-charge limit (Woolley, 2020). We note that the polarization in Eq. (44) does not require one to specify an arbitrary fixed center of the charge distribution. As we describe in note VI.2 of the Supplemental Material (245), it is possible to extend this treatment to arbitrary numbers of charges, and this provides a description of atoms and molecules that, unlike conventional molecular QED [see Craig and Thirunamachandran (1998)], does not depend on arbitrary fixed molecular centers.

Following the arguments of Belinfante (1962), one can use the solution in Eq. (33) to provide a novel derivation of the Coulomb-gauge polarization $\mathbf{P}_{\mathrm{L}} = -\mathbf{E}_{\mathrm{L}}$. For concreteness, we again consider the atomic charge density in Eq. (1). We consider the straight line $C(\mathbf{o},\mathbf{x})$ and choose the origin $\mathbf{o}$ as a point at spatial infinity, which yields the polarization

$$\mathbf{P}(\mathbf{x}) = -q\int_{-\infty}^{0} d\sigma\,\hat{\mathbf{n}}[\delta(\sigma\hat{\mathbf{n}}-\mathbf{x}) - \delta(\mathbf{r}+\sigma\hat{\mathbf{n}}-\mathbf{x})]. \tag{48}$$

When one sets $\mathbf{y} = -\sigma\hat{\mathbf{n}}$, with $|\mathbf{y}| = -\sigma$, and expresses the associated volume element as $d^3y = dyd\Omega|\mathbf{y}|^2$, the average of Eq. (48) over all directions $\hat{\mathbf{n}}$ can be computed as

$$\begin{aligned}\int\frac{d\Omega}{4\pi}\mathbf{P}(\mathbf{x}) &= -\frac{q}{4\pi}\int d^3y\frac{\mathbf{y}}{|\mathbf{y}|^3}[\delta(\mathbf{y}+\mathbf{x}) - \delta(\mathbf{y}-\mathbf{r}+\mathbf{x})]\\ &= \frac{q}{4\pi}\left[\frac{\mathbf{x}}{|\mathbf{x}|^3} - \frac{\mathbf{x}-\mathbf{r}}{|\mathbf{x}-\mathbf{r}|^3}\right] = -\mathbf{E}_{\mathrm{L}}(\mathbf{x}).\end{aligned} \tag{49}$$

We see, therefore, that the Coulomb gauge specifies a delocalized polarization, in which polarizations localized along the straight line with direction $\hat{\mathbf{n}}$ stretching between the charges and spatial infinity are then averaged over all directions $\hat{\mathbf{n}}$.

The solution in Eq. (41) suggests an interpretation of the paths on which the polarization field is localized as "lines of force" in the sense of Faraday (1846). For a single charge at position $\mathbf{r}$, Dirac has interpreted the path $C(\mathbf{o},\mathbf{r})$ as a single line of force between the charge and the origin $\mathbf{o}$ (Dirac, 1955). It has been suggested that a novel QED might be constructed in which the paths on which the polarization field is localized are themselves taken as the dynamical variables of the theory. A suitable averaging procedure over all paths would be required to eliminate the dependence on any choice of path (Dirac, 1955; Woolley, 2020).

2. Fourier transform

To further understand the significance of the freedom to choose $\mathbf{g}_{\mathrm{T}}$, it is convenient to introduce the unconstrained function $\mathbf{G}$, which is essentially completely arbitrary, as

$$\tilde{\mathbf{g}}_{\mathrm{T}}(\mathbf{k},\mathbf{x}) = \sum_\sigma \mathbf{e}_\sigma(\mathbf{k})[\mathbf{e}_\sigma(\mathbf{k})\cdot\tilde{\mathbf{G}}(\mathbf{k},\mathbf{x})], \tag{50}$$

where $\mathbf{e}_\sigma(\mathbf{k}), \sigma = 1,2$ are orthonormal vectors spanning the plane orthogonal to $\mathbf{k}$. Restricting our attention to the α gauges of Sec. II.A amounts to restricting $\mathbf{G}$ as

$$\tilde{\mathbf{G}}_\alpha(\mathbf{k},\mathbf{x}) = \alpha\tilde{\mathbf{G}}_1(\mathbf{k},\mathbf{x}) = -\frac{\alpha\mathbf{x}}{\sqrt{(2\pi)^3}}\int_0^1 d\lambda\, e^{-i\mathbf{k}\cdot\lambda\mathbf{x}}, \quad (51)$$

where now only α is freely choosable. The multipolar gauge $\alpha = 1$ specifies polarization $\mathbf{P}_{\mathrm{T}1}$, which is singular at the origin and therefore often regularized at small distances (Cohen-Tannoudji, Dupont-Roc, and Grynberg, 1989; Vukics, Grießer, and Domokos, 2015; Grießer, Vukics, and Domokos, 2016). This is achieved through the introduction of a form factor such as a Lorentzian with frequency cutoff k_M to give

$$\tilde{\mathbf{G}}_{\alpha M}(\mathbf{k},\mathbf{x}) = -\frac{\alpha\mathbf{x}}{\sqrt{(2\pi)^3}}\frac{k_M^2}{k^2+k_M^2}\int_0^1 d\lambda\, e^{-i\mathbf{k}\cdot\lambda\mathbf{x}}. \quad (52)$$

For k_M finite the field $\mathbf{P}_{\mathrm{T}\alpha}$ is no longer singular at $\mathbf{0}$. This regularization of $\mathbf{P}_{\mathrm{T}\alpha}$ actually constitutes a choice of gauge; that is, we now have a two-parameter gauge function uniquely specified by a gauge vector (α, k_M). Only for $\alpha = 0$ do we have $\mathbf{P}_{\mathrm{T}\alpha} = \mathbf{0}$ and $\chi_0 = 0$, such that regularization of $\mathbf{P}_{\mathrm{T}}$ has no effect on the Hamiltonian. Note that if $\mathbf{P}_{\mathrm{L}}$ is similarly regularized, then for $\alpha = 1$ the ensuing total polarization $\mathbf{P}_1$ is no longer point localized, but instead exponentially localized. Regularization of $\mathbf{P}_{\mathrm{L}} = -\nabla^{-2}\rho$ is not, however, a choice of gauge. The procedure instead amounts to a relaxation of the strict point-particle limit of $\rho(\mathbf{x})$, given by $k_M \to \infty$.

More generally than in Eq. (52), we may let

$$\tilde{\mathbf{G}}_{\{\alpha\}}(\mathbf{k},\mathbf{x}) = -\frac{\alpha(\mathbf{k})^*\mathbf{x}}{\sqrt{(2\pi)^3}}\int_0^1 d\lambda\, e^{-i\mathbf{k}\cdot\lambda\mathbf{x}}, \quad (53)$$

which from Eqs. (37) and (36) yields

$$\chi_g(\mathbf{x}) = \int d^3k \sum_\sigma \alpha(\mathbf{k})\mathbf{e}_\sigma(\mathbf{k})\cdot\tilde{\mathbf{A}}_{\mathrm{T}}(\mathbf{k})\mathbf{e}_\sigma(\mathbf{k})\cdot\tilde{\mathbf{G}}_1(\mathbf{k},\mathbf{x})^*, \quad (54)$$

$$\tilde{\mathbf{P}}_{\mathrm{T}g}(\mathbf{k})^* = -\int d^3x \sum_\sigma \alpha(\mathbf{k})\mathbf{e}_\sigma(\mathbf{k})\mathbf{e}_\sigma(\mathbf{k})\cdot\tilde{\mathbf{G}}_1(\mathbf{k},\mathbf{x})^*\rho(\mathbf{x}), \quad (55)$$

where $\tilde{\mathbf{G}}_1$ is as given in Eq. (51). The field χ_g depends on photonic degrees of freedom through $\tilde{\mathbf{A}}_{\mathrm{T}}$ and couples to the material momentum $\mathbf{p}$ within the Hamiltonian, while the field $\tilde{\mathbf{P}}_{\mathrm{T}g}$ depends on the material degrees of freedom through ρ and couples to the photonic momentum $\tilde{\mathbf{\Pi}}$ within the Hamiltonian. Thus, Eq. (53) enables broad control over the physical nature of the light-matter coupling because, while it is restricted in its $\mathbf{x}$ dependence, $\alpha(\mathbf{k}) = \alpha(-\mathbf{k})^*$ is essentially arbitrary. As an example, we see in Sec. VI that the gauge $\alpha(\mathbf{k}) = \omega_m/(\omega+\omega_m)$, where ω_m is a material frequency, is noteworthy. It can be interpreted as defining a canonical harmonic dipole that automatically subsumes the virtual photons dressing the system ground state (Drummond, 1987; Stokes *et al.*, 2012; Stokes and Nazir, 2019). The Green's function $\mathbf{g}_{\mathrm{T}}(\mathbf{x},\mathbf{x}')$ may yet be more general than the previously listed forms. In particular, the specification of the previous fixed dependence on the second argument $\mathbf{x}'$ stems from the line-integral solution in Eq. (41), which is not the most general form of $\mathbf{g}_{\mathrm{T}}$, as shown by Healy (1978). Furthermore, the gauge function χ_g need not even be restricted as in Eq. (37). This broad generality warrants further study but is not considered here.

E. Sharing out the constrained degrees of freedom: Regularization and localization

The choice of $\mathbf{G}$ determines the physical meaning of the canonical degrees of freedom. To see how, we focus on the simple choices given by Eqs. (51) and (52). We begin by considering the "unregularized" one-parameter gauges with $\mathbf{G}_\alpha$ defined by Eq. (51). We first consider the potential $\mathbf{A}_\alpha$ and the momentum $\mathbf{p}$ determined physically by $\mathbf{A}_\alpha$. According to Eq. (9), $\mathbf{A}_\alpha$ is a function of $\mathbf{A}_0 = \mathbf{A}_{\mathrm{T}} = (\boldsymbol{\nabla}\times)^{-1}\mathbf{B}$, so it can be expressed as the following convex sum of the extremal potentials $\mathbf{A}_0$ and $\mathbf{A}_1$:

$$\begin{aligned}\mathbf{A}_\alpha(\mathbf{x}) &= (1-\alpha)\mathbf{A}_0(\mathbf{x}) + \alpha\mathbf{A}_1(\mathbf{x}) \\ &= \int d^3x' \frac{(1-\alpha)\boldsymbol{\nabla}'\times\mathbf{B}(\mathbf{x}')}{4\pi|\mathbf{x}-\mathbf{x}'|} - \alpha\int_0^1 d\lambda\,\lambda\mathbf{x}\times\mathbf{B}(\lambda\mathbf{x}).\end{aligned} \quad (56)$$

Equation (56) shows that the potential $\mathbf{A}_\alpha(\mathbf{r})$, as it appears in the Hamiltonian, is nonlocal in any gauge, but it is most localized in the multipolar gauge ($\alpha = 1$) because all points $\mathbf{x}$ for which $\mathbf{A}_1(\mathbf{r})$ depends on the local field $\mathbf{B}(\mathbf{x})$ are inside the atom ($|\mathbf{x}| \le |\mathbf{r}|$). More precisely, $\mathbf{A}_1(\mathbf{r})$ depends on $\mathbf{B}$ only at points on the straight line connecting $\mathbf{0}$ to $\mathbf{r}$. The value of α within the vector potential $\mathbf{A}_\alpha$ dictates the balance between this more localized contribution and the nonlocal contribution $(1-\alpha)\mathbf{A}_0$ given by the $\mathbf{x}'$ integral in Eq. (56). The quantity $q\mathbf{A}_0(\mathbf{r}) = q\mathbf{A}_{\mathrm{T}}(\mathbf{r})$ is the momentum associated with the longitudinal electric field of the charge q at $\mathbf{r}$, viz. (Cohen-Tannoudji, Dupont-Roc, and Grynberg, 1989),

$$\mathbf{K}_{\mathrm{long}} \coloneqq \int d^3x \mathbf{E}_{\mathrm{L}\mathbf{r}}\times\mathbf{B} = q\mathbf{A}_{\mathrm{T}}(\mathbf{r}), \quad (57)$$

where $\mathbf{E}_{\mathrm{L}\mathbf{r}}(\mathbf{x}) \coloneqq -q\boldsymbol{\nabla}(4\pi|\mathbf{x}-\mathbf{r}|)^{-1}$, which is consistent with Eq. (56).

To see how $\mathbf{A}_\alpha$ determines the physical nature of $\mathbf{p}$, which defines the canonical atom, consider the EDA implemented as

$$\tilde{\mathbf{G}}_\alpha(\mathbf{k},\mathbf{r}) \approx -\frac{\alpha\mathbf{r}}{\sqrt{(2\pi)^3}}, \quad (58)$$

which implies

$$\begin{aligned}\mathbf{A}_\alpha(\mathbf{r}) &\coloneqq \mathbf{A}_{\mathrm{T}}(\mathbf{r}) - \alpha\boldsymbol{\nabla}_{\mathbf{r}}\int_0^1 d\lambda\,\mathbf{r}\cdot\mathbf{A}_{\mathrm{T}}(\lambda\mathbf{r}) \\ &\approx \mathbf{A}_{\mathrm{T}}(\mathbf{0}) - \alpha\boldsymbol{\nabla}_{\mathbf{r}}[\mathbf{r}\cdot\mathbf{A}_{\mathrm{T}}(\mathbf{0})] = (1-\alpha)\mathbf{A}_{\mathrm{T}}(\mathbf{0}).\end{aligned} \quad (59)$$

According to Eq. (59), the multipolar vector potential at the position of the dipole $\mathbf{A}_1(\mathbf{0})$ vanishes at dipole order. The dipole canonical momentum is defined by $\mathbf{p} = m\dot{\mathbf{r}} + q\mathbf{A}_\alpha(\mathbf{0})$, where $\mathbf{A}_\alpha(\mathbf{0}) = (1-\alpha)\mathbf{A}_{\mathrm{T}}(\mathbf{0})$ [Eq. (59)]. For $\alpha = 1$ we have $\mathbf{p} = m\dot{\mathbf{r}}$, such that $\mathbf{E}_{\mathrm{L}}$ makes no contribution to the canonical

pair $\{\mathbf{r},\mathbf{p}\}$, which is therefore "bare." For $\alpha=0$, the momentum $\mathbf{p}=m\dot{\mathbf{r}}+\mathbf{K}_{\text{long}}$ is fully dressed by $\mathbf{E}_{\text{L}\mathbf{r}}$. Thus, the gauge α controls the extent to which the canonical dipole is dressed by the electrostatic field of the dynamical charge q at $\mathbf{r}$.

We now repeat the previous analysis in the case of the other quantity that is determined by the gauge α, namely, $\mathbf{P}_{\text{T}\alpha}$. We then see how this quantity determines the second canonical momentum $\mathbf{\Pi}$. The total α-gauge polarization is $\mathbf{P}_\alpha=\mathbf{P}_{\text{L}}+\alpha\mathbf{P}_{\text{T1}}$, where $\mathbf{P}_{\text{L}}=-\mathbf{E}_{\text{L}}=\mathbf{P}_0$ defines the nonlocal Coulomb-gauge polarization and $\mathbf{P}_{\text{T1}}$ is the transverse part of the multipolar polarization. The total multipolar polarization $\mathbf{P}_1$ is given in Eq. (15), showing that it is a line integral that vanishes at all points $\mathbf{x}$ that are not on the straight line from $\mathbf{0}$ to $\mathbf{r}$. Therefore, outside the atom ($|\mathbf{x}|>|\mathbf{r}|$) we have $\mathbf{P}_{\text{T1}}=-\mathbf{P}_{\text{L}}=\mathbf{E}_{\text{L}}$. The α-gauge polarization can be written analogously to Eq. (56) as a convex sum of the extremal polarizations $\mathbf{P}_0$ and $\mathbf{P}_1$,

$$\mathbf{P}_\alpha=(1-\alpha)\mathbf{P}_0+\alpha\mathbf{P}_1. \tag{60}$$

The polarization $\mathbf{P}_\alpha$ is nonlocal in any gauge, but it is most localized in the multipolar gauge $\alpha=1$ because all points $\mathbf{x}$ for which $\mathbf{P}_1(\mathbf{x})\neq\mathbf{0}$ are inside the atom ($|\mathbf{x}|\leq|\mathbf{r}|$). Within $\mathbf{P}_\alpha$, the value of α dictates the balance between this more localized contribution and the nonlocal contribution $(1-\alpha)\mathbf{P}_0=-(1-\alpha)\mathbf{E}_{\text{L}}$.

As before, we can approximate the stationary atom as a dipole at the origin $\mathbf{0}$ using Eq. (58) to obtain $\mathbf{P}_{\text{T1}}(\mathbf{x})=q\mathbf{r}\cdot\delta_{\text{T}}(\mathbf{x})$, where $q\mathbf{r}$ is the dipole moment. Within the fixed gauge α, the field canonical momentum operator is defined by $\mathbf{\Pi}=-\mathbf{E}_{\text{T}}-\alpha\mathbf{P}_{\text{T1}}=-\mathbf{E}_{\text{T}}-\alpha\mathbf{E}_{\text{L}}$, where the second equality holds for $\mathbf{x}\neq\mathbf{0}$. Thus, the value of α controls the extent to which the canonical pair $\{\mathbf{A}_{\text{T}},\mathbf{\Pi}\}$ includes the electrostatic field $\mathbf{E}_{\text{L}}=\mathbf{E}-\mathbf{E}_{\text{T}}$. For $\alpha=0$ we have $\mathbf{\Pi}=-\mathbf{E}_{\text{T}}$, so $\mathbf{E}_{\text{L}}$ is completely absent from the field canonical degrees of freedom. For $\alpha=1$ we have $\mathbf{\Pi}=-\mathbf{E}$ for $\mathbf{x}\neq\mathbf{0}$, so the situation is reversed; $\mathbf{E}_{\text{L}}$ is fully included in the field canonical degrees of freedom for all $\mathbf{x}\neq\mathbf{0}$. This holds beyond the EDA, but the condition $\mathbf{x}\neq\mathbf{0}$ must be replaced by $|\mathbf{x}|>|\mathbf{r}|$, specifying all points outside the atom. Gauss's law implies gauge redundancy by constraining $\mathbf{E}$, and this lies at the heart of gauge ambiguities in ultrastrong-coupling QED. The gauge α controls the weight with which $\mathbf{E}_{\text{L}}$ is shared between the two canonical pairs $\{\mathbf{A}_{\text{T}},\mathbf{\Pi}\}$ and $\{\mathbf{r},\mathbf{p}\}$.

We can also consider the regularization of the previous theory at short distances around the distribution center $\mathbf{0}$ using $\mathbf{G}_{\alpha M}$ in Eq. (52), which within the EDA is

$$\tilde{\mathbf{G}}_{\alpha M}(\mathbf{k},\mathbf{r})\approx-\frac{\alpha\mathbf{r}}{\sqrt{(2\pi)^3}}\frac{k_M^2}{k^2+k_M^2}. \tag{61}$$

The transverse (α,k_M)-gauge polarization within the EDA is therefore

$$\mathbf{P}_{\text{T}\alpha M}(\mathbf{x})=\alpha q\mathbf{r}\cdot\delta_M^{\text{T}}(\mathbf{x}), \tag{62}$$

where $\delta_M^{\text{T}}(\mathbf{x})$ denotes the regularized transverse delta function (Cohen-Tannoudji, Dupont-Roc, and Grynberg, 1989)

$$\delta_{M,ij}^{\text{T}}(\mathbf{x})=\frac{2}{3}\delta_{ij}\delta(\mathbf{x})-\frac{\beta(x)}{4\pi x^3}(\delta_{ij}-3\hat{x}_i\hat{x}_j), \tag{63}$$

$$\beta(x)=1-(1+k_Mx+\tfrac{1}{2}k_M^2x^2)e^{-k_Mx}. \tag{64}$$

The function $\beta(x)$ controls the singularity at $\mathbf{0}$ but is unity for $x\gg1/k_M$. The transverse delta function $\delta^{\text{T}}(\mathbf{x})$ is strictly recovered in the limit $k_M\to\infty$. In the (α,k_M) gauge the parameter α functions as before, while the additional gauge parameter k_M controls the rate of exponential localization of what was previously the singular pointlike multipolar dipole. It is now the case that only for $x\gg1/k_M$ do we have $\mathbf{P}_1(\mathbf{x})=\mathbf{0}$. Thus, there are now many "multipolar gauges" specified by the gauge vectors $(1,k_M)$, each of which possesses a different degree of exponential dipolar localization.

The (α,k_M)-gauge vector potential is within the EDA

$$\mathbf{A}_{\alpha M}(\mathbf{r})\approx\mathbf{A}_{\text{T}}(\mathbf{0})-\alpha\int\frac{d^3k}{\sqrt{(2\pi)^3}}\frac{k_M^2}{k^2+k_M^2}\tilde{\mathbf{A}}_{\text{T}}(\mathbf{k}), \tag{65}$$

such that $\mathbf{A}_1(\mathbf{0})=\mathbf{0}$ is recovered in the limit $k_M\to\infty$. More generally, vanishing of $\mathbf{A}_{1M}(\mathbf{r})$ to dipole order requires that $\tilde{\mathbf{A}}_{\text{T}}(\mathbf{k})\approx\mathbf{0}$ for $k\geq k_M$. For this to be the case, the modes $k\geq k_M$ must not be populated. This is the case if the bare atom (as occurs in the free theory) is small compared to the characteristic wavelengths of the populated modes. In other words, the EDA places a lower bound on the cutoff k_M in order that gauges $(1,k_M)$ possess the property $\mathbf{A}_{1M}(\mathbf{0})=\mathbf{0}$ that at dipole order characterizes the usual multipolar gauge $(\alpha,k_M)=(1,\infty)$.

F. Discussion on gauge fixing, forms of rotation, forms of coupling, and common pitfalls

1. Gauge freedom and gauge fixing

We have defined the gauge principle according to modern gauge-field theory and we have given a formulation of canonical (Hamiltonian) nonrelativistic QED in an arbitrary gauge. One of the main objectives of this review is to clarify what gauge freedom, gauge fixing, and gauge ambiguities are within the following theory.

- *Gauge freedom* in electrodynamics is a freedom to choose $\mathbf{A}_{\text{L}}$. Once $\mathbf{A}_{\text{L}}$ is fixed, the scalar potential $\phi_{\mathbf{A}_{\text{L}}}$ is also fixed up to a constant by $-\nabla\phi_{\mathbf{A}_{\text{L}}}=\mathbf{E}_{\text{L}}+\partial_t\mathbf{A}_{\text{L}}$. *Gauge fixing* means specifying $\mathbf{A}_{\text{L}}$ in terms of gauge-invariant quantities.

We have provided a formulation of QED in which $\mathbf{A}_{\text{L}}$ is fixed by Eq. (37) [note II in the Supplemental Material (245)] as $\mathbf{A}_{\text{L}}(\mathbf{x})=\nabla\chi_g(\mathbf{x},\mathbf{A}_{\text{T}})$, meaning that it is fixed up to a choice of the non-operator-valued function $\mathbf{g}_{\text{T}}$. The corresponding vector and scalar potentials are given in accordance with their fundamental definitions by $\mathbf{A}_g=\mathbf{A}_{\text{T}}+\nabla\chi_g$ and $\phi_g=\phi-\partial_t\chi_g$, where $\mathbf{E}_{\text{L}}=-\nabla\phi$.

2. Equality of multipolar and Poincaré gauges

QED in multipolar form and its relation to the Poincaré gauge has been a recent topic of debate (Rousseau and

Felbacq, 2017, 2018; Andrews *et al.*, 2018; Vukics, Kónya, and Domokos, 2021). Rousseau and Felbacq (2017) employed Dirac's constrained quantization procedure to derive the nonrelativistic QED Hamiltonian in the Poincaré gauge. They claimed that the multipolar Hamiltonian would not produce the same results as the Coulomb-gauge Hamiltonian and that it did not coincide with the correct Poincaré gauge Hamiltonian. Andrews *et al.* (2018) and Vukics, Kónya, and Domokos (2021) disputed this, concluding that criticisms of the multipolar framework given by Rousseau and Felbacq (2017) are not valid. A reply to Vukics, Kónya, and Domokos (2021) was offered by Rousseau and Felbacq (2018), who argued that the conclusions of Vukics, Kónya, and Domokos (2021) are not correct, maintaining their conclusion that the Poincaré gauge Hamiltonian does not coincide with the multipolar Hamiltonian.

We clarify the relation between the multipolar theory and the Poincaré gauge in note VI of the Supplemental Material (245). We show in notes II and IX of the Supplemental Material (245) that Dirac's quantization procedure does yield the well-known multipolar theory. We also show that the latter can be obtained via a gauge-fixing transformation from the Coulomb gauge and that, even in the case of multiple charge distributions, the multipolar theory is obtained by choosing what we term ζ-Poincaré gauge-fixing conditions. Complete reconciliation of our results with those of Rousseau and Felbacq (2017, 2018), Andrews *et al.* (2018), and Vukics, Kónya, and Domokos (2021) was provided by Stokes and Nazir (2021a) and in note IX of the Supplemental Material (245) through the construction of Dirac brackets within the theory of a single electron atom. This reveals precisely where misunderstanding has occurred, while also fully clarifying the status of the multipolar (Poincaré gauge) theory.

3. Dipolar coupling

An aspect of light-matter interactions that is especially poorly understood concerns the field that a dipole couples to within the multipolar gauge. Common misidentifications are exacerbated by the development of the theory via semiclassical treatments, as features heavily in textbook quantum optics (Scully and Zubairy, 1997; Schleich, 2001; Gerry and Knight, 2004). In such treatments the gauge principle implies that the EDA of the semiclassical Power-Zienau-Woolley (PZW) transformation applied to $\mathbf{p} - q\mathbf{A}_{\mathrm{T}}(\mathbf{0})$ within the Scrödinger equation yields the bilinear coupling $-q\mathbf{r}\cdot\mathbf{E}_{\mathrm{T}}(\mathbf{0})$, where $\mathbf{E}_{\mathrm{T}} = -\partial_t \mathbf{A}_{\mathrm{T}}$ (Scully and Zubairy, 1997; Schleich, 2001; Gerry and Knight, 2004). However, according to the fully quantum description the correct bilinear component of the coupling is $-q\mathbf{r}\cdot\mathbf{D}_{\mathrm{T}}(\mathbf{0})$, where $\mathbf{D}_{\mathrm{T}} = \mathbf{E}_{\mathrm{T}} + \mathbf{P}_{\mathrm{T}}$. The field $\mathbf{P}_{\mathrm{T}}$ is singular at $\mathbf{0}$, so the fully quantum description provides a coupling that is infinitely different from the result of a semiclassical approach. Nevertheless, the notation $-q\mathbf{r}\cdot\mathbf{E}_{\mathrm{T}}(\mathbf{0})$ remains prevalent even in textbooks that employ fully quantum treatments (Loudon, 2000). Further confusion stems from the fact that $\mathbf{E}_{\mathrm{T}}$ is often written simply as $\mathbf{E}$ even when $\mathbf{E}_{\mathrm{L}} \neq \mathbf{0}$, such that the notation $-q\mathbf{r}\cdot\mathbf{E}(\mathbf{0})$ is also encountered in textbooks (Agarwal, 2012) and, more recently, in the ultrastrong-coupling light-matter physics literature (Settineri *et al.*, 2021).

Furthermore, it is not commonly recognized that within the EDA $\mathbf{D}_{\mathrm{T}} = \mathbf{E}$, but only for $\mathbf{x} \neq \mathbf{0}$; see Sec. II.E. The interchanging of fields $\mathbf{D}_{\mathrm{T}}$, $\mathbf{E}_{\mathrm{T}}$, and $\mathbf{E}$, which are related but not equal, may lead to the misidentification of fields both at and away from the dipole's position. We emphasize that neither $-q\mathbf{r}\cdot\mathbf{E}_{\mathrm{T}}(\mathbf{0})$ nor $-q\mathbf{r}\cdot\mathbf{E}(\mathbf{0})$ is a correct interaction, and neither is it true that $\mathbf{E}_{\mathrm{T}} = \mathbf{E}$, whereas it is true that $\mathbf{D}_{\mathrm{T}} = \mathbf{E}$ at points $\mathbf{x}$ outside of the charge distribution, which within the EDA means for $\mathbf{x} \neq \mathbf{0}$. In the weak-coupling regime one can often afford to misidentify the physical fields involved in light-matter interactions, but this may lead to erroneous results in sufficiently strong-coupling regimes.

Similarly, confusion can arise in nonrelativistic QED due to claims that a dipolar coupling such as $-q\mathbf{r}\cdot\mathbf{E}_{\mathrm{T}}(\mathbf{0})$ may be preferable to a Coulomb-gauge coupling because it is expressible solely in terms of a gauge-invariant electric field. Scully and Zubairy (1997) provided a typical semiclassical derivation of $-q\mathbf{r}\cdot\mathbf{E}_{\mathrm{T}}(\mathbf{0})$ and referred to the semiclassical multipolar gauge as the E gauge. They stated that the E-gauge interaction is gauge invariant, in contrast to a linear $\mathbf{p}\cdot\mathbf{A}_{\mathrm{T}}$ interaction found in the Coulomb gauge. It was argued that only in the E gauge is the unperturbed dipolar Hamiltonian

$$H_m = \frac{\mathbf{p}^2}{2m} + V(\mathbf{r}), \tag{66}$$

a physical quantity. However, in the Coulomb gauge both $\mathbf{p} = m\dot{\mathbf{r}} + \mathbf{K}_{\mathrm{long}}$ and $\mathbf{A}_{\mathrm{T}}$ are also gauge invariant. Indeed, there is no means by which the requirement of gauge invariance can be leveraged as an argument to prefer one gauge over another. The theory in any gauge can be expressed entirely in terms of gauge-invariant quantities by the definition of gauge fixing, as explained in Sec. II.F.1.

We have already seen that the Hamiltonian always represents the total energy [Eq. (54) in note III of the Supplemental Material (245) and Eq. (16)] (Stokes and Nazir, 2019). If one prefers to eliminate only $\mathbf{A}_\alpha$ from the expression for the Hamiltonian but retain its explicit dependence on the canonical momenta, then this is easily achieved in any gauge α using Eq. (56). In particular, the Coulomb-gauge theory for which $\mathbf{\Pi} = -\mathbf{E}_{\mathrm{T}}$ can be expressed solely in terms of electric and magnetic fields. The latter property is not unique to the multipolar theory.

4. Active and passive perspectives of unitary rotations

A generic feature of linear spaces is that rotations therein may be implemented in an active or passive way. A vector $\mathbf{v} = \sum_i v_i \mathbf{e}_i$ within Hermitian inner-product space V may be actively rotated using a unitary transformation R into a new vector $\mathbf{v}' = R\mathbf{v} = \sum_i v_i' \mathbf{e}_i$. Expressed in the same basis $\{\mathbf{e}_i\}$, the new vector has components $v_i' = \sum_j R_{ij} v_j$, where $R_{ij} = \langle \mathbf{e}_i, R\mathbf{e}_j \rangle$. Alternatively, the original vector $\mathbf{v}$ may be expressed in a rotated basis $\{\mathbf{e}_i' = R^\dagger \mathbf{e}_i\}$ to give $\mathbf{v} = \sum_i v_i' \mathbf{e}_i'$. In both cases the same numerical components $\{v_i'\}$ are obtained from the rotation. Note that the passive rotation $R^\dagger$ of basis vectors $\mathbf{e}_i$ is in the opposite direction of the active rotation R of $\mathbf{v}$.

The same considerations apply when one unitarily rotates a Hamiltonian expressed in a canonical operator "basis." In Sec. II.A and note III of the Supplemental Material (245), an active perspective of unitary rotations has been adopted whereby the canonical operators $\mathbf{y} = (\psi, \psi^\dagger, \mathbf{A}_\mathrm{T}, \mathbf{\Pi})$ are viewed as fixed, while the Hamiltonian can be rotated to a new but equivalent form using a gauge-fixing transformation as $H_{g'}(\mathbf{y}) = U_{gg'} H_g(\mathbf{y}) U^\dagger_{gg'}$ [in the particle-based α-gauge formalism of Sec. II.A we instead have $\mathbf{y} = (\mathbf{r}, \mathbf{p}, \mathbf{A}_\mathrm{T}, \mathbf{\Pi})$ and $H_{\alpha'}(\mathbf{y}) = R_{\alpha\alpha'} H_\alpha(\mathbf{y}) R^\dagger_{\alpha\alpha'}$]. The transformation of the Hamiltonian can be implemented via transformation of the canonical operators in the sense that $U_{gg'} H_g(\mathbf{y}) U^\dagger_{gg'} = H_g(U_{gg'} \mathbf{y} U^\dagger_{gg'})$; cf. Eqs. (51) and (52) in note III of the Supplemental Material (245) and also Eqs. (27) and (28).

The active perspective is commonly found and was adopted by Cohen-Tannoudji, Dupont-Roc, and Grynberg (1989). From this point of view, any operator that does not commute with gauge-fixing transformations, such as $\mathbf{\Pi}$, will represent a different physical observable before and after such a transformation (Cohen-Tannoudji, Dupont-Roc, and Grynberg, 1989). Conversely, a given physical observable will be represented by a different operator before and after transformation. For example, the energy E is represented by $H_g(\mathbf{y})$ in gauge g and by $H_{g'}(\mathbf{y})$ in gauge g'. The eigenvalue equation $H_g(\mathbf{y})|E^n_g\rangle = E^n|E^n_g\rangle$ implies that the vector $|E^n_g\rangle$ represents, within the gauge g, the physical state $\mathcal{S}^n$ in which the system possesses energy E^n. Meanwhile, in the gauge g' the same state $\mathcal{S}^n$ is represented by a different vector $|E^n_{g'}\rangle = U_{gg'}|E^n_g\rangle$ because the energy is represented by a different operator $H_{g'}(\mathbf{y})$.

Alternatively, a passive perspective of rotations may be adopted whereby different canonical operators are associated with different gauges as $\mathbf{y}_g = U_{gg'} \mathbf{y}_{g'} U^\dagger_{gg'}$. Notice that the rotation between canonical operators associated with different gauges g and g' is in the opposite direction of the rotation between the Hamiltonians associated with g and g' obtained via the active perspective. Nevertheless, the same relationship between Hamiltonian functions is obtained within the passive viewpoint by noting that $H_g(\mathbf{y}_g) = H_g(U_{gg'} \mathbf{y}_{g'} U^\dagger_{gg'}) = U_{gg'} H_g(\mathbf{y}_{g'}) U^\dagger_{gg'} = H_{g'}(\mathbf{y}_{g'})$. The passive perspective is also commonly found in the literature (Power and Thirunamachandran, 1983a, 1983b, 1983c, 1992, 1993, 1999a; Craig and Thirunamachandran, 1998). Therein, the Hamiltonian $H_g(\mathbf{y}_g) = H_{g'}(\mathbf{y}_{g'})$ is unique and uniquely represents the energy E. Similarly, the eigenvector $|E^n\rangle$ uniquely represents the physical state $\mathcal{S}^n$ of definite energy E^n. Conversely, each set of canonical operators $\mathbf{y}_g$ explicitly represents a different set of physical observables. This again contrasts the active perspective wherein the physical difference between the same canonical operators $\mathbf{y}$ in different gauges was implicit.

Either an active or passive perspective can be chosen, but the associations between operators and observables and between vectors and states will generally depend on the perspective adopted. The importance of such associations and their relation to gauge freedom is discussed in Secs. III.A and III.B. Here, unless otherwise stated, we adopt an active perspective of unitary rotations.

5. Gauge-symmetry transformations versus gauge-fixing transformations

Confusion can stem from the fact that the PZW transformation R_{01} commutes with $A_0 = \phi$ and $\mathbf{A}_0 = \mathbf{A}_\mathrm{T}$, so it cannot directly implement a gauge transformation [cf. Eqs. (61) and (62) of note IV of the Supplemental Material (245)] as noted by Andrews *et al.* (2018). The situation becomes clear upon recognition that the PZW transformation is not a gauge-symmetry transformation S_χ, but instead an example of a gauge-fixing transformation $U_{gg'}$. The distinction between these types of gauge transformation was recognized some tine ago in relativistic physics (Lenz *et al.*, 1994), but it is perhaps less well known in quantum optics and atomic physics. Within the final unconstrained theory all gauge-symmetry transformations have been reduced to the identity, expressing the fact that once the gauge has been fixed there is no longer any redundancy within the state space or operator algebra. The redundant degrees of freedom $\mathbf{A}_\mathrm{L}$ have been fixed as known functions of the gauge-invariant degrees of freedom. The gauge-fixing transformation $U_{gg'}$ transforms between alternative isomorphic realizations of the physical state space that result from different choices of gauge $\mathbf{A}_\mathrm{L} = \nabla\chi_g$ and $\mathbf{A}_\mathrm{L} = \nabla\chi_{g'}$.

Although $U_{gg'}$ cannot transform $(\phi_g, \mathbf{A}_g)$ directly, it does so indirectly. To see this, note that $H_g(\mathbf{y})$ is shorthand for $H(\mathbf{g}_\mathrm{T}, \mathbf{y})$, where the function H is unique. The concrete choice of function $\mathbf{g}_\mathrm{T}$ used to evaluate H is left open. In other words, $H_{g'}(\mathbf{y})$ defined by $H_{g'}(\mathbf{y}) \coloneqq U_{gg'} H_g(\mathbf{y}) U^\dagger_{gg'}$ is given by $H_{g'}(\mathbf{y}) \equiv H(\mathbf{g}'_\mathrm{T}, \mathbf{y})$. By construction the functional form of the Hamiltonian in terms of $\mathbf{g}_\mathrm{T}$, as well as all resulting dynamical equations written in terms of $(\phi_g, \mathbf{A}_g)$, are the same for every possible concrete choice of $\mathbf{g}_\mathrm{T}$ (gauge). Thus, the following is true in the final unconstrained theory.

- Gauge freedom is the freedom to transform between different Hamiltonians H_g and $H_{g'}$ resulting from different fixed choices of gauge $\mathbf{g}_\mathrm{T}$ and $\mathbf{g}'_\mathrm{T}$.

Gauge invariance means that formulations corresponding to different choices of $\mathbf{g}_\mathrm{T}$ must be physically equivalent. The *unitarity* of gauge-fixing transformations $U_{gg'}$ ensures that this is the case because the quantum-theoretic definition of physical equivalence is unitary equivalence; cf. Sec. III.A.

6. Minimal coupling

A final common pitfall that we want to address concerns the nature of the minimal coupling prescription and its relation to the Coulomb gauge. In Sec. II.A we saw that $R_{\alpha\alpha'}$ implements a gauge change within the Hamiltonian by transforming between distinct minimal coupling prescriptions [Eqs. (27) and (28)]. This shows that the minimal coupling replacement is not synonymous with the Coulomb gauge.

It is unfortunate that the term "minimal coupling" has so often been reserved exclusively for the Coulomb-gauge Hamiltonian H_0 because this nomenclature is in direct opposition to the fundamental meaning of minimal coupling. The gauge principle implies the existence of a potential whose

gauge $\mathbf{A}_{\mathrm{L}}$ can be chosen freely. Different fixed gauges correspond to different fixed minimal coupling replacements, as shown by Eqs. (22) and (27). This fact is obscured by the almost universal practice of expressing the multipolar potential $\mathbf{A}_1$ in terms of $\mathbf{B}$ within the Hamiltonian via Eq. (56). It is then not obvious that the multipolar Hamiltonian results from the minimal coupling replacement $\mathbf{p} \to \mathbf{p} - q\mathbf{A}_1(\mathbf{r})$. Meanwhile, despite it being possible to express the Coulomb-gauge potential $\mathbf{A}_0$ in terms of $\mathbf{B}$, the Coulomb-gauge Hamiltonian is nearly always left as a function of $\mathbf{A}_0$. The minimal coupling prescription $\mathbf{p} \to \mathbf{p} - q\mathbf{A}_0(\mathbf{r})$ is therefore immediately apparent therein. The combined effect of these conventions may be the false impression that only the Coulomb-gauge Hamiltonian results from minimal coupling replacement. In fact, in any gauge α, the Hamiltonian includes a minimal coupling replacement $\mathbf{p} \to \mathbf{p} - q\mathbf{A}_\alpha(\mathbf{r})$ and the potential $\mathbf{A}_\alpha$ is expressible as a function of the magnetic field $\mathbf{B}$.

Further obfuscation occurs within the EDA, which states that $\mathbf{A}_{\mathrm{T}}(\mathbf{x}) \approx \mathbf{A}_{\mathrm{T}}(\mathbf{0})$ whenever $|\mathbf{x}| \leq |\mathbf{r}|$. This implies that χ_α in Eq. (11) is approximated as in Eq. (30). Thus, choosing the multipolar gauge $\mathbf{A}_1$ means choosing $\mathbf{A}_{\mathrm{L}} = \nabla\chi_1$ such that $\mathbf{A}_{\mathrm{L}}(\mathbf{r}) = -\mathbf{A}_{\mathrm{T}}(\mathbf{r})$ within the EDA, giving $\mathbf{A}_1(\mathbf{r}) \approx \mathbf{0}$; cf. Eq. (59). The position $\mathbf{r}$ is where the potential $\mathbf{A}_1$ is evaluated within the Hamiltonian; cf. Eq. (22). Thus, the dipole approximation of the kinetic energy part of the multipolar-gauge Hamiltonian is independent of the potential and the canonical momentum $\mathbf{p}$ becomes purely mechanical ($\mathbf{p} = m\dot{\mathbf{r}}$). This again may lead to the false impression that the multipolar Hamiltonian is not a minimal coupling Hamiltonian.

According to the gauge principle all Hamiltonians H_α are equally valid, and any one of them can be taken as the starting point for a canonical description of QED. It is not the case that only one gauge's Hamiltonian, such as H_0, is compatible with the gauge principle. Indeed, such a conclusion would contradict the gauge principle. In particular, it is not the case that $H_0(t)$ is a fundamentally preferable starting point when one considers time-dependent interactions and that any other Hamiltonian must be obtained from it via a time-dependent gauge transformation. This fact appears to contradict recent articles (Stefano *et al.*, 2019; Settineri *et al.*, 2021). Time-dependent interactions are discussed in detail in Sec. V.

III. SUBSYSTEM GAUGE RELATIVITY

Quantum theory provides postulates for the association of physical states and observables with their mathematical representations, and for the calculation of predictions of observable properties. The notion of a quantum system is an inherently relative one (Zanardi, 2001; Barnum *et al.*, 2004; Zanardi, Lidar, and Lloyd, 2004; Harshman and Wickramasekara, 2007; Viola and Barnum, 2007; Harshman and Ranade, 2011). Understanding quantum subsystem properties in light of this remains a topic of current interest (Cai *et al.*, 2021; Hoehn *et al.*, 2022). The partition of a quantum system into subsystems is dictated by the set of operationally accessible interactions and measurements (Zanardi, Lidar, and Lloyd, 2004). The importance of this fact in QED beyond traditional regimes is addressed in this section.

A. Quantum subsystem relativity

We begin by examining fundamental concepts relating to composite quantum systems and subsequently relate them to gauge freedom. In quantum theory all predictions are obtained from the inner product; therefore, the following associations,

$$\begin{aligned} \text{physical state } \mathcal{S} &\leftrightarrow \text{vector } |\psi\rangle, \\ \text{physical observable } \mathcal{O} &\leftrightarrow \text{operator } O, \end{aligned} \tag{67}$$

are equivalent to the associations

$$\begin{aligned} \text{physical state } \mathcal{S} &\leftrightarrow \text{vector } |\psi'\rangle = U|\psi\rangle, \\ \text{physical observable } \mathcal{O} &\leftrightarrow \text{operator } O' = UOU^\dagger, \end{aligned} \tag{68}$$

where U is any unitary operator (Isham, 1995). In other words, the associations $\{\text{state} \leftrightarrow \text{vector}\}$ and $\{\text{observable} \leftrightarrow \text{operator}\}$ can be made only relative to a Hilbert space frame. The unitary group is the symmetry group of the inner product $\langle\cdot|\cdot\rangle$ defined over $\mathcal{H}$, meaning that U transforms between two distinct Hilbert space frames (bases). This is analogous to moving between frames within Minkowski spacetime $E^{1,3}$ using a Lorentz transformation Λ belonging to the Lorentz group, which is the symmetry group of the (indefinite) Minkowski inner product. The definition of a composite quantum system uses the tensor product $\otimes$, which extends the inner product in the way required in order that probabilities associated with independent subsystems are statistically independent. Specifically, $(\langle\psi_{\mathrm{A}}| \otimes \langle\psi_{\mathrm{B}}|)(|\varphi_{\mathrm{A}}\rangle \otimes |\varphi_{\mathrm{B}}\rangle) \equiv \langle\psi_{\mathrm{A}}|\varphi_{\mathrm{A}}\rangle\langle\psi_{\mathrm{B}}|\varphi_{\mathrm{B}}\rangle$.

To understand how the relativity of associations between operators and observables affects the meaning of quantum subsystems, we consider a composite system of two spins A and B with Hilbert space $\mathcal{H} = \mathcal{H}_{\mathrm{A}} \otimes \mathcal{H}_{\mathrm{B}}$. We denote spin observables in some specified directions for A and B by $\mathcal{O}_{\mathrm{A}}$ and $\mathcal{O}_{\mathrm{B}}$, respectively, and we let these observables be represented in frame X by operators $\sigma_{\mathrm{A}} \otimes I_{\mathrm{B}}$ and $I_{\mathrm{A}} \otimes \sigma_{\mathrm{B}}$, where I_{A} and I_{B} are identity operators over $\mathcal{H}_{\mathrm{A}}$ and $\mathcal{H}_{\mathrm{B}}$, respectively. For notational economy one often writes $O_{\mathrm{A}} \otimes I_{\mathrm{B}}$ ($I_{\mathrm{A}} \otimes O_{\mathrm{B}}$) simply as O_{A} (O_{B}). Spin can take two values denoted as $s = +, -$. The (eigen)state $\mathcal{S}_s$ is the *physical state* in which the *physical observables* $\mathcal{O}_{\mathrm{A}}$ and $\mathcal{O}_{\mathrm{B}}$ simultaneously possess value s. It is represented in frame X by the vector $|s_{\mathrm{A}}\rangle \otimes |s_{\mathrm{B}}\rangle =: |s_{\mathrm{A}}, s_{\mathrm{B}}\rangle$, where $\sigma_{\mathrm{Z}}|s_{\mathrm{Z}}\rangle = s|s_{\mathrm{Z}}\rangle$ with $\mathrm{Z} = \mathrm{A}, \mathrm{B}$. Now consider the unitary transformation U for which

$$U|+_{\mathrm{A}}, +_{\mathrm{B}}\rangle = \frac{1}{\sqrt{2}}(|+_{\mathrm{A}}, +_{\mathrm{B}}\rangle + |-_{\mathrm{A}}, -_{\mathrm{B}}\rangle), \tag{69}$$

which connects frame X to a new frame Y but does not have the form $U_{\mathrm{A}} \otimes U_{\mathrm{B}}$. In frame Y the observables $\mathcal{O}_{\mathrm{A}}$ and $\mathcal{O}_{\mathrm{B}}$ are represented by the operators $\sigma'_{\mathrm{A}} := U\sigma_{\mathrm{A}}U^\dagger$ and $\sigma'_{\mathrm{B}} := U\sigma_{\mathrm{B}}U^\dagger$, respectively, such that the state $\mathcal{S}_s$ is represented by the vector $U|s_{\mathrm{A}}, s_{\mathrm{B}}\rangle$. This ensures that the physical prediction $\langle\mathcal{O}_{\mathrm{Z}}\rangle_{\mathcal{S}_s}$ is frame independent. The frame can therefore be chosen freely. However, in frame Y the operator $\sigma_{\mathrm{Z}} \neq \sigma'_{\mathrm{Z}}$ evidently does not represent the observable $\mathcal{O}_{\mathrm{Z}}$. It must therefore represent some other physical observable, which we denote by O_{Z}.

We now define the subalgebras $\mathcal{A}_{\mathrm{Z}} \coloneqq \{O_{\mathrm{Z}} : O_{\mathrm{Z}}\ \text{Hermitian}\}$ with $\mathrm{Z} = \mathrm{A}, \mathrm{B}$. The mathematical quantum subsystem Z may be defined as the following pair: $\mathrm{Z} = (\mathcal{H}_{\mathrm{Z}}, \mathcal{A}_{\mathrm{Z}})$. Operationally, meanwhile, any physical system must be specified through a collection of observable properties. And yet, whether or not a given observable property belongs to the set of observables that defines the quantum subsystem Z depends on the Hilbert space frame. For example, the observable $\mathcal{O}_{\mathrm{Z}}$ is represented by $\sigma_{\mathrm{z}} \in \mathcal{A}_{\mathrm{Z}}$ in frame X and by $U\sigma_{\mathrm{Z}}U^{\dagger} \notin \mathcal{A}_{\mathrm{Z}}$ in frame Y, whereas the observable O_{Z} is represented by $\sigma_{\mathrm{Z}} \in \mathcal{A}_{\mathrm{Z}}$ in frame Y and by $U^{\dagger}\sigma_{\mathrm{Z}}U \notin \mathcal{A}_{\mathrm{Z}}$ in frame X. It follows that $\mathrm{Z} = (\mathcal{H}_{\mathrm{Z}}, \mathcal{A}_{\mathrm{Z}})$ represents a distinct collection of states and observables in the two different frames X and Y.

Any question about the physics of the system must be posed in terms of states and observables. For example, we can ask the following question: Is the physical state $\mathcal{S}_{+}$ an entangled state? There are two answers: Yes, $\mathcal{S}_{+}$ is entangled with respect to the observables O_{A} and O_{B}, and no, $\mathcal{S}_{+}$ is not entangled with respect to the observables $\mathcal{O}_{\mathrm{A}}$ and $\mathcal{O}_{\mathrm{B}}$. The first answer is deduced using frame Y, where we have the representations $\mathsf{O}_{\mathrm{Z}} \leftrightarrow \sigma_{\mathrm{Z}}$ with $\mathrm{Z} = \mathrm{A}, \mathrm{B}$, and in terms of the eigenvectors of σ_{Z} the state $\mathcal{S}_{+}$ is represented by the entangled vector $(|+_{\mathrm{A}}, +_{\mathrm{B}}\rangle + |-_{\mathrm{A}}, -_{\mathrm{B}}\rangle)/\sqrt{2}$. The second answer is deduced using frame X, where we have $\mathcal{O}_{\mathrm{Z}} \leftrightarrow \sigma_{\mathrm{Z}}$, and in terms of the eigenvectors of σ_{Z} the state $\mathcal{S}_{+}$ is represented by the vector $|+_{\mathrm{A}}, +_{\mathrm{B}}\rangle$.

Both answers to the question are physically meaningful, and they are certainly compatible statements regarding states and observables. The same physical state $\mathcal{S}_{+}$ is simultaneously entangled and not entangled because the term "entanglement" is referring to different physical observable properties in the two different answers to the question. We can further ask whether the entanglement in the state $\mathcal{S}_{+}$ is physically relevant. The answer is yes if we are able to access observables O_{A} and O_{B}, and the answer is no if we are able to access only the observables $\mathcal{O}_{\mathrm{A}}$ and $\mathcal{O}_{\mathrm{B}}$. This again is a statement about physical states and observables, but it also concerns which observables are actually measurable in a given experiment.

In QED gauge-fixing transformations are unitary, so a gauge can be understood as a frame within the Hilbert space. We have seen that the choice of frame can be labeled with a parameter α such that the Hilbert space has the form $\mathcal{H}[\alpha] = \mathcal{H}_{\mathrm{matter}}[\alpha] \otimes \mathcal{H}_{\mathrm{light}}[\alpha]$. Gauge transformations mix the matter and light canonical operators of the theory, which possess the forms $O_{\mathrm{matter}} \otimes I_{\mathrm{light}} \in \mathcal{A}_{\mathrm{matter}}[\alpha]$ and $I_{\mathrm{matter}} \otimes O_{\mathrm{light}} \in \mathcal{A}_{\mathrm{light}}[\alpha]$, respectively. Thus, the matter and light mathematical subsystems defined as the pairs $(\mathcal{H}_{\mathrm{matter}}[\alpha], \mathcal{A}_{\mathrm{matter}}[\alpha])$ and $(\mathcal{H}_{\mathrm{light}}[\alpha], \mathcal{A}_{\mathrm{light}}[\alpha])$, respectively, are defined using physically different collections of observables for each different gauge α. The matter subsystem constitutes a different operational subsystem in each different gauge, as does the light subsystem.

B. Gauge ambiguities and gauge invariance

Quantum theory provides predictions for observables, and the unitarity of the gauge-fixing transformations $U_{gg'}$ $(R_{\alpha\alpha'})$ guarantees the gauge invariance of these predictions. We define gauge invariance as follows.

- A prediction is gauge invariant if it is independent of the gauge in which it is calculated. If all predictions pertaining to an observable are gauge invariant, then the observable is gauge invariant.

In general, an observable $\mathcal{O}$ is represented in the fixed gauge α by a generally α-dependent function o_{α} of the canonical operators $\mathbf{y} = \{\mathbf{r}, \mathbf{A}_{\mathrm{T}}, \mathbf{p}, \mathbf{\Pi}\}$. A physical state $\mathcal{S}$ is represented by an α-dependent vector $|\psi_{\alpha}\rangle$. In the gauge α', the same observable $\mathcal{O}$ is represented by the operator $o_{\alpha}(R_{\alpha\alpha'}\mathbf{y}R^{\dagger}_{\alpha\alpha'}) \equiv R_{\alpha\alpha'}o_{\alpha}(\mathbf{y})R^{\dagger}_{\alpha\alpha'} \eqqcolon o_{\alpha'}(\mathbf{y})$, and the same state $\mathcal{S}$ is represented by the vector $|\psi_{\alpha'}\rangle = R_{\alpha\alpha'}|\psi_{\alpha}\rangle$. The average $\langle\mathcal{O}\rangle_{\mathcal{S}}$ can be calculated in any gauge

$$\langle\psi_{\alpha}|o_{\alpha}(\mathbf{y})|\psi_{\alpha}\rangle = \langle\mathcal{O}\rangle_{\mathcal{S}} = \langle\psi_{\alpha'}|o_{\alpha'}(\mathbf{y})|\psi_{\alpha'}\rangle. \tag{70}$$

This gauge invariance holds as a consequence of the unitarity of gauge-fixing transformations, so it should be clear that it will hold independently of any restriction on the form of the gauge. An example of a gauge-invariant observable is the total energy $\mathcal{O} = E$, which in the gauge α is represented by the Hamiltonian $H_{\alpha}(\mathbf{y})$.

Although QED is fundamentally gauge invariant, the task remains to decide which observables are relevant to us. For example, consider the observables $\mathbf{E}_{\mathrm{T}}$ and $\mathbf{P}_{\mathrm{T}}$, where hereafter we use $\mathbf{P} \coloneqq \mathbf{P}_1$ to denote the multipolar polarization. The transformation $R_{\alpha\alpha'}$ commutes with $\mathbf{P}_{\mathrm{T}}$, so this observable possesses the same operator representation in every gauge (Cohen-Tannoudji, Dupont-Roc, and Grynberg, 1989). The same is not true for $\mathbf{E}_{\mathrm{T}}$. Consider the physical observable $\mathcal{O} \coloneqq -\mathbf{E}_{\mathrm{T}} - \alpha\mathbf{P}_{\mathrm{T}}$, where α denotes a fixed real number. As a fixed linear combination of gauge-invariant observables, $\mathcal{O}$ is gauge invariant. If we now choose our gauge parameter to have the same fixed value α, then the observable $\mathcal{O}$ is represented by the operator $\mathbf{\Pi}$.

We emphasize that gauge freedom is not a freedom to define $\mathcal{O}$. It is a freedom to decide whether the parameter that fixes the redundancy $\mathbf{A}_{\mathrm{L}}$ within our description equals the number α that defines $\mathcal{O}$. If the gauge parameter is instead chosen to have a value $\alpha' \neq \alpha$, then the observable $\mathcal{O}$ is represented by the operator $\mathbf{\Pi}' = R_{\alpha\alpha'}\mathbf{\Pi}R^{\dagger}_{\alpha\alpha'} = \mathbf{\Pi} - (\alpha - \alpha')\mathbf{P}_{\mathrm{T}}$. The operator $\mathbf{\Pi}$ represents the different gauge-invariant physical observable $\mathcal{O}' \coloneqq -\mathbf{E}_{\mathrm{T}} - \alpha'\mathbf{P}_{\mathrm{T}}$. A physical state $\mathcal{S}$ is represented by the vectors $|\psi\rangle$ and $|\psi'\rangle = R_{\alpha\alpha'}|\psi\rangle$ in the gauges α and α', respectively. Thus, the averages of $\mathcal{O}$ and $\mathcal{O}'$ in the state $\mathcal{S}$ are $\langle\mathcal{O}\rangle_{S} = \langle\psi|\mathbf{\Pi}|\psi\rangle$ and $\langle\mathcal{O}'\rangle_{S} = \langle\psi'|\mathbf{\Pi}|\psi'\rangle$. It is important to recognize that both predictions satisfy gauge invariance as defined in Eq. (70). The same operator $\mathbf{\Pi}$ represents different observables $\mathcal{O}$ and $\mathcal{O}'$ in the two averages, whereas different vectors represent the same physical state $\mathcal{S}$.

For a fixed α the combination $\mathbf{\Pi} = -\mathbf{E}_{\mathrm{T}} - \alpha\mathbf{P}_{\mathrm{T}}$ is a gauge-invariant observable, but by definition of $\mathbf{\Pi}$, here α is the gauge parameter. Thus, while it is true that in each gauge $\mathbf{\Pi}$ represents a physical observable and it is also true that every observable possesses unique physical predictions that can be calculated in any gauge, it is not true that the operator $\mathbf{\Pi}$ represents the same physical observable in any two different gauges, and predictions pertaining to different observables are

different: for example, two different observables will not generally possess the same average value. As discussed throughout this review, the task of determining which gauge-invariant predictions are relevant in which situations is not necessarily straightforward, because it depends on the interpretation of virtual processes, dressing, and localization. We therefore conclude the following (Stokes and Nazir, 2021b).

- Gauge ambiguities arise not because it is unclear how to obtain gauge-invariant predictions but because it is not always clear which gauge-invariant observables are operationally relevant. The gauge invariance of a prediction is necessary but not sufficient to ensure its operational relevance.

On a practical level, simply verifying the fundamental gauge invariance of predictions does not imply that gauge freedom can be ignored. For example, Settineri *et al.* (2021) (Sec. V) noted that "of course detectable subsystem excitations and correlations have to be gauge invariant, since the results of experiments cannot depend on the gauge. On this basis we can define gauge invariant excitations and qubit-field entanglement." We note, however, that providing gauge-invariant definitions is straightforward and has never been a problem. Indeed, given the unitarity of gauge-fixing transformations, gauge invariance is automatic. "Ambiguities" occur not because gauge invariance breaks down, but because there are many different gauge-invariant definitions of excitations and qubit-field entanglement. The latter can be defined relative to any gauge; see Sec. III.C. Gauge invariance is necessary, but it is not a sufficient basis for providing physically relevant theoretical definitions. Any conceptual ambiguities that result from the availability of many different physical definitions can be called gauge ambiguities, but they are not due to a breakdown of gauge invariance, which is a fundamental requirement.

C. Definition of subsystem gauge relativity

We adopt the viewpoint that the relevant definition of any system is determined by experimental capability. Operationally, a system comprises a set of observable properties that can be measured. On the other hand, theoretically there is a continuous infinity of different gauge-invariant transverse fields, all of which are represented by the operator $\mathbf{\Pi}$. Any of these fields can be used to define a boson called a photon. Mathematically, photons are defined directly in terms of $\mathbf{\Pi}$ via

$$a_\lambda(\mathbf{k}) \coloneqq \frac{1}{\sqrt{2\omega}}\mathbf{e}_\lambda(\mathbf{k})\cdot[\omega\tilde{\mathbf{A}}_{\mathrm{T}}(\mathbf{k})+i\tilde{\mathbf{\Pi}}(\mathbf{k})], \tag{71}$$

where $\omega \coloneqq |\mathbf{k}|$ and $\mathbf{e}_\lambda(\mathbf{k})$ is a unit polarization vector orthogonal to $\mathbf{k}$ (Fourier transforms are denoted with a tilde). From $\tilde{\mathbf{\Pi}} = -\tilde{\mathbf{E}}_{\mathrm{T}} - \alpha\tilde{\mathbf{P}}_{\mathrm{T}}$, it is clear that for each different fixed value of α the photon number operator $n = \sum_{\mathbf{k}\lambda} a^\dagger_\lambda(\mathbf{k})a_\lambda(\mathbf{k})$ represents a different gauge-invariant observable.

- Photons defined using the gauge-invariant observable $\mathcal{O} = -\mathbf{E}_{\mathrm{T}} - \alpha\mathbf{P}_{\mathrm{T}}$, which in the gauge α is represented by the operator $\mathbf{\Pi}$, are said to be defined relative to the gauge α.

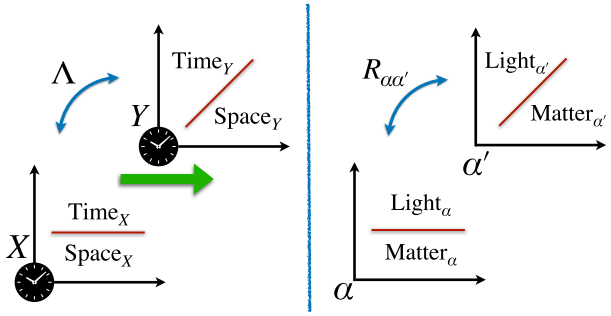

FIG. 3. The analogy between the relativity of space and time when partitioning spacetime, and the relativity of QED subsystems when partitioning the QED Hilbert space. Left panel: the Lorentz transformation Λ mixes space$_X$ and time$_X$ in transforming to the comoving frame Y. The relevant definition of time for the prediction of time intervals measured by a clock at rest in frame X is time$_X$. Right panel: the unitary gauge-fixing transformation $R_{\alpha\alpha'}$ mixes matter$_\alpha$ and light$_\alpha$ in moving to frame α'.

The eigenstates of the corresponding number operator n are a basis for the light Hilbert space, which is therefore defined relative to a choice of gauge. We can express this relativity symbolically by writing the subsystem label "light" or "photons" as a function of the observable that defines it, for instance, in the gauge α "light" $=$ light$(\mathbf{E}_{\mathrm{T}} + \alpha\mathbf{P}_{\mathrm{T}}) =:$ light$_\alpha$ and "photons" $=$ photons$_\alpha$. As an example, suppose that in a given experiment the observable $\mathbf{E}_{\mathrm{T}}$ is measurable. In this situation light$_0$ is then a relevant mathematical subsystem. This relativity for the case of photons applies to any subsystem property defined in terms of the canonical momenta. To summarize, according to the postulates of quantum theory, QED subsystems are defined relative to the choice of gauge (Stokes and Nazir, 2019).

D. Implications of subsystem gauge relativity

Predictions are necessarily gauge invariant when they pertain entirely to gauge-invariant objects. An example is the mechanical momentum $m\dot{\mathbf{r}} = \mathbf{p} - q\mathbf{A}(\mathbf{r})$, which is represented by the gauge-covariant derivative $-i\nabla - q\mathbf{A}(\mathbf{r})$ acting on position space wave functions. Scully and Zubairy (1997) therefore argued that only this momentum is physical, unlike the canonical momentum $\mathbf{p}$. Similarly, Schwinger (1951) favored the use of only gauge-covariant quantities in the calculation of relativistic vacuum effects. Yet, once the gauge has been fixed every operator within the theory represents an observable that is a known function of manifestly gauge-invariant observables (Fig. 2). Physical predictions will therefore be gauge invariant [cf. Eq. (70)] provided that approximations that ruin gauge invariance are avoided and that they are calculated properly. For example, when dealing with time-dependent interactions one must take into account the time dependence of gauge transformations, as noted by Settineri *et al.* (2021) and Stokes and Nazir (2021b).

Subsystem gauge relativity means that the light and matter quantum subsystems are defined by different gauge-invariant observables in each different gauge. A subsystem property such as the degree of light-matter entanglement constitutes two different gauge-invariant physical predictions when

calculated in two different gauges. This is a form of linear-space relativity analogous to that encountered in theories of space and time (Fig. 3).

Within sufficiently strong-coupling or non-Markovian regimes, the relativity of light and matter quantum subsystems cannot be ignored. Unlike in special relativity, determining which theoretical definition of a photon is the most relevant one for predicting experimental outcomes is not necessarily straightforward, because the task is intimately related to the interpretation of virtual processes and spacetime localization properties. It is also far from clear that the most relevant definition of photon is independent of the given experiment. Settineri *et al.* (2021) assumed that a photodetector registers photons defined by the gauge-invariant transverse electric field $\mathbf{E}_{\mathrm{T}}$. Given this assumption about which physical observable is relevant, one can calculate the rate of photodetection as a unique physical prediction in any gauge for both time-dependent and time-independent interactions. In Glauber's original theory, however, the total electric field $\mathbf{E}=\mathbf{E}_{\mathrm{T}}+\mathbf{E}_{\mathrm{L}}$ was used (Glauber, 1963; Milonni, James, and Fearn, 1995) and this field is transverse only when there are no charges present. Indeed, it has been argued that the transverse displacement field $\mathbf{D}_{\mathrm{T}}=\mathbf{E}_{\mathrm{T}}+\mathbf{P}_{\mathrm{T}}$ provides a more relevant definition because its source component equals the source component of $\mathbf{E}$ away from the source, and it is therefore local, unlike $\mathbf{E}_{\mathrm{T}}$ (Cohen-Tannoudji, Dupont-Roc, and Grynberg, 1989; Biswas *et al.*, 1990; Milonni, James, and Fearn, 1995; Power and Thirunamachandran, 1997, 1999a, 1999b; Sabín *et al.*, 2011). In particular, it has been known for six decades that photons defined relative to the multipolar gauge, i.e., in terms of $\mathbf{D}_{\mathrm{T}}$, are able to provide a natural line shape prediction that is in sufficient agreement with early experiments to rule out the corresponding prediction for the same experiments when photons are defined using $\mathbf{E}_{\mathrm{T}}$ (Power and Zienau, 1959; Fried, 1973; Davidovich and Nussenzveig, 1980; Milonni, Cook, and Ackerhalt, 1989; Woolley, 2000; Stokes, 2013). For these specific experiments the multipolar-gauge subsystems are evidently more operationally relevant than the Coulomb-gauge subsystems. Predictions of radiation spectra are discussed further in Secs. VI.B and VII.D.

The multipolar gauge $\alpha=1$ defines a dipole$_1$ that is purely mechanical, i.e., completely bare; see Sec. II.E. However, one often views physical atoms as being dressed by virtual photons, and this is more consistent with definitions provided by $\alpha\neq 1$, where the dipole is instead a delocalized dressed object. Only the localized dipole$_1$ does not respond instantaneously to a test charge placed away from its center at $\mathbf{0}$ (Cohen-Tannoudji, Dupont-Roc, and Grynberg, 1989; Biswas *et al.*, 1990; Milonni, James, and Fearn, 1995; Power and Thirunamachandran, 1997, 1999a, 1999b; Sabín *et al.*, 2011). In gauges $\alpha\neq 1$, the extent of the apparently instantaneous but typically small response of a test charge distribution to the field of the α-gauge dipole could be interpreted as simply a measure of the dressed dipole's delocalization due to its own virtual cloud of photons (Hegerfeldt, 1994). These points are discussed in the context of photodetection theory in Sec. VI.

For given values of the remaining model parameters, it is often possible to choose an intermediate value of α denoted as α_{JC}, which lies between 0 and 1, and for which ground-state virtual photons are highly suppressed (Stokes and Nazir, 2019). This representation is defined in Secs. IV.F, V.C.1, and VI.A.3, where the choice of notation α_{JC} is explained. The representation can be interpreted as one in which virtual photons have been absorbed into the definitions of the quantum subsystems. The physical meanings of the different mathematical definitions of light and matter are evidently closely related to virtual photons and processes.

Finally, we note that a prosaic implication of subsystem gauge relativity is that approximations performed on the subsystems can ruin the gauge invariance of the theory. A well-known example is the truncation of the material system to a finite number of levels (De Bernardis *et al.*, 2018; Roth, Hassler, and DiVincenzo, 2019; Stefano *et al.*, 2019; Stokes and Nazir, 2019). Because matter is defined differently in different gauges, the truncation generally constitutes a significantly different physical procedure in different gauges. This is discussed in detail in Sec. IV.

E. Canonical transformations in quantum field theory and unitary inequivalence

In the preceding development of nonrelativistic QED, the gauge-fixing transformations $U_{gg'}$ defined in Eq. (40) possess the form e^{iS}, with S Hermitian, and one can verify that $U_{gg'}$ preserves the canonical commutation relations. However, the gauge invariance identified in Sec. III.B comes with a certain caveat, namely, that the transformation $U_{gg'}$ is unitary only in form. By this we mean that although formally $U_{gg'}U_{gg'}^{\dagger}=I$, rigorously establishing the unitarity of canonical transformations in quantum field theory is nontrivial because one often encounters generators S that are too poorly behaved to avoid the occurrence of infinite terms during the course of formal manipulations. This point is directly relevant when one considers $U_{gg'}$. Here we follow the intuitive heuristic discussion conducted by Umezawa (1995).

Consider the formally unitary transformation

$$U[\theta]=\exp\left[\int d^3k\sum_{\lambda}[\theta_\lambda(\mathbf{k})^{*}a_\lambda(\mathbf{k})-\theta_\lambda(\mathbf{k})a_\lambda^{\dagger}(\mathbf{k})]\right], \tag{72}$$

where θ is an arbitrary function $\mathbf{k}\rightarrow\theta(\mathbf{k})$. If one assumes that $[\theta_\lambda(\mathbf{k}),\theta_{\lambda'}(\mathbf{k}')]=0$, then since $a'_\lambda(\mathbf{k})=U[\theta]a_\lambda(\mathbf{k})U[\theta]^{\dagger}=a_\lambda(\mathbf{k})+\theta_\lambda(\mathbf{k})$ the transformation is canonical, that is, $[a_\lambda(\mathbf{k}),a_\lambda^{\dagger}(\mathbf{k}')]=\delta_{\lambda\lambda'}\delta(\mathbf{k}-\mathbf{k}')\Leftrightarrow[a'_\lambda(\mathbf{k}),a'^{\dagger}_\lambda(\mathbf{k}')]=\delta_{\lambda\lambda'}\delta(\mathbf{k}-\mathbf{k}')$. Denoting the vacuum annihilated by $a_\lambda(\mathbf{k})$ by $|0\rangle$ and assuming that $\langle 0|0\rangle=1$, we see that according to Eq. (72) the vacuum $|0'\rangle=U[\theta]|0\rangle$ annihilated by $a'(\mathbf{k})$ is also formally normalized ($\langle 0'|0'\rangle=1$). One finds in addition, however, that

$$|0'\rangle=\tau\exp\left[-\int d^3k\sum_{\lambda}\theta_\lambda(\mathbf{k})a_\lambda^{\dagger}(\mathbf{k})\right]|0\rangle, \tag{73}$$

where

$$\tau=\exp\left[-\frac{1}{2}\int d^3k\sum_{\lambda}|\theta_\lambda(\mathbf{k})|^2\right], \tag{74}$$

such that if $\int d^3k|\theta_\lambda(\mathbf{k})|^2=\infty$, then the prefactor $\tau=\langle 0|0'\rangle$ is vanishingly small. It would then follow that $\langle\psi|0'\rangle=0$, where $|\psi\rangle$ is any Fock state generated by applying the operators $a_\lambda^\dagger(\mathbf{k})$ to the vacuum $|0\rangle$. From this it would follow that the vacuum $|0'\rangle$ and the Fock states generated from it using the operators $a'^\dagger_\lambda(\mathbf{k})$ cannot be expressed as linear combinations of the Fock states generated using $a_\lambda^\dagger(\mathbf{k})$ and $|0\rangle$. The two bases are then said to be inequivalent (Umezawa, 1995).

We now turn our attention to the PZW gauge-fixing transformation R_{01}, which can be written in the form of Eq. (72) with $\theta_\lambda(\mathbf{k})=i\mathbf{e}_\lambda(\mathbf{k})\cdot\tilde{\mathbf{P}}(\mathbf{k})/\sqrt{2\omega}$, where $\tilde{\mathbf{P}}$ denotes the Fourier transform of the multipolar polarization. In this case

$$-\ln\tau=\frac{1}{2}\langle 0'|\int d^3k\sum_\lambda a_\lambda^\dagger(\mathbf{k})a_\lambda(\mathbf{k})|0'\rangle$$
$$=\int d^3k|\tilde{\mathbf{P}}_{\mathrm{T}}(\mathbf{k})|^2/4\omega \tag{75}$$

is half the average number of photons$_0$ in the vacuum $|0'\rangle$. Woolley (2020) found that for a two-charge system $\tau\to 0$ in the point-charge limit; that is, the vacua of the Coulomb and multipolar gauges do indeed become orthogonal. Physically, the vacua of the Coulomb gauge ($\alpha=0$) and the multipolar gauge ($\alpha=1$) must contain an infinite number of photons$_1$ (multipolar photons) and photons$_0$ (Coulomb-gauge photons), respectively, so the two vacua cannot be simultaneously meaningful. According to a simple second-order perturbation theory calculation, the ground state of the Hamiltonian in Eq. (22) contains both photons$_0$ and photons$_1$. Identifying the ground state of the Hamiltonian as the physical vacuum is the underlying idea of the Jaynes-Cummings (JC) gauge mentioned in Sec. III.D. This is discussed further in Secs. IV.F, V.C.1, and VI.A.3.

Unitary inequivalence results from the singular nature of $\mathbf{P}(\mathbf{x})$ within the PZW transformation. This localization of $\mathbf{P}(\mathbf{x})=\int d^3k\tilde{\mathbf{P}}(\mathbf{k})e^{i\mathbf{k}\cdot\mathbf{x}}/\sqrt{(2\pi)^3}$ requires that all wave vectors $\mathbf{k}$ are retained within the Fourier transformation. However, in nonrelativistic QED one can argue *a priori* that relativistic modes are not properly described (Cohen-Tannoudji, Dupont-Roc, and Grynberg, 1989). The multiplication of $\tilde{\mathbf{P}}_{\mathrm{T}}(\mathbf{k})$ by a form factor such as $\ell_M(k)=k_M^2/(k^2+k_m^2)$, as described in Secs. II.D.2 and II.E, removes the contributions of relativistic wave vectors such that $\int d^3k|\tilde{\mathbf{P}}_{\mathrm{T}}(\mathbf{k})|^2<\infty$. Similarly, $\langle 0|0'\rangle$ vanishes only in the point-charge limit (Woolley, 2020), yet the elimination of relativistic wave vector contributions to the point-charge density ρ is equivalent to considering extended charge distributions, which yield a more rigorously well-defined quantum theory (Spohn, 2004). In this review we consider formally unitary gauge-fixing transformations and assume that gauge invariance as defined in Sec. III.B holds, but with the understanding that when dealing with quantum fields strict unitarity may require one to invoke suitable regularization procedures.

We note finally that although after a suitable regularization that gives $\langle 0|0'\rangle\neq 0$ the vacua $|0\rangle$ and $|0'\rangle$ contain only a finite number of photons$_1$ and photons$_0$, respectively, the two vacua remain physically distinct and it remains to determine which, if either, is relevant in a given situation. As discussed in more detail in Sec. V, the gauge nonrelativistic property of the QED S matrix under only formally unitary gauge-fixing transformations $U=e^{iS}$ can be proved quite generally (Craig and Thirunamachandran, 1998; Woolley, 2000). These points demonstrate that while the occurrence of unitarily inequivalent representations of the CCR algebra is of importance with regard to the technical challenge of establishing the strict gauge invariance of predictions, it is of far less importance with regard to the occurrence or otherwise of gauge relativity.

F. Modal restrictions and transversality

Restrictions on the number of photonic modes are extremely common in light-matter physics. However, retaining all modes is necessary to maintain spacetime localization and causal wave propagation. In particular, the Green's function for the wave operator receives contributions from all $\mathbf{k}$-space modes. A modal restriction should be understood as a statement about which frequencies are dominant within a given light-matter interaction Hamiltonian.

1. Significance of transversality

We begin by noting that the transversality of canonical fields is closely related to gauge freedom. Only transverse fields can be used to define unconstrained physical photons as in Eq. (71). This feature is fundamental and persists in the presence of background media [see note VII of the Supplemental Material (245)], such as those relevant in numerous artificial photonic systems that realize large coupling strengths (Ciuti, Bastard, and Carusotto, 2005; Ciuti and Carusotto, 2006; Todorov *et al.*, 2010; Bamba and Ogawa, 2012, 2013, 2014a). Relativistic particles can be specified via the unitary representations of the Poincaré group (Bargmann and Wigner, 1948), which are labeled by two numbers, "mass" $m\geq 0$ and integer or half odd integer "spin" s. Massless fields possess only two independent helicities $-s,+s$ obtained from the projection of the spin s onto the axis of particle motion (Hassani, 2013). In particular, the massless spin-1 Maxwell field supports the two independent polarizations of a photon. Scalar and longitudinal photons can also be defined, as in the Lorenz gauge (Lorenz, 1867), but such photons are not unconstrained. They satisfy a nondynamical constraint (Lorenz subsidiary condition) whose derivative in time is Gauss's law (Cohen-Tannoudji, Dupont-Roc, and Grynberg, 1989).

Gauss's law generates gauge-symmetry transformations and its derivative in time is the continuity equation for electric charge, which is the conserved quantity associated with gauge symmetry. It specifies $\mathbf{E}_{\mathrm{L}}$ as a function of ρ, telling us that longitudinal photons are not independent. Specifically, an analog of Eq. (71) may be written as

$$a_{\mathrm{L}}(\mathbf{k}):=-\frac{i}{\sqrt{2\omega}}\hat{\mathbf{k}}\cdot\tilde{\mathbf{E}}(\mathbf{k})=-\frac{\tilde{\rho}(\mathbf{k})}{\sqrt{2\omega^3}}. \tag{76}$$

Although $\mathbf{E}_{\mathrm{T}}$ is the part of the electric field not constrained by Gauss's law, it is by fundamental assumption that the total electric field $\mathbf{E}$ is local. It follows that the fields $\mathbf{E}_{\mathrm{L}}$ and,

notably, $\mathbf{E}_{\mathrm{T}} = \mathbf{E} - \mathbf{E}_{\mathrm{L}}$ are both nonlocal [cf. Eq. (7)], and away from a localized source they respond instantaneously to changes in the source (Cohen-Tannoudji, Dupont-Roc, and Grynberg, 1989; Craig and Thirunamachandran, 1998). The multipolar-gauge momentum $\mathbf{\Pi} = -\mathbf{D}_{\mathrm{T}}$ offers the best possible representation of the nontransverse local field $\mathbf{E}$ by an unconstrained transverse field that can then be used to define unconstrained photons (Cohen-Tannoudji, Dupont-Roc, and Grynberg, 1989). Specifically, $\mathbf{P}_{\mathrm{L}} = -\mathbf{E}_{\mathrm{L}}$ implies that $\mathbf{D} \coloneqq \mathbf{E} + \mathbf{P} = (\mathbf{E}_{\mathrm{T}} + \mathbf{E}_{\mathrm{L}}) + (\mathbf{P}_{\mathrm{T}} - \mathbf{E}_{\mathrm{L}}) \equiv \mathbf{D}_{\mathrm{T}}$ and since $\mathbf{P}$ vanishes outside of a charge distribution we have $\mathbf{D} \equiv \mathbf{D}_{\mathrm{T}} = \mathbf{E}$ at all such points. It is not the case, however, that $\mathbf{E} = \mathbf{E}_{\mathrm{T}}$ or that $\mathbf{P} = \mathbf{P}_{\mathrm{T}}$.

In the case of a dipole at $\mathbf{0}$, the multipolar polarization is $\mathbf{P} = q\mathbf{r}\delta(\mathbf{x})$, whereas $\mathbf{P}_{\mathrm{T}}(\mathbf{x}) = q\mathbf{r}\cdot\delta^{\mathrm{T}}(\mathbf{x})$. The transverse dyadic $\delta^{\mathrm{T}}(\mathbf{x})$ is not purely singular: rather, it decays as $1/x^3$ away from 0. From elementary electrostatics we know that $\mathbf{E}_{\mathrm{L}}$ decays as $1/x^3$ away from a dipole at $\mathbf{0}$, and for a dipole we do indeed have $\mathbf{P}_{\mathrm{T}} = \mathbf{E}_{\mathrm{L}}$ for $\mathbf{x} \neq \mathbf{0}$ [i.e., $\mathbf{P}(\mathbf{x}) = q\mathbf{r}\delta(\mathbf{x}) = 0$ for $\mathbf{x} \neq \mathbf{0}$]. For any α the field $\mathbf{\Pi}$ can be expanded in terms of photons using Eq. (71). However, for different α these fields are related by the nonlocal field $\mathbf{P}_{\mathrm{T}}$.

For a transverse field, the mode functions $\mathbf{f}_\lambda(\mathbf{k},\mathbf{x}) = \mathbf{e}_\lambda(\mathbf{k})e^{i\mathbf{k}\cdot\mathbf{x}}/\sqrt{(2\pi)^3}$ of a canonical mode expansion are not complete with respect to the usual inner product in $L^2(\mathbb{R}^3)$, because $\{\mathbf{e}_\lambda(\mathbf{k})\}$ is an orthonormal basis in the two-dimensional plane orthogonal to $\mathbf{k}$. They instead furnish the following representation of the transverse delta function:

$$\int d^3k \sum_{\lambda=1,2} \mathbf{f}_\lambda(\mathbf{k},\mathbf{x})^* \mathbf{f}_\lambda(\mathbf{k},\mathbf{x}') = \delta^{\mathrm{T}}(\mathbf{x}-\mathbf{x}'). \tag{77}$$

To obtain a representation of $\delta(\mathbf{x}-\mathbf{x}')$, one must include the vector $\hat{\mathbf{k}}$ in Fourier space to obtain the three-dimensional basis $\{\hat{\mathbf{k}}, \mathbf{e}_\lambda(\mathbf{k})\}$. If the longitudinal eigenfrequency is set to vanish ($\omega_{\mathrm{L}} \equiv 0$), then one can expand $\mathbf{\Pi}$ using the complete set of mode functions. However, the operators $a_{\mathrm{L}}(\mathbf{k})$ have completely arbitrary definitions and cannot contribute to physical predictions.

2. Modal restriction

Ultrastrong coupling between light and matter arises in artificial systems in which the set of photonic modes is altered and often restricted. Theoretically, care must be taken when carrying out such restrictions. To demonstrate this we choose the multipolar gauge such that $\mathbf{\Pi} = -\mathbf{D}_{\mathrm{T}}$, implying that the Coulomb-gauge momentum $-\mathbf{E}_{\mathrm{T}}$ is represented by the operator $\mathbf{\Pi}' = -\mathbf{E}_{\mathrm{T}} = R_{10}\mathbf{\Pi}R_{10}^\dagger = \mathbf{\Pi} + \mathbf{P}_{\mathrm{T}}$. Coulomb and multipolar-gauge transverse photonic operators $a'_\lambda(\mathbf{k})$ and $a_\lambda(\mathbf{k})$ are defined as in Eq. (71) using $\mathbf{\Pi}' = -\mathbf{E}_{\mathrm{T}}$ and $\mathbf{\Pi} = -\mathbf{D}_{\mathrm{T}}$, respectively. They are therefore related by

$$a'_\lambda(\mathbf{k}) = R_{10}a_\lambda(\mathbf{k})R_{10}^\dagger = a_\lambda(\mathbf{k}) + i\frac{q\mathbf{r}\cdot\mathbf{f}_\lambda(\mathbf{k},\mathbf{0})}{\sqrt{2\omega_\lambda}}. \tag{78}$$

For the unphysical longitudinal mode operators any relation can be specified. We note, however, that the right-hand side of Eq. (78) would be undefined for $\lambda = \mathrm{L}$ because $\omega_{\mathrm{L}} \equiv 0$. The total electric field is given by $\mathbf{E} = \mathbf{D}_{\mathrm{T}} - \mathbf{P} = -\mathbf{\Pi} - \mathbf{P} = -\mathbf{\Pi}' - \mathbf{P}_{\mathrm{L}}$, and $\mathbf{P}(\mathbf{x}) = q\mathbf{r}\delta(\mathbf{x})$ is fully localized. The electric field $\mathbf{E}$ is completely independent of the $a_{\mathrm{L}}(\mathbf{k})$, as any physical field must be.

When the modes are confined to a volume v with periodic boundary conditions, the mode functions become discrete [$\mathbf{f}_\lambda(\mathbf{k},\mathbf{x}) \to \mathbf{f}_{\mathbf{k}\lambda}(\mathbf{x})$] such that factors of $(2\pi)^3$ are replaced by v. For a field $\mathbf{F}$ the component associated with the wave vector $\mathbf{k}$ or mode $\mathbf{k}\lambda$ can be read off by expressing $\mathbf{F}$ as $\mathbf{F}(\mathbf{x}) = \sum_{\mathbf{k}} \mathbf{F}_{\mathbf{k}} = \sum_{\mathbf{k}\lambda} \mathbf{F}_{\mathbf{k}\lambda}$. For the transverse and longitudinal polarization fields we have $\mathbf{P}_{\mathrm{T}\mathbf{k}}(\mathbf{x}) = q\mathbf{e}_{\mathbf{k}\lambda}[(\mathbf{e}_{\mathbf{k}\lambda}\cdot\mathbf{r})/v]\cos\mathbf{k}\cdot\mathbf{x}$ and $\mathbf{P}_{\mathbf{L}\mathbf{k}} = -\mathbf{E}_{\mathbf{L}\mathbf{k}} = q\hat{\mathbf{k}}[(\hat{\mathbf{k}}\cdot\mathbf{r})/v]\cos(\mathbf{k}\cdot\mathbf{x})$, respectively, such that the restricted total polarization is $\mathbf{P}_{\mathbf{k}}(\mathbf{x}) = (q\mathbf{r}/v)\cos(\mathbf{k}\cdot\mathbf{x})$. For the total electric field we have $\mathbf{E}_{\mathbf{k}}(\mathbf{x}) = -\mathbf{\Pi}_{\mathbf{k}}(\mathbf{x}) - \mathbf{P}_{\mathbf{k}}(\mathbf{x})$. These single-mode restrictions can be implemented at the position $\mathbf{0}$ of a single dipole via the α-gauge theory presented in Secs. II.B and III. Since all algebraic and kinematic relations are preserved, so too is gauge invariance. The dipole approximated fields in Eqs. (29) and (31) are assumed to point in the direction $\boldsymbol{\varepsilon}$ of the mode polarization and, in this direction, have the following components (Stokes and Nazir, 2019, 2020b):

$$A_\alpha = (1-\alpha)A = \frac{1-\alpha}{\sqrt{2\omega v}}(a^\dagger + a), \tag{79}$$

$$P_{\mathrm{T}\alpha} = \frac{\alpha q x}{v}. \tag{80}$$

In Eqs. (79) and (80) $x = \boldsymbol{\varepsilon}\cdot\mathbf{r}$ and $A = \boldsymbol{\varepsilon}\cdot\mathbf{A}_{\mathrm{T}}(\mathbf{0})$, where $\boldsymbol{\varepsilon}$ is the unit polarization vector of the single transverse mode retained. The Hamiltonian reduces to a simple form that has now been used in a number of works (Roth, Hassler, and DiVincenzo, 2019; Stefano *et al.*, 2019; Stokes and Nazir, 2019, 2020b, 2021b); cf. Sec. IV.A. The gauge-fixing transformations in Eq. (32) remain unitary, becoming $R_{\alpha\alpha'} = \exp(i[\alpha-\alpha']qxA)$ (Stokes and Nazir, 2019, 2021b).

The restriction to a finite number of modes within the Hamiltonian of a light-matter system evidently must be understood as an assumption about which modes are dominant within the dipole-field interaction. This may be valid at the position of the dipole center $\mathbf{0}$ in the form $\mathbf{V}(\mathbf{0}) = \sum_{\mathbf{k}} \mathbf{V}_{\mathbf{k}}(\mathbf{0}) \approx \mathbf{V}_{\mathbf{k}}(\mathbf{0})$. However, the dipole's center $\mathbf{0}$ is also where the field cannot be measured by an external detector. For any $\mathbf{x}$ the field $\mathbf{E}_{\mathbf{k}}$ equals neither $-\mathbf{E}_{\mathrm{T}\mathbf{k}}$ nor $-\mathbf{D}_{\mathrm{T}\mathbf{k}}$. Because of Gauss's law, the electric field, whether restricted or not, cannot be expressed solely in terms of physical transverse photons. In particular, since $\mathbf{\Pi}_{\mathbf{k}}$ is orthogonal to $\mathbf{k}$, one cannot obtain $\mathbf{E}_{\mathbf{k}}$ by means of a unitary operator acting on $\mathbf{\Pi}_{\mathbf{k}}$.

The fully localized physical polarization $\mathbf{P}(\mathbf{x}) = \sum_{\mathbf{k}} \mathbf{P}_{\mathbf{k}}(\mathbf{x})$ cannot be elicited in a restricted space of wave vectors. A modal restriction at an arbitrary point $\mathbf{x} \neq \mathbf{0}$ will therefore violate the property $\mathbf{P}_{\mathrm{T}} = -\mathbf{P}_{\mathrm{L}}$ of the full theory. Naively restricting the polarization and electric fields to only one transverse mode $\mathbf{k}\lambda$ means that $\mathbf{P}_{\mathrm{L}\mathbf{k}}(\mathbf{x}) \equiv \mathbf{0}$, and we obtain $\mathbf{E}_{\mathbf{k}}(\mathbf{x}) \equiv -\mathbf{\Pi}'_{\mathbf{k}\lambda}(\mathbf{x}) = -\mathbf{\Pi}_{\mathbf{k}}(\mathbf{x}) - \mathbf{P}_{\mathrm{T}\mathbf{k}}(\mathbf{x})$. This yields a theory without $\mathbf{E}_{\mathrm{L}}$, which can therefore be valid only in the far field. In the far field where $\mathbf{E}_{\mathrm{L}} = \mathbf{P}_{\mathrm{T}}$ vanishes, we have $-\mathbf{\Pi}' = \mathbf{E}_{\mathrm{T}} \approx \mathbf{E} = -\mathbf{\Pi}$ whether or not the modes are restricted. If we

instead use the fact that $\mathbf{\Pi}(\mathbf{x}) = -\mathbf{E}(\mathbf{x})$ for $\mathbf{x} \neq \mathbf{0}$ and then restrict our attention to one transverse mode, we obtain the differing result $\mathbf{E}_{\mathbf{k}}(\mathbf{x}) \equiv -\mathbf{\Pi}_{\mathbf{k}}(\mathbf{x}) = -\mathbf{\Pi}'_{\mathbf{k}}(\mathbf{x}) + \mathbf{P}_{\mathrm{T}\mathbf{k}}(\mathbf{x})$. This single-mode limit respects the equalities $\mathbf{E} = -\mathbf{\Pi} = -\mathbf{\Pi}' + \mathbf{P}_{\mathrm{T}}$ holding for $\mathbf{x} \neq \mathbf{0}$ in the unrestricted theory. Within the light-matter interaction, Hamiltonian fields are evaluated at $\mathbf{x} = \mathbf{0}$, so these considerations do not apply.

Evidently, different implementations of a modal restriction can result in altogether different identifications of the same physical field, so care must be taken. In the previously mentioned case regarding the electric field $\mathbf{E}$, we have fundamentally that at all points $\mathbf{x}$ outside of a charge distribution, which is where the field can be measured by an external detector, the multipolar polarization vanishes, implying that at such points $\mathbf{\Pi}(\mathbf{x}) = -\mathbf{E}(\mathbf{x})$ in and only in the multipolar gauge. We should not expect a modal restriction in which this is no longer the case to offer a generally robust approximation of the unrestricted theory for describing measurements involving $\mathbf{E}(\mathbf{x})$. In particular, the Glauber intensity at $(t, \mathbf{x})$ is given within the single-mode limit that respects the fundamental equalities of the multimode theory as

$$\langle \mathbf{E}^{(-)}_{\mathbf{k}\lambda}(t, \mathbf{x}) \cdot \mathbf{E}^{(+)}_{\mathbf{k}\lambda}(t, \mathbf{x}) \rangle = \frac{\omega}{2v} \langle a^{\dagger}_{\mathbf{k}\lambda}(t) a_{\mathbf{k}\lambda}(t) \rangle, \tag{81}$$

where $a_{\mathbf{k}\lambda}$ is the multipolar-gauge photonic operator. Irrespective of the modal restrictions, the Glauber intensity is not proportional to the photon number operator defined relative to the Coulomb gauge except in the far field, where $\mathbf{E} \approx \mathbf{E}_{\mathrm{T}}$. Photodetection is discussed in more detail in Sec. V.

G. Simple extension to superconducting circuits

The arbitrary-gauge formalism is readily adapted to describe circuit QED systems, which we now review. Vool and Devoret (2017) provided an introductory review of circuit QED, while a more recent review was given by Blais *et al.* (2021). Conventional descriptions of superconducting circuits employ the lumped-element model, which results from Kirchoff's assumptions applied to Maxwell theory. Consider a node defined as the meeting point of N conducting wire branches, outside of which there is no current. Bounding the node is a closed surface $\mathcal{S}$ containing a region v with outward normal $\hat{\mathbf{n}}$. The continuity equation $\partial_\nu j^\nu = 0$ and the divergence theorem yield

$$\sum_{\mu=1}^{N} I_\mu(t) \equiv \sum_{\mu=1}^{N} \int_{\mathcal{S}_\mu} dS \hat{\mathbf{n}} \cdot \mathbf{J}(t, \mathbf{x}) = -\frac{dQ(t)}{dt}, \tag{82}$$

$$Q(t) = \int_v d^3x \, \rho(t, \mathbf{x}), \tag{83}$$

where $\mathcal{S}_\mu$ is the subsurface of $\mathcal{S}$ intersecting the μth wire, I_μ is the current entering v through the μth wire, and $Q(t)$ is the total charge within the region v containing the node. Equation (82) assumes that $\mathbf{J}(t, \mathbf{x}) = 0$ for all $\mathbf{x} \in \mathcal{S} / \cup_\mu \mathcal{S}_\mu$ (there is no current outside the conducting wires). Kirchoff further assumed a local steady-state current condition within v, namely, $dQ(t)/dt = 0$, yielding the current law

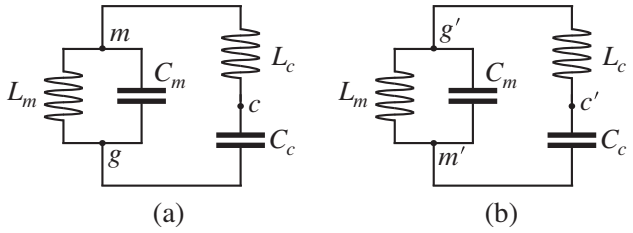

FIG. 4. Circuit diagram for a parallel LC oscillator coupled to a series LC oscillator. There are three nodes within the circuit. The two panels each provide a distinct labeling of the nodes corresponding to different specifications of the ground flux. As a result, they depict two different divisions of the circuit into subsystems. Specifically, these are the two extreme cases of (a) fully inductive coupling whereby the ground flux is specified as the flux associated with the node that is labeled g and (b) fully capacitive coupling whereby the ground flux is specified as the flux associated with the node that is labeled g'.

$$\sum_{\mu=1}^{N} I_\mu(t) = 0. \tag{84}$$

Arbitrary lumped-element circuits can be considered collections of nodes joined by (super)conducting branches, with Kirchoff's law [Eq, (84)] satisfied at each node. As a nontrivial example we consider the coupled LC-oscillator circuit depicted in Fig. 4. As basic dynamical variables we take the node fluxes denoted as ϕ_k. The current into node k through a branch $j \to k$ with an inductor connecting node k to node j is $I_{j\to k} = (\phi_k - \phi_j)/L$, where L is the inductance of the inductor. The current into node k through a branch $j \to k$ with a capacitor connecting node k to node j is $I_{j\to k} = C(\ddot{\phi}_k - \ddot{\phi}_j)$, where C is the capacitance of the capacitor. Since only flux differences are of importance, we can specify the flux zero point arbitrarily. This is the so-called ground flux such that $\phi_g = 0$. As special cases, we can choose this flux zero point to be the flux of one of the circuit nodes depicted in Fig. 4. Figures 4(a) and 4(b) give two different specifications of which node possesses the ground flux.

In the circuit of Fig. 4(a) there are two nonground nodes labeled m and c. Kirchoff's law [Eq. (84)] yields the following equations of motion:

$$0 = I_{g\to m} + I_{c\to m} = C_m \ddot{\phi}_m + \frac{\phi_m}{L_m} + \frac{\phi_m - \phi_c}{L_c}, \tag{85}$$

$$0 = I_{g\to c} + I_{m\to c} = C_c \ddot{\phi}_c + \frac{\phi_c - \phi_m}{L_c}. \tag{86}$$

Equations (85) and (86) are obtained from the Lagrangian

$$L = \frac{1}{2}\left[C_m \dot{\phi}_m^2 - \frac{\phi_m^2}{L_m} + C_c \dot{\phi}_c^2 - \frac{(\phi_c - \phi_m)^2}{L_c}\right] \tag{87}$$

or the corresponding Hamiltonian

$$H = \frac{1}{2}\left[\frac{q_m^2}{C_m} + \frac{\phi_m^2}{L_m} + \frac{q_c^2}{C_c} + \frac{(\phi_c - \phi_m)^2}{L_c}\right], \tag{88}$$

where $q_x = \partial L/\partial\dot{\phi}_x$ are the node charges conjugate to the ϕ_x, with $x = m, c$. A node flux and its conjugate charge satisfy a canonical Lie bracket relation that generates the dynamics in conjunction with the Hamiltonian. In particular, in the quantum theory $[\phi_x, q_{x'}] = i\delta_{xx'}$.

We now consider the relabeling of the nodes depicted in Fig. 4(b). The ground node has the flux $\phi_{g'} = 0$, and the nonground nodes m' and c' are now connected by the capacitance C_c rather than by the inductance L_c. Since the physical currents through the branches must stay the same, we obtain the following coordinate relations:

$$\phi_{m'} = -\phi_m, \tag{89}$$

$$\phi_{c'} = \phi_c - \phi_m. \tag{90}$$

Either ϕ_m or $\phi_{m'}$ can be used as a coordinate with $\phi_{c'}$. We choose ϕ_m. The sum of Eqs. (85) and (86) can be expressed as

$$0 = C_m\ddot{\phi}_m + C_c(\ddot{\phi}_m + \ddot{\phi}_{c'}) + \frac{\phi_m}{L_m}, \tag{91}$$

and this equation together with Eq. (86) is obtained from the Lagrangian (87) or the Hamiltonian (88), with ϕ_m and $\phi_{c'}$ taken as dynamical coordinates. At the Hamiltonian level the primed and unprimed canonical operators are related by a gauge-fixing transformation as

$$q_{m'} = R_{10}q_mR_{10}^\dagger, \tag{92}$$

$$\phi_{c'} = R_{10}\phi_cR_{10}^\dagger, \tag{93}$$

where $R_{10} \coloneqq e^{-iq_c\phi_m}$. This is analogous to the PZW transformation between the charge (Coulomb) gauge and the flux (multipolar) gauge.

Note that within the previous derivation we have adopted a passive view of rotations within the operator algebra (see Sec. II.F.4), by which we mean that the same Hamiltonian has been expressed in terms of alternative canonical operators that belong to different gauges. Equivalently, we may adopt an active perspective as in previous sections, where the Hamiltonian H is actively rotated using gauge-fixing transformations, thereby yielding new Hamiltonians that are all expressed in terms of the same canonical operators. The extension to arbitrary gauges is straightforward via the gauge-fixing transformation $R_{\alpha\alpha'} \coloneqq e^{-i(\alpha-\alpha')q_c\phi_m}$. We note that gauges specified by $\alpha \neq 0, 1$ do not correspond to a definite specification of one of the nodes within Fig. 4 as possessing the ground flux $\phi_g = 0$. Instead, the ground flux is specified as some combination of the fluxes associated with the three nodes.

The basic nonlinear element in superconducting circuits is the Josephson junction (Josephson, 1962). These junctions are typically realized using two conducting materials separated by a thin insulator gap. Quantum mechanically, electron tunneling across the junction is possible, with the tunneling charge flowing in units of Cooper pairs as $Q = 2qN$, where N denotes the number of Cooper pairs on one side of the junction. The junction Hamiltonian is

$$H_J = -\frac{E_J}{2}\sum_N(|N\rangle\langle N+1| + |N+1\rangle\langle N|), \tag{94}$$

where the energy E_J determines the coupling strength across the junction. Introducing the phase variable ϕ_m conjugate to Q through Fourier transformation as $|\phi_m\rangle = \sum_{N=-\infty}^{\infty} e^{2iq\phi_mN}|N\rangle$, one can express the junction Hamiltonian as $H_J = -E_J\cos[2q\phi_m]$.

The previous formalism is easily extended to arbitrary circuits constructed from capacitors, inductors, and Josephson junctions. For example, by adding a Josephson junction connecting the ground node g to the node m in Fig. 4, one obtains the light-matter Hamiltonian

$$H' = H - E_J\cos[2q\phi_m]. \tag{95}$$

The Hamiltonian H' possesses the same structure as the cavity QED Hamiltonian considered in Sec. II.A in which the material potential is arbitrary.

We have seen that the choice of gauge is determined by the choice of ground flux, and that arbitrary choices of gauge selected by a parameter α may be considered. Gauge-fixing transformations are directly analogous to those encountered in conventional QED, and as such they are nonlocal with respect to Hilbert space tensor-product structure. A circuit may be dividing into physically distinct canonical subcircuits arbitrarily, and this division is directly controlled by the choice of gauge.

IV. MATERIAL TRUNCATION AND GAUGE NONINVARIANCE

Material energy level truncation is a commonly adopted procedure that nevertheless breaks the gauge invariance of QED by fundamentally modifying the algebra of material operators. This was discussed in the context of strong and ultrastrong-coupling by De Bernardis, Jaako, and Rabl (2018), De Bernardis *et al.* (2018), Roth, Hassler, and DiVincenzo (2019), Stefano *et al.* (2019), Stokes and Nazir (2019, 2020a, 2020b, 2021b), Taylor *et al.* (2020), Ashida, İmamoğlu, and Demler (2021), and Settineri *et al.* (2021). Here we review the implications of the resulting gauge noninvariances, which as explained in Sec. III.B are related to, but not synonymous with, gauge ambiguities. We review various proposed theoretical approaches for obtaining truncated models.

A. Single dipole interacting with a single cavity mode

The EDA and single-mode approximation can be performed while preserving all algebraic properties of the theory, thereby preserving gauge invariance (Stokes and Nazir, 2019, 2020b, 2021b). The dipole is assumed to be located at the origin $\mathbf{0}$ and for simplicity the canonical operators are assumed to point in the direction $\boldsymbol{\varepsilon}$ of polarization of the single mode. We define $x = \boldsymbol{\varepsilon}\cdot\mathbf{r}$ and $A = \boldsymbol{\varepsilon}\cdot\mathbf{A}_\mathrm{T}$ and denote by p and Π the corresponding dipole and cavity canonical momenta, such that $[x, p] = i$ and $[A, \Pi] = i/v$, with v the cavity volume. Details of the EDA and single-mode restriction are given in Secs. II.B and III.F.2, respectively.

The α gauge continues to be specified by its vector potential $\mathbf{A}_\alpha = \boldsymbol{\varepsilon} A_\alpha$ and material polarization $\mathbf{P}_{\mathrm{T}\alpha} = \boldsymbol{\varepsilon} P_{\mathrm{T}\alpha}$, which are given by Eqs. (79) and (80), respectively. The definition of gauge freedom given by Eqs. (27) and (28) now reads

$$R_{\alpha\alpha'} p R^\dagger_{\alpha\alpha'} = p - (\alpha - \alpha') qA, \tag{96}$$

$$R_{\alpha\alpha'} \Pi R^\dagger_{\alpha\alpha'} = \Pi - (\alpha - \alpha') \frac{qx}{v}. \tag{97}$$

Since gauge-fixing transformations remain unitary, the gauge invariance of the theory is preserved. The Hamiltonian is as always the total energy (Stokes and Nazir, 2019, 2021b),

$$H_\alpha = \mathcal{H}_m(A_\alpha) + \mathcal{H}_{\mathrm{ph},\alpha}, \tag{98}$$

$$\mathcal{H}_m(A_\alpha) \coloneqq \frac{1}{2} m\dot{x}^2 + V(x) = \frac{1}{2m}(p - qA_\alpha)^2 + V(x), \tag{99}$$

$$\mathcal{H}_{\mathrm{ph},\alpha} \coloneqq \frac{v}{2}(E_{\mathrm{T}}^2 + \omega A^2) = \frac{v}{2}[(\Pi + P_{\mathrm{T}\alpha})^2 + \omega^2 A^2], \tag{100}$$

where $\dot{x} = -i[x, H_\alpha]$ and $E_{\mathrm{T}} = -\dot{A}_{\mathrm{T}} = i[A_{\mathrm{T}}, H_\alpha]$. All three energies are gauge invariant,

$$\mathcal{X}_{\alpha'} = R_{\alpha\alpha'} \mathcal{X}_\alpha R^\dagger_{\alpha\alpha'}, \tag{101}$$

where $\mathcal{X}_\alpha = H_\alpha, \mathcal{H}_m(A_\alpha), \mathcal{H}_{\mathrm{ph},\alpha}$. Note also that, as discussed in Sec. II.B, within (and only within) the EDA the α-gauge mechanical momentum may be obtained from the canonical momentum $\mathbf{p}$ using $R_{1\alpha}$. For $\alpha = 1$, Eq. (101) with $\mathcal{X} = \mathcal{H}_m(A_\alpha)$ then has the following appearance of a unitary transformation applied to the free material Hamiltonian [Eq. (66)] (Stefano *et al.*, 2019):

$$\mathcal{H}_m(A_\alpha) = R_{1\alpha} H_m R^\dagger_{1\alpha}. \tag{102}$$

Equation (102) holds in and only in the EDA.

The transverse electromagnetic energy can be similarly written as $\mathcal{H}_{\mathrm{ph},\alpha} = R_{0\alpha} H_{\mathrm{ph}} R^\dagger_{0\alpha}$, where $H_{\mathrm{ph}} = \mathcal{H}_{\mathrm{ph},0} = (v/2)(\Pi^2 + \omega^2 A^2)$. We see therefore that within the present simplified setting the Hamiltonian can be written as

$$H_\alpha = R_{1\alpha} H_m R^\dagger_{1\alpha} + R_{0\alpha} H_{\mathrm{ph}} R^\dagger_{0\alpha}. \tag{103}$$

Equation (103) is an approximate special case of the more general and fundamental expression

$$H_\alpha = R_{\alpha'\alpha} \mathcal{H}_m(A_{\alpha'}) R^\dagger_{\alpha'\alpha} + R_{\alpha''\alpha} \mathcal{H}_{\mathrm{ph},\alpha''} R^\dagger_{\alpha''\alpha}, \tag{104}$$

which follows immediately from Eqs. (27) and (28). Equation (104) reduces to Eq. (103) when we choose $\alpha' = 1$ and $\alpha'' = 0$, and we make use of $\mathcal{H}_{\mathrm{ph},0} = H_{\mathrm{ph}}$ and $\mathcal{H}_m(A_1) = H_m$, which holds only because of the approximations and simplifying assumptions made. Note that without the latter the derivation of $\mathcal{H}_m(A_\alpha)$ via the unitary transformation of H_m is impossible.

B. Material truncation

We now consider truncating the material Hilbert space (De Bernardis, Jaako, and Rabl, 2018; De Bernardis *et al.*, 2018; Roth, Hassler, and DiVincenzo, 2019; Stefano *et al.*, 2019; Stokes and Nazir, 2019, 2020a; Settineri *et al.*, 2021). Since the canonical momentum p represents a different physical observable for each different value of α, the same is true of H_m. Therefore, projecting onto a finite number of eigenstates of H_m is a gauge-dependent procedure. Eigenvalues of H_m are denoted as ϵ_n. The projection P onto the first two levels $|\epsilon^0\rangle$ and $|\epsilon^1\rangle$ of H_m gives $PH_mP = \omega_m \sigma^+\sigma^- + \epsilon_0$ and $PqxP = d\sigma^x$, where $\sigma^+ = |\epsilon^1\rangle\langle\epsilon^0|$, $\sigma^- = |\epsilon^0\rangle\langle\epsilon^1|$, and $\sigma^x = \sigma^+ + \sigma^-$. The first transition energy is denoted as $\omega_m = \epsilon_1 - \epsilon_0$, and the transition dipole moment $d = \langle\epsilon^0|qx|\epsilon^1\rangle$ is assumed to be real. More generally, P may project onto any finite number of levels.

There are many ways to define two-level models. In general, a truncation of H_α is a P-dependent map $M_P : H_\alpha \to M_P(H_\alpha)$, such that $M_P(H_\alpha) : P\mathcal{H} \to P\mathcal{H}$ is an Hermitian operator on $P\mathcal{H}$ (Stokes and Nazir, 2020a). If, unlike the H_α's, the $M_P(H_\alpha)$'s are not equivalent for different α's, then truncation has broken the gauge invariance of the theory. To obtain what we refer to as the standard α-gauge two-level model, one replaces x and p with their projected counterparts PxP and PpP to obtain

$$M_P(H_\alpha) = H_\alpha^2 = PH_mP + PH_{\mathrm{ph}}P + V^\alpha(PxP, PpP), \tag{105}$$

where $V^\alpha(x,p) = H_\alpha - H_m - H_{\mathrm{ph}}$ is the interaction Hamiltonian. The terminology "standard" is used because this definition of M_P is capable of yielding the standard quantum Rabi model (QRM) that is ubiquitous in light-matter physics. Specifically, a standard QRM is obtained by choosing $\alpha = 1$ in Eq. (105). More generally, for distinct values of α the Hamiltonians H_α^2 are not equivalent to each other (De Bernardis *et al.*, 2018; Stokes and Nazir, 2019, 2021b), because P represents a different physical projection in each gauge.

Of importance when defining two-level models is the recognition that for an Hermitian operator O, projection $P \neq I$, and nonlinear function f we have

$$Pf(O)P \neq f(POP). \tag{106}$$

Thus, for a general material operator $O(x,p)$ we have $PO(x,p)P \neq O(PxP, PpP)$. This becomes an equality if and only if O is linear in x and p (Stokes and Nazir, 2019). As a result, various alternative truncating maps have been identified in the literature (De Bernardis, Jaako, and Rabl, 2018; De Bernardis *et al.*, 2018; Stefano *et al.*, 2019; Stokes and Nazir, 2019, 2020a; Taylor *et al.*, 2020; Settineri *et al.*, 2021).

Two further methods were proposed by Stefano *et al.* (2019); see also Taylor *et al.* (2020). Both methods require the EDA and involve replacing the unitary transformation $R_{\alpha\alpha'}$ in Eq. (103) with a two-level model counterpart. There are two different two-level model versions of $R_{\alpha\alpha'} : \mathcal{H} \to \mathcal{H}$, which are defined as

$$\mathcal{G}_{\alpha\alpha'} = PR_{\alpha\alpha'}P = P\exp[iq(\alpha-\alpha')xA]P, \tag{107}$$

$$\mathcal{T}_{\alpha\alpha'} = \exp[iq(\alpha-\alpha')PxPA] \neq \mathcal{G}_{\alpha\alpha'}, \tag{108}$$

where the final inequality holds because $e^{PxP} \neq Pe^{x}P$; cf. Eq. (106). Moreover, we cannot expect this inequality to become an approximate equality even for highly anharmonic material systems. An arbitrary operator O that is not necessarily diagonal in momentum space is defined by

$$[O\psi](p,A) = \int dp'dA'O(p,p',A,A')\psi(p',A'), \tag{109}$$

where ψ is the wave function of the composite system represented in momentum space for the matter subsystem and in position space (A space) for the photonic mode. It is straightforward to show that $R_{\alpha\alpha'}$ enacts a gauge transformation of the momentum arguments of O as

$$[R_{\alpha\alpha'}OR^{\dagger}_{\alpha\alpha'}\psi](p,A) = \int dp'dA'O(p-q[\alpha-\alpha']A, p' - q[\alpha-\alpha']A', A, A')\psi(p',A'). \tag{110}$$

We may write this more succinctly using the shorthand notation $R_{\alpha\alpha'}O(p)R^{\dagger}_{\alpha\alpha'} = O(p-q[\alpha-\alpha']A)$, in which it is to be understood that the gauge transformation applies to both momentum arguments of a generally nondiagonal operator. Since here both O and ψ are arbitrary, these results apply in particular to a projected operator $F = POP$ and a projected vector $P\psi$. Furthermore, since $\mathcal{G}_{\alpha\alpha'} = PR_{\alpha\alpha'}P$ and $P = P^2$, it follows again using shorthand notation that

$$\mathcal{G}_{\alpha\alpha'}F(p)\mathcal{G}^{\dagger}_{\alpha\alpha'} = PF(p-q[\alpha-\alpha']A)P. \tag{111}$$

Therefore, $\mathcal{G}_{\alpha\alpha'}$ implements a gauge transformation [as defined by Eqs. (96) and (97)] within a projected operator and then reprojects the result. By replacing $R_{\alpha\alpha'}$ in Eq. (103) [or Eq. (104)] with $\mathcal{G}_{\alpha\alpha'}$, one obtains the following new kind of two-level model:

$$\tilde{H}^2_{\alpha} = \mathcal{G}_{1\alpha}PH_mP\mathcal{G}^{\dagger}_{1\alpha} + \mathcal{G}_{0\alpha}PH_{\mathrm{ph}}P\mathcal{G}^{\dagger}_{0\alpha}. \tag{112}$$

These models are not equivalent for different α.

The other two-level model transformation $\mathcal{T}_{\alpha\alpha'}$, which is given in Eq. (108), is unitary (unlike $\mathcal{G}_{\alpha\alpha'}$), but it does not implement a gauge change [in the sense of Eqs. (96) and (97)], even when one considers a projected operator $POP = F$ in the sense previously described, that is,

$$\mathcal{T}_{\alpha\alpha'}F(p)\mathcal{T}_{\alpha\alpha'} \neq PF[p-(\alpha-\alpha')qA]P. \tag{113}$$

A two-level model unitary transformation cannot implement the minimal coupling replacement $p \to p - qA$, because the required operator algebra cannot be supported by the truncated space (Weyl, 1927). In general, the transformations $R_{\alpha\alpha'}$, $PR_{\alpha\alpha'}$, $\mathcal{G}_{\alpha\alpha'}$, and $\mathcal{T}_{\alpha\alpha'}$ trivially coincide in (and only in) the limit $P \to I$, which is the limit of no truncation.

By replacing $R_{\alpha\alpha'}$ in Eq. (103) [or Eq. (104)] with $\mathcal{T}_{\alpha\alpha'}$, one obtains the two-level models

$$h^2_1(\alpha) = \mathcal{T}_{1\alpha}PH_mP\mathcal{T}^{\dagger}_{1\alpha} + \mathcal{T}_{0\alpha}PH_{\mathrm{ph}}P\mathcal{T}^{\dagger}_{0\alpha} = \mathcal{T}_{1\alpha}H^2_1\mathcal{T}^{\dagger}_{1\alpha}, \tag{114}$$

where the second equality shows that these models are equivalent to the standard multipolar-gauge QRM H^2_1. In particular, $h^2_1(1) = H^2_1$. We note that the entire class $\{h^2_1(\alpha)\}$ results from truncation within the multipolar gauge (Stokes and Nazir, 2020a) (see also Sec. IV.C), so we refer to this class as a multipolar-gauge equivalence class. As discussed in Sec. IV.C, the transformations $\mathcal{T}_{\alpha\alpha'}$ refer to a phase-invariance principle defined entirely within a truncated space in terms of $x_P = PxP \neq x$.

Although it is clear that $\mathcal{T}_{\alpha\alpha'} \neq \mathcal{G}_{\alpha\alpha'}$, it is instructive to consider how the associated two-level models in Eqs. (112) and (114) differ. When the dimensionless coupling parameter is defined as $\eta = d/\sqrt{2\omega v}$ and $\bar{x} = \langle\epsilon^0|x|\epsilon^1\rangle = d/q$, if we assume that $PxQ \ll PxP$, where $Q = I - P$, and we neglect the terms PxQ and QxP in the exponent of R_{10}, we obtain

$$\begin{aligned}\mathcal{G}_{10} &\approx P\exp[i\eta(\sigma^x + QxQ/\bar{x})(a^{\dagger}+a)]P \\ &= P\exp[i\eta\sigma^x(a^{\dagger}+a)]P = \mathcal{T}_{10}.\end{aligned} \tag{115}$$

However, as already noted, such a naive approximation cannot be justified, even for a sufficiently anharmonic material system. To see this, note that by employing this approximation and then following exactly the same steps as previously detailed one obtains $\mathcal{T}_{\alpha\alpha'} \approx PR_{\alpha\alpha'}$. From this one obtains $H^2_0 \approx h^2_1(0)$, where the left-hand side is the standard Coulomb-gauge Rabi model and the right-hand side is equivalent to the standard multipolar-gauge Rabi model H^2_1. Since it is known that the spectra of H^2_0 and H^2_1 are markedly different (De Bernardis *et al.*, 2018; Stefano *et al.*, 2019; Stokes and Nazir, 2019), it follows that in general one cannot neglect terms PxQ and QxP in the exponent of $R_{\alpha\alpha'}$ even for highly anharmonic material systems. The multipolar-gauge models $h^2_1(\alpha)$ are indeed significantly different than $\tilde{H}^2_0$, exemplifying the importance of inequality (106) (Stokes and Nazir, 2020a).

The approximate equality $\mathcal{T}_{\alpha\alpha'} \approx \mathcal{G}_{\alpha\alpha'}$ does result if the exponentials on both sides are expanded to linear order in q. In this case the two-level models $\tilde{H}^2_{\alpha}$ are then the same as the models $h^2_1(\alpha)$ and they must be equivalent to each other for different α. However, a first-order expansion of the model $h^2_1(\alpha)$ simply gives back the standard two-level model H^2_{α} with quadratic terms neglected. It follows that in the weak-coupling regime all two-level models are the same $\tilde{H}^2_{\alpha} = h^2_1(\alpha) = H^2_{\alpha}$. This is the only regime in which such an equivalence can generally be obtained. As the coupling strength increases, the first-order expansion in q becomes progressively worse, so $\mathcal{T}_{\alpha\alpha'}$ and $\mathcal{G}_{\alpha\alpha'}$ become progressively different. Thus, if a gauge's truncation were found to be accurate for a particular observable in a particular situation, then as the coupling strength increases, truncation in any other gauge could be expected to become progressively less accurate by comparison. The relative optimality of different two-level models is discussed in Sec. IV.F.

C. Phase invariance with respect to truncated position

The Supplemental Material of Stefano *et al.* (2019) (note 1) provided an alternative derivation of the multipolar equivalence class $\{h_1^2(\alpha)\}$ via the imposition of a phase-invariance principle defined using the truncated operator $x_P \coloneqq PxP$. More generally, as shown by Stokes and Nazir (2020a), this principle can be applied in any gauge α, and it yields an equivalence class $\{h_\alpha^2(\alpha')\}$.

In the first quantized setting the gauge principle asserts that the mechanical energy $\mathcal{H}_m(A_\alpha)$ in Eq. (99) satisfies local phase invariance (gauge invariance),

$$\langle\psi|\mathcal{H}_m(A_\alpha)|\psi\rangle = \langle\psi'|\mathcal{H}_m(A'_\alpha)|\psi'\rangle, \tag{116}$$

where $|\psi'\rangle = e^{iq\chi}|\psi\rangle$ and $A'_\alpha = A_\alpha + \nabla\chi$. In particular, the equality $\langle\psi_\alpha|\mathcal{H}_m(A_\alpha)|\psi_\alpha\rangle = \langle\psi_{\alpha'}|\mathcal{H}_m(A_{\alpha'})|\psi_{\alpha'}\rangle$, in which $|\psi_{\alpha'}\rangle = R_{\alpha\alpha'}|\psi_\alpha\rangle$, expresses gauge invariance within the α-gauge framework and is a special case of Eq. (116) obtained by letting $\chi = \chi_{\alpha'} - \chi_\alpha$.

To define the class $\{h_1^2(\alpha)\}$, the gauge-fixing transformation $R_{1\alpha}$ was replaced by $\mathcal{T}_{1\alpha}$ in Eq. (102), and the multipolar-gauge mechanical energy $\mathcal{H}_m(A_1) = H_m$ was replaced by its projection $P\mathcal{H}_m(A_1)P$. More generally, however, Eqs. (102) and $\mathcal{H}_m(A_1) = H_m$ are special cases of Eqs. (101) and (99), respectively. If we replace $R_{\alpha\alpha'}$ with $\mathcal{T}_{\alpha\alpha'}$ and $\mathcal{H}_{m,\alpha}(A)$ with $\mathcal{H}_m^2(A_\alpha) \coloneqq P\mathcal{H}_m(A_\alpha)P$ on the right-hand side of Eq. (101), we obtain the following truncated α'-"gauge" mechanical energy:

$$\mathcal{H}_{m,\alpha}^2(A_{\alpha'}) \coloneqq \mathcal{T}_{\alpha\alpha'}\mathcal{H}_m^2(A_\alpha)\mathcal{T}_{\alpha\alpha'}^\dagger. \tag{117}$$

This truncated energy satisfies a form of phase invariance analogous to Eq. (116) but defined with respect to the truncated position operator $x_P \coloneqq PxP$. The phase transformation is defined by

$$U_{x_P} = e^{iq\chi(x_P)} = e^{i\beta}e^{id\Lambda\sigma^x}, \tag{118}$$

where β and Λ are constants depending on the choice of function χ. The global phase $e^{i\beta}$ can be ignored. Letting $|\psi_2\rangle = P|\psi\rangle$ denote an arbitrary truncated state, we have

$$\langle\psi_2|\mathcal{H}_{m,\alpha}^2(A_{\alpha'})|\psi_2\rangle = \langle\psi_2'|\mathcal{H}_{m,\alpha}^2(A'_{\alpha'})|\psi_2'\rangle, \tag{119}$$

where $A'_{\alpha'} = A_{\alpha'} + \partial_{x_P}\chi(x_P) = A_{\alpha'} + \Lambda$ and $|\psi_2'\rangle = U_{x_P}|\psi_2\rangle$. Thus, we see that $\mathcal{H}_{m,\alpha}^2(A_{\alpha'})$ is the mechanical energy of the α' gauge, where here the term gauge does not possess the same meaning as in the nontruncated theory but instead refers to x_P-phase invariance within the α-gauge truncated mechanical energy. Subsequently, a gauge transformation of $A_{\alpha'}$ under this principle is $A'_{\alpha'} = A_{\alpha'} + \Lambda$.

To obtain the complete α'-dependent Hamiltonian, one adds the transverse electromagnetic energy $\mathcal{H}_{\mathrm{ph},\alpha'}$, which is defined in Eq. (100), to the mechanical energy. This gives the total energy. Noting that $E_\mathrm{T} = -\Pi - \alpha' d\sigma^x/v = -\Pi - P_{\mathrm{T}\alpha'}$ is the transverse electric field after truncation, the truncated transverse electromagnetic energy $\mathcal{H}_{\mathrm{ph},\alpha'}^2$ may be defined as

$$\begin{aligned}\mathcal{H}_{\mathrm{ph},\alpha'}^2 &\coloneqq \frac{v}{2}\left[\left(\Pi + \frac{\alpha' d\sigma^x}{v}\right)^2 + \omega^2 A^2\right] \\ &= \mathcal{T}_{\alpha\alpha'}\mathcal{H}_{\mathrm{ph},\alpha}^2\mathcal{T}_{\alpha\alpha'}^\dagger = \mathcal{T}_{0\alpha'}H_\mathrm{ph}\mathcal{T}_{0\alpha'}^\dagger.\end{aligned} \tag{120}$$

The second equality in Eq. (120) follows because truncation does not alter the algebra of photonic operators, such that $\mathcal{T}_{\alpha\alpha'}$ transforms Π in the same manner as a gauge transformation. Equations (117) and (120) yield the full α'-dependent two-level model as the total energy

$$h_\alpha^2(\alpha') = \mathcal{H}_{m,\alpha}^2(A_{\alpha'}) + \mathcal{H}_{\mathrm{ph},\alpha'}^2 = \mathcal{T}_{\alpha\alpha'}H_\alpha^2\mathcal{T}_{\alpha\alpha'}^\dagger. \tag{121}$$

Thus, the equivalence class $\{h_\alpha^2(\alpha')\}$ can be obtained as the class of Hamiltonians satisfying x_P-phase invariance after truncation within the α gauge. The class $\{h_1^2(\alpha)\}$ derived by Stefano *et al.* (2019), Taylor *et al.* (2020), and Settineri *et al.* (2021) is the special case resulting from the application of the x_P-phase-invariance principle to the multipolar-gauge truncated theory. This has the appearance of an application to the free theory only due to approximations that have implied that $A_1 \equiv 0$ such that $p - qA_1 = p$, and therefore that $\mathcal{H}_m(A_\alpha) \equiv R_{1\alpha}\mathcal{H}_m(A_1)R_{1\alpha}^\dagger = R_{1\alpha}H_mR_{1\alpha}^\dagger$.

D. Relating models belonging to different equivalence classes

Further insight into the nature of the models $h_\alpha^2(\alpha')$ may be obtained by asking how any given standard two-level model must be modified in order that it coincides with the standard two-level model found using a different gauge. For example, we consider the term $q^2\mathbf{A}^2/2m$ of the Coulomb-gauge Hamiltonian. The coefficient $q^2/2m$ satisfies the Thomas-Reiche-Kuhn sum rule

$$\sum_n \omega_{nl}d_{nl}^i d_{ln}^j = i\frac{q^2}{2m}\langle\epsilon^l|[p_i, r_j]|\epsilon^l\rangle = \delta_{ij}\frac{q^2}{2m}. \tag{122}$$

Equation (122) rests directly on the CCR algebra, which as already noted can be supported only in an infinite-dimensional Hilbert space. Equation (122) is independent of the level l appearing on the left-hand side. However, if on the left-hand side we restrict ourselves to two levels $n, l = 0, 1$ with energy difference ω_m, then for $l = 1$ Eq. (122) reads

$$\sum_n \omega_{n1}d_{n1}^i d_{1n}^j = -\omega_m d_{10}^i d_{01}^j, \tag{123}$$

whereas for $l = 0$ Eq. (122) reads

$$\sum_n \omega_{n0}d_{n0}^i d_{0n}^j = +\omega_m d_{10}^i d_{01}^j. \tag{124}$$

The result obtained now depends on whether l is the ground or the excited state. As first noted by Stokes and Nazir (2018, 2019), if one takes the two-level projection of the Coulomb-gauge self-energy term, namely, $q^2\mathbf{A}^2(|\epsilon^0\rangle\langle\epsilon^0| + |\epsilon^1\rangle\langle\epsilon^1|)/2m$, and one applies Eqs. (123) and (124) to the excited-state projection $q^2|\epsilon^1\rangle\langle\epsilon^1|/2m$ and the ground-state projection $q^2|\epsilon^0\rangle\langle\epsilon^0|/2m$, respectively, then one arrives at the following

modified term, which now constitutes a nontrivial light-matter interaction:

$$\frac{q^2}{2m}\mathbf{A}^2 \leftrightarrow -\omega_m(\mathbf{d}\cdot\mathbf{A})^2\sigma^z, \tag{125}$$

where $\mathbf{d} \coloneqq \mathbf{d}_{10}$ and $\sigma^z = |\epsilon^1\rangle\langle\epsilon^1| - |\epsilon^0\rangle\langle\epsilon^0|$. As noted by Stefano *et al.* (2019) the modification (125) is *ad hoc*. It results in a model that no longer has the interaction of the Coulomb gauge. However, to second order in q the model obtained coincides with the multipolar-gauge model $h_1^2(0)$ (Stefano *et al.*, 2019). In this sense the truncated gauge principle can reveal what nonunitary modifications are required in order to relate nonequivalent truncated theories.

As already noted, to first order in q all two-level models are equivalent without any modification. To second order in q, forcing equivalence requires a nonunitary modification of at least one of the models involved. The modification (125) suffices to give the Coulomb-gauge model H_0^2 from the model $h_1^2(0)$ if and only if all higher-order terms in the expansion of $h_1^2(0)$ in powers of the coupling strength are neglected. This shows that as the coupling strength increases, increasingly drastic nonunitary modifications will be needed to transform a given model into one that belongs to a different equivalence class. This perspective is another way to understand the increasing difference with increasing coupling strength between the transformations $PR_{\alpha\alpha'}$ and $\mathcal{G}_{\alpha\alpha'}$ and the rotation $\mathcal{T}_{\alpha\alpha'}$.

E. Representing observables after truncation

It has been argued in the literature that the transformation $\mathcal{T}_{10}$ constitutes a two-level model gauge transformation, and that, since $\mathcal{T}_{10}$ is unitary, this resolves any gauge noninvariance due to truncation (Stefano *et al.*, 2019; Taylor *et al.*, 2020; Settineri *et al.*, 2021). However, the inequality (113) states that $\mathcal{T}_{1\alpha}$ does not generally implement a gauge change, as defined by Eqs. (96) and (97). The action of $\mathcal{T}_{\alpha\alpha'}$ coincides with that of the gauge transformation $R_{\alpha\alpha'}$ followed by projection P only when acting on operators that commute with $R_{\alpha\alpha'}$ (functions of x and A) and linear functions of Π, for which $PR_{\alpha\alpha'}\Pi R^\dagger_{\alpha\alpha'}P = \mathcal{T}_{\alpha\alpha'}\Pi\mathcal{T}^\dagger_{\alpha\alpha'}$. As first shown by Stokes and Nazir (2020a) (reviewed in Sec. IV.C), the invariance of the models related by $\mathcal{T}_{\alpha\alpha'}$ is x_P-phase invariance [as defined by Eq. (116)] rather than gauge invariance [as defined by Eq. (70)]. This is not merely a matter of semantics but an important mathematical distinction.

According to the general quantum postulates given in Sec. III.A for the identification of states and observables with vectors and operators, different gauges constitute different such associations within the starting theory (pretruncation). If we assume that in gauges α and α', the observable $\mathcal{O}$ is represented by operators o_α and $o_{\alpha'}$, and if we assume that after truncation $\mathcal{O}$ is represented by $M_P(o_\alpha)$ and $M_P(o_{\alpha'})$, then these truncated representations of $\mathcal{O}$ are not connected by a unitary operator in general (Fig. 5). A truncating map M_P does not preserve the algebra of material operators, and thus it cannot preserve the unitary relation between distinct associations of operators with observables (gauges) made within the starting theory.

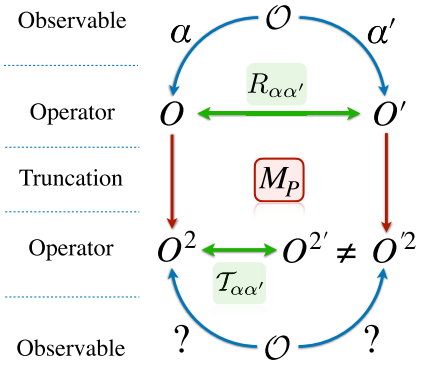

FIG. 5. Breakdown of gauge invariance under a truncating map M_P. The equivalent representations O and O' of the same observable $\mathcal{O}$ are not equivalent after the application of M_P. A two-level unitary such as $\mathcal{T}_{\alpha\alpha'}$ cannot produce from the truncated α-gauge theory the same observable $\leftrightarrow$ operator association as defined by a distinct gauge α'. The "correct" association $\mathcal{O} \leftrightarrow O^2$ after truncation can be defined only by identifying a gauge in which the truncation O^2 is accurate. Subsequently, any two-level unitary operator can be used to define an equivalent truncated representation.

The word or words used to label the freedom to choose among unitarily equivalent representations of an observable within quantum theory is immaterial. In particular, the label gauge freedom has been used for this purpose within truncated theories (Stefano *et al.*, 2019; Settineri *et al.*, 2021). Specifically, within a starting theory the different representations of observable $\mathcal{O}$ that compose the equivalence class

$$C(\mathcal{O}) = \{ROR^\dagger : R \text{ unitary}\} \tag{126}$$

can be referred to as different gauges, and similarly, in a truncated theory subsequently obtained using a map M_P, the truncated representations of $\mathcal{O}$ belonging to

$$C^2(M_P,\mathcal{O}) = \{U^2O^2(U^2)^\dagger : O^2 = M_P(O) \text{ and } U^2 \text{ unitary}\} \tag{127}$$

could also be referred to as different gauges. However, given a rotation U^2 within the truncated space, in general we have

$$U^2M_P(O)(U^2)^\dagger \neq M_P(ROR^\dagger) \tag{128}$$

for any rotation R. In other words,

$$M_P(C(\mathcal{O})) \neq C^2(M_P,\mathcal{O}). \tag{129}$$

This proves that identifying the equivalences that occur within the truncated and nontruncated theories would be erroneous. Specifically, one must not surreptitiously and incorrectly equate the left- and right-hand sides of inequality (129) simply because one has chosen to refer to both the elements of $C(\mathcal{O})$ and the elements of $C^2(M_P,\mathcal{O})$ using the same label (gauges).

The definition of the class $C^2(M_P,\mathcal{O})$ relies upon an accurate truncation $O^2 = M_P(O)$ having first been found; that is, $C^2(M_P,\mathcal{O})$ cannot be defined until a map M_P has first been applied to one of the elements of $C(\mathcal{O})$ to give O^2. Yet, applying M_P to different elements of $C(\mathcal{O})$ will give different

(nonequivalent) operators O^2; that is, the left-hand side of inequality (129) is not a unitary equivalence class. Thus, given a map M_P, every different (but equivalent) element of $C(\mathcal{O})$ defines a different equivalence class $C^2(M_P,\mathcal{O})$. These different equivalence classes are not equivalent, which constitutes gauge noninvariance. The fact that each $C^2(M_P,\mathcal{O})$ is an equivalence class constitutes x_P-phase invariance. Within $C^2(M_P,\mathcal{O})$ any two elements are connected by an x_P-phase transformation. Thus, gauge noninvariance and x_P-phase invariance are necessarily simultaneously satisfied by truncated models. It follows that the two invariances cannot coincide and exhibiting one of these invariances cannot resolve a breakdown of the other. We note that, although we have focused on two-level truncations, the preceding general analysis holds for any $P \neq I$.

In summary, the possibility of applying unitary rotations after truncation does not eliminate the problem of first determining a gauge and a map M_P that combined provide an accurate representation O^2 of the observable of interest $\mathcal{O}$. This problem arises because a truncating map M_P breaks gauge invariance.

F. Optimality of truncations

We now discuss which two-level models are known to be accurate in which situations. Subsequently, we discuss the importance of two-level model predictions for gauge ambiguities. Material truncation should be expected to offer a robust approximation when the material system is sufficiently anharmonic that the orthogonal subspace $Q\mathcal{H}$ is well separated from $P\mathcal{H}$, where $P\mathcal{H} \oplus Q\mathcal{H} = \mathcal{H}$ is the full Hilbert space. Such regimes may or may not be of experimental importance when one considers specific implementations of light-matter physics models.

We first suppose we have a highly anharmonic system at arbitrary coupling strength and only a single radiation mode. The Coulomb-gauge coupling involves the canonical momentum $\mathbf{p}$, which possesses matrix elements in the material basis $\{|\epsilon^n\rangle\}$ that scale with material transition frequencies as

$$q\mathbf{p}_{nl} = im\omega_{nl}\mathbf{d}_{nl}. \tag{130}$$

As first explained by De Bernardis *et al.* (2018), transitions to higher states are not suppressed within the Coulomb gauge, because the increasing energy gap is compensated for by an increasing coupling matrix element. In contrast, the multipolar coupling involves only the dipole moment. Therefore, for sufficiently strong coupling where two-level models are not equivalent, the Coulomb-gauge truncation will generally perform poorly in comparison to the multipolar-gauge truncation as a general approximation of the nontruncated theory. These points were also elaborated on by Stokes and Nazir (2019) via a Schrieffer-Wolff-type analysis. As an illustrative example, we take a double-well dipole with potential $V(\theta,\phi) = -\theta r^2/2 + \phi r^4/4$, where θ and ϕ control the shape of the double well (De Bernardis *et al.*, 2018; Stefano *et al.*, 2019; Stokes and Nazir, 2020b). The material Hamiltonian is therefore (De Bernardis *et al.*, 2018)

$$H_m^\alpha = \frac{\mathcal{E}}{2}\left(-\partial_\zeta^2 - \beta\zeta^2 + \frac{\zeta^4}{2}\right), \tag{131}$$

where we have defined the dimensionless variable $\zeta = r/r_0$ with $r_0 = (1/[m\phi])^{1/6}$, along with $\mathcal{E} = 1/mr_0^2$ and $\beta = \theta m r_0^4$. We first consider the case of resonance $\delta = \omega/\omega_m = 1$ together with a high anharmonicity $\mu = (\omega_{21} - \omega_m)/\omega_m$ of $\mu = 70$. We compare the unique spectrum of the nontruncated Hamiltonian H_α to the different approximations given by the QRMs H_1^2 and H_0^2, as well as with the nonstandard Coulomb-gauge model $\tilde{H}_0^2$ defined by Eq. (112). We note that for each α the standard two-level model H_α^2 can be selected as the representative of its unitary equivalence class $\{h_\alpha^2(\alpha')\}$ without loss of generality. As shown in Fig. 6, the multipolar-gauge QRM H_1^2 is accurate for predicting transition spectra in this regime while the Coulomb-gauge models H_0^2 and $\tilde{H}_0^2$ are qualitatively similar and inaccurate for strong enough couplings. There are a number of factors determining the optimality of a truncation.

For example, when the detuning $\delta = \omega/\omega$ is large (small) the Coulomb-gauge two-level model coupling $\eta' = (\omega_m/\omega)d/\sqrt{2\omega v}$ is weaker (stronger) than the corresponding multipolar-gauge coupling $\eta = d/\sqrt{2\omega v} = \delta\eta'$. The two-level model Hamiltonian $PH_\alpha P$ constitutes the first-order (in V^α) contribution to a more general effective Hamiltonian defined over the two-level subspace $P\mathcal{H}$ (Wilson and Hubac, 2010). If the model $PH_\alpha P$ is found to be inaccurate, then higher-order corrections can be calculated perturbatively using various forms of perturbation theory (Wilson and Hubac, 2010). In particular, the second-order contribution is straightforwardly obtainable for a two-level system and single mode and should yield a two-level model with improved accuracy. In a single-mode theory, such higher-order contributions will tend to be larger toward the Coulomb-gauge value $\alpha = 0$ because as noted the energy gap to the orthogonal subspace $Q\mathcal{H}$ is compensated for by the form of the Coulomb-gauge coupling.

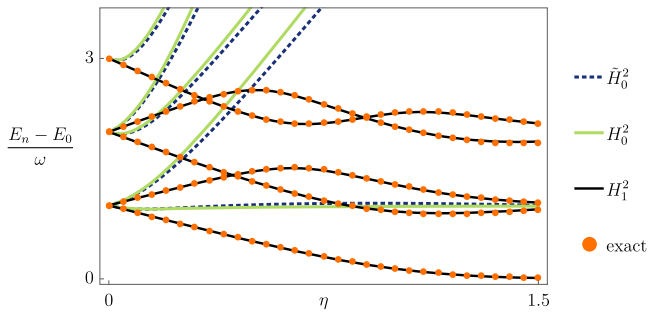

FIG. 6. Transition spectra (relative to the ground energy G) of two-level models are compared to the exact transition spectrum (points), assuming a material anharmonicity of $\mu := (\omega_{21} - \omega_m)/\omega_m = 70$ and a resonance $\delta := \omega/\omega_m = 1$. The multipolar-gauge QRM (black curves) is generally accurate in this regime, in the sense that one must go to high energy levels before discrepancies with the exact spectrum are found. The two Coulomb-gauge two-level models H_0^2 (lighter curves) and $\tilde{H}_0^2$ (dashed curves) are generally inaccurate and are qualitatively similar.

When more radiation modes are considered, the optimal gauge may often be shifted away from the multipolar gauge toward the Coulomb gauge (Roth, Hassler, and DiVincenzo, 2019). The multipolar and Coulomb-gauge linear interactions scale as $\sqrt{\omega}$ and $1/\sqrt{\omega}$, respectively. The introduction of more radiation modes causes the multipolar-gauge truncation to become suboptimal because the effects of nonresonant modes are more pronounced in this gauge, as further discussed in Sec. VI.A. Results illustrating this effect within the strong-coupling regime were given by Roth, Hassler, and DiVincenzo (2019). When more dipoles are considered but only a single radiation mode is retained, the multipolar-gauge truncation is again typically optimal at sufficiently large anharmonicity, and accuracy increases with the number of dipoles considered (Stokes and Nazir, 2020b).

Rouse *et al.* (2021) addressed the issue of gauge noninvariance due to truncation using a novel description in terms of dual coordinates. This is reviewed in note X of the Supplemental Material (245). It was found that approximations within the multipolar gauge ($\alpha = 1$) will typically most accurately represent the physics of small, bound dipoles interacting with a single mode. A wide range of system types are considered along with the effects of both material truncation and the EDA.

Ashida, İmamoğlu, and Demler (2021) identified a Pauli-Fierz-type representation obtained from the Coulomb gauge by unitary transformation. For a one-dimensional material system coupled to a single cavity mode with frequency ω, the transformation is defined by $U = e^{-i\xi_g p\pi}$. Here $\pi = i(c^\dagger - c)$, with c a renormalized cavity annihilation operator for a photon with frequency $\tilde{\omega}$, where $\tilde{\omega} = \sqrt{\omega^2 + g^2}$ and $x_{\tilde{\omega}} g$ is a bare coupling strength defined using the Coulomb-gauge Hamiltonian with $x_{\tilde{\omega}} = 1/\sqrt{m\tilde{\omega}}$. The renormalized coupling $\xi_g := g x_{\tilde{\omega}}/\tilde{\omega}$ is a nonconstant function of the bare coupling parameter g with a maximum value close to $g = 1$.

The idea of the Pauli-Fierz representation is to eliminate the component of the transverse field tied to material charges (Cohen-Tannoudji, Dupont-Roc, and Grynberg, 2010). The Hamiltonian within the transformed frame is $H^U = H_m(p, r + \xi_g \pi) + \tilde{\omega} c^\dagger c$, where $H_m(p, r) := p^2/2m_{\text{eff}} + V(r)$ and the effective mass is defined by $m_{\text{eff}} = m[1 + 2(g/\omega)^2]$. For an increasing g the renormalized frequency $\tilde{\omega}$ is increasingly dominant, while the coupling ξ_g eventually begins to decrease. For sufficiently large g the eigenvectors of H^U become approximately separable despite remaining highly entangled in the Coulomb gauge.

If V has local minima near which it can be expanded as $\delta V \propto r^2$, then since m_{eff} increases quadratically with g the eigenfunctions of $H_m(p, r)$ are increasingly localized around the potential minima and the low-lying spectrum of H^U is that of a harmonic oscillator with narrowing level spacing $\delta E \propto 1/g$. Ashida, İmamoğlu, and Demler (2021) further argued that truncation is increasingly well justified within H^U at larger g due to increased localization of the eigenstates of $H_m(p, r)$ that results from the dependence on m_{eff}. Ashida, İmamoğlu, and Demler (2021) studied a double-well dipole as an application example. They found that for a shallow double well even the multipolar-gauge truncation fails quite severely at extreme-coupling strengths, and even in the case of only a single mode, whereas truncation in the Pauli-Fierz frame remains accurate. Ashida, İmamoğlu, and Demler (2021) also provided a multimode generalization of their Pauli-Fierz-type transformation.

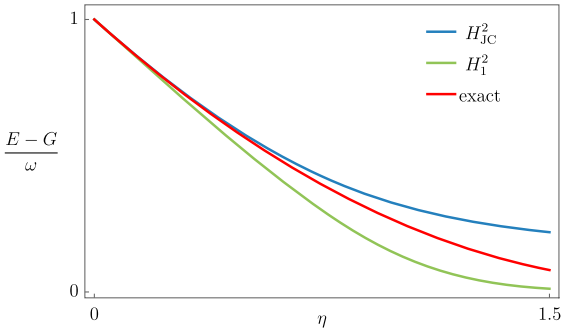

FIG. 7. First transition energies of the two-level models H_1^2 (lower curve) and H_{JC}^2 (upper curve) compared to the exact transition energy (middle curve), assuming a material anharmonicity of $\mu \approx 3$ and a resonance $\delta = 1$. The α_{JC}-gauge two-level model can be more accurate than the multipolar-gauge QRM in the ultrastrong-coupling regime.

The relative accuracy of material truncations performed in different regimes and gauges is now well understood, at least for simple light-matter systems. In particular, truncation will break down as a general approximation for sufficiently harmonic material systems. However, in simple models the accuracy of a given truncation is of limited importance because the truncation is straightforwardly avoidable.

Truncation is most significant in its capacity to reveal important qualitative physical implications. In particular, the onset of ultrastrong coupling (USC) has often been identified through a departure from Jaynes-Cummings physics due to the breakdown of the rotating-wave approximation (RWA). In the USC regime the qualitative low-energy physics of the Jaynes-Cummings model (JCM) is markedly different than that of the QRM. For example, the JCM predicts no ground-state entanglement and no ground-state photon population for all coupling strengths. The contrary predictions of the QRM have previously been regarded as defining ultrastrong-coupling phenomenology. However, Stokes and Nazir (2019) showed that there is a gauge choice that yields a Jaynes-Cummings model without performing the RWA. The corresponding gauge parameter α_{JC} varies with the coupling and detuning parameters of the theory, but this is certainly permissible. It simply amounts to choosing a nonconstant gauge function; see Sec. II.D.

For a material harmonic oscillator two-level truncation is essentially as poor a general approximation as it can ever be, yet for this system the ground state of the truncated model is exact in the JC gauge ($P|G_{\text{JC}}\rangle = |G_{\text{JC}}\rangle$); see Sec. V.C.1. As a result, there are gauges $\alpha \neq 1$ in which the two-level truncation of material systems with low anharmonicity remains accurate for low-energy states, despite truncation in any gauge generally breaking down for higher levels. Stokes and Nazir (2019) exemplified an experimentally realistic regime of a fluxonium LC-oscillator system with anharmonicity $\mu \approx 3.15$, such that two-level models remain accurate for predictions up to the first excited state and for which the JC-gauge two-level

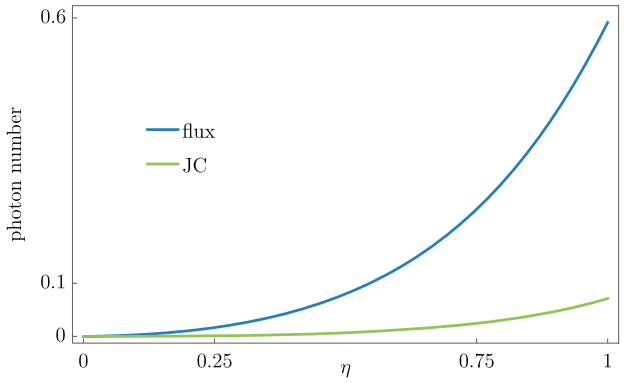

FIG. 8. The exact ground-state average numbers of flux-gauge (upper curve) and JC-gauge (lower curve) photons with coupling strength η for a fluxonium system assuming an anharmonicity of $\mu \approx 3$ and a resonance of $\delta = 1$. The number of JC-gauge photons is much lower than the number of flux-gauge photons. An appreciable JC-gauge photon population occurs only for large couplings approaching the deep-strong limit $\eta = 1$.

model is usually more accurate. It follows that low-energy weak-coupling phenomenology can persist even within the USC regime, such that the phenomenology previously viewed as definitive in the USC regime need not hold even within gauge-invariant nontruncated models. Essentially the same findings are obtained for a double-well dipole, as illustrated in Fig. 7.

We now turn our attention to photon number observables. The dipole-cavity Hamiltonian in Eq. (98) possesses a cavity self-energy term $q^2A_\alpha^2/2m$ that vanishes only for $\alpha = 1$ ($A_1 = 0$). Since it acts nontrivially only within the photonic Hilbert space, this term is unaffected by material truncation. It can be absorbed into the cavity Hamiltonian using a local Bogoliubov transformation. Thus, each gauge $\alpha \neq 1$ possesses two possible definitions of photon number that do and do not include this renormalization, respectively. In the JC gauge the renormalized photon number predicted by the JC-gauge two-level model is identically zero in the ground state because, in terms of the corresponding photonic operators, the JC-gauge

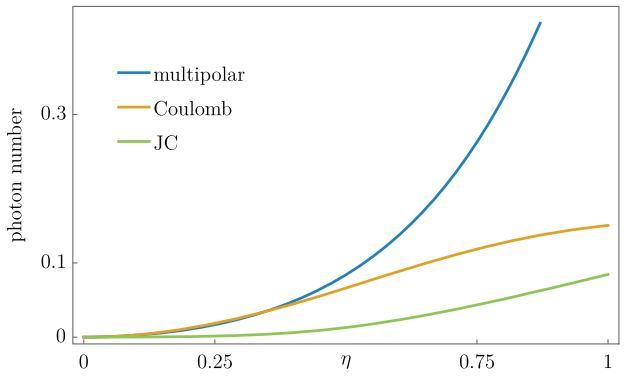

FIG. 9. The exact ground-state average numbers of multipolar-gauge (upper curve), Coulomb-gauge (middle curve), and JC-gauge (lower curve) photons for a double-well dipolar system assuming an anharmonicity of $\mu \approx 70$ and a resonance of $\delta = 1$. The number of JC-gauge photons is appreciable only well into the USC regime $\eta > 1/2$.

two-level model takes the Jaynes-Cummings form. On the other hand, the ground-state average of the bare JC-gauge photon number (which does not include the A_{JC}^2 term) can possess nonzero values for sufficiently large coupling strengths, even when the average is found using the JC-gauge truncated model. Moreover, when the two-level truncation is avoided, both renormalized and nonrenormalized (bare) JC-gauge photon numbers can be nonzero in the ground state due to counter-rotating terms connecting to dipole levels above the first.

Figures 8 and 9 show the exact, i.e., nontruncated, ground-state bare photon numbers defined relative to the multipolar (flux), Coulomb (charge), and JC gauges for fluxonium and double-well dipole systems, respectively. In the cases of the Coulomb and JC gauges these photon numbers do not include in their definitions the A_α^2-type terms. In particular, for sufficiently large coupling strengths, the predicted JC-gauge photon number average is nonzero due to both the A_{JC}^2 term and counterrotating terms to higher dipolar levels. To illustrate different regimes of anharmonicity, we have assumed that $\mu \approx 3$ for the fluxonium system and $\mu \approx 70$ for the double-well dipole. In both cases the ground-state $\mathrm{photon}_{\mathrm{JC}}$ population is highly suppressed when compared to the ground-state photon_0 and photon_1 populations. All of these predictions are gauge invariant, having been obtained from the nontruncated theory.

V. TIME-DEPENDENT INTERACTIONS AND ADIABATIC SWITCHING

Time-dependent interactions arise in a number of contexts in light-matter physics. Here the notion of a process in which material charges exchange photons is elementary. The concept arises from scattering theory wherein the interaction $V = H - h$, where h is called the unperturbed Hamiltonian, is adiabatically switched on and off over an infinite duration. Such an idealization may not, however, be applicable in extreme light-matter interaction regimes. Gauge freedom in scattering theory has been discussed extensively in the context of atomic line shape and level-shift phenomena (Lamb, 1952; Low, 1952; Power and Zienau, 1959; Fried, 1973; Bassani, Forney, and Quattropani, 1977; Kobe, 1978; Cohen-Tannoudji, Dupont-Roc, and Grynberg, 1989; Baxter, Babiker, and Loudon, 1990; Woolley, 1998, 2000; Stokes, 2013). We explain why subsystem gauge relativity can be ignored in calculating the S matrix (Cohen-Tannoudji, Dupont-Roc, and Grynberg, 1989). We then directly demonstrate that conventional quantum optical approximations mimic the S matrix, and thereby eliminate subsystem gauge relativity. Only within such approximations do atoms and photons defined as quantum subsystems become ostensibly unique concepts for a given definition of h. It should also be noted, however, that different definitions of h are available and might be considered. We also discuss nonadiabatic switching of ultrastrong couplings whereby subsystem gauge relativity becomes important generally.

A. Adiabatic switching and a unique invariance property of the S matrix

As explained in Secs. III.B–III.D, the task we are faced with is the determination of which gauge-invariant subsystem

definitions are relevant in any given situation. However, if the S matrix is applicable in providing all physical predictions, we are able to completely ignore this question. The subsystems become ostensibly unique within scattering theory because of the adiabatic interaction-switching condition therein. Feynman diagrams can be used as a mnemonic when calculating the terms in a perturbative expansion of the Hamiltonian resolvent used to define the S matrix, which is the primary source of predictions in particle physics. This gives rise to the notions of "real" and "virtual" processes.

The α-gauge Hamiltonian can be partitioned as $H_\alpha = h + V^\alpha$, where $h = H_m + H_{\mathrm{ph}}$ is the unperturbed Hamiltonian and V^α is the interaction. The unperturbed energy eigenvalues and eigenvectors are defined by $h|\epsilon^n\rangle = \epsilon^n|\epsilon^n\rangle$. The vectors $\{|\epsilon^n\rangle\}$ are each a tensor product of an eigenvector of H_m and an eigenvector of H_{ph} (photon number state). Suppose that physical states $\mathcal{S}_n$ and $\mathcal{S}_m$ are represented in gauge α by vectors $|\epsilon^n\rangle$ and $|\epsilon^m\rangle$. The same states are represented in gauge α' by vectors $|\epsilon'^n\rangle = R_{\alpha\alpha'}|\epsilon^n\rangle$ and $|\epsilon'^m\rangle = R_{\alpha\alpha'}|\epsilon^m\rangle$, respectively; therefore, the bare eigenvectors of h represent different physical states in each gauge (subsystems are gauge relative). The evolution operator generated by H_α between times t_i and t_f is denoted $U_\alpha(t_i, t_f)$. Evolutions in different gauges are related by $U_{\alpha'}(t_i, t_f) = R_{\alpha\alpha'}U_\alpha(t_i, t_f)R^\dagger_{\alpha\alpha'}$. The probability amplitude $A(\mathcal{S}_n, t_f; \mathcal{S}_m, t_i)$ to find the system at time t_f in state $\mathcal{S}_n$ given that at time t_i its state was $\mathcal{S}_m$ is given by the corresponding evolution operator matrix element and is the following gauge-invariant prediction:

$$A(\mathcal{S}_n, t_f; \mathcal{S}_m, t_i) = \langle\epsilon^n|U_\alpha(t_i, t_f)|\epsilon^m\rangle = \langle\epsilon'^n|U_{\alpha'}(t_i, t_f)|\epsilon'^m\rangle \quad \text{(gauge invariance)}. \tag{132}$$

It is equally clear that for $\alpha \neq \alpha'$ we have

$$\langle\epsilon^n|U_\alpha(t_i, t_f)|\epsilon^m\rangle \neq \langle\epsilon^n|U_{\alpha'}(t_i, t_f)|\epsilon^m\rangle \quad \text{(gauge relativity)}. \tag{133}$$

Inequality (133) simply exemplifies the expected result that an eigenvector of h represents a different physical state in each different gauge. The left-hand side is α dependent, while the right-hand side is α' dependent, but both predictions are gauge-invariant amplitudes of the form specified by Eq. (132). We refer to this α dependence despite the gauge-invariance of both predictions as gauge relativity.

In scattering theory it is assumed that $V^\alpha = 0$ in the remote past and distant future $t = \pm\infty$, such that at these times $H = h$, so the unperturbed energy eigenvectors uniquely represent the total energy eigenstates. It is then assumed that the interaction is switched on and off adiabatically between $t = \pm\infty$. Subsequently, the S matrix is formally defined by (Cohen-Tannoudji, Dupont-Roc, and Grynberg, 1989; Weinberg, 2005)

$$S_{nm} = \lim_{t\to\infty}\langle\epsilon^n|U_{\alpha I}(-t, t)|\epsilon^m\rangle, \tag{134}$$

where $U_{\alpha I}$ denotes the corresponding evolution operator in the interaction picture defined by h. In contrast to inequality (133), the S matrix possesses the noteworthy property that it is independent of α despite being defined in terms of the same unperturbed vectors for every α. In other words, a special property of the S matrix is that it is *gauge nonrelative*. In calculating S_{nm}, we do not have to transform the eigenvectors of h in order to ensure that we are using the same physical states in each gauge, as in Eq. (132) (Cohen-Tannoudji, Dupont-Roc, and Grynberg, 1989). "Photonic" and "material" excitations represented by the eigenstates of h become (at least ostensibly) unique in scattering theory, so we do not have to confront the question of which subsystem definitions are the most relevant.

A general proof of this unique invariance property of the S matrix was given for nonrelativistic QED by Woolley (1998, 2000). Essential for the proof is that the unperturbed operator h is kept the same in each gauge. The S matrix can also be expressed in the following form (Woolley, 1998, 2000; Cohen-Tannoudji, Dupont-Roc, and Grynberg, 2010):

$$S_{nm} = \delta_{nm} - 2\pi i T_{nm}\delta(\epsilon_n - \epsilon_m), \tag{135}$$

where T is called the transition matrix, whose elements in the unperturbed basis naturally depend on α. However, when it is evaluated on energy shell as expressed by the delta function in the S-matrix element S_{nm}, all α dependence drops out (Woolley, 1998, 2000). This bare-energy conservation property is thereby seen as crucial in ensuring that the gauge relativity of the subsystems can be ignored when one calculates the S matrix.

We can define any process that conserves h as real. A virtual process is then one that is not real. In the S matrix, the latter can occur only as intermediate processes constituting part of a real process. More generally, however, the S matrix can be understood as an infinite-time limit of the more general matrix given by (Cohen-Tannoudji, Dupont-Roc, and Grynberg, 2010)

$$S^{(\tau)}_{nm} = \delta_{nm} - 2\pi i T_{nm}\delta^{(\tau)}(\epsilon_n - \epsilon_m). \tag{136}$$

The function $\delta^{(\tau)}(\epsilon_n - \epsilon_m)$ has a peak at $\epsilon_n = \epsilon_m$ with a width of the order of $1/\tau$, which is often taken as expressing the conservation of bare energy to within $1/\tau$ (Cohen-Tannoudji, Dupont-Roc, and Grynberg, 2010). This is the heuristic energy-time uncertainty relation, which it should be noted is significantly different than the rigorous Heisenberg uncertainty relation for conjugate operators.

The processes described by the matrix $S^{(\tau)}$ are not purely real (zero energy uncertainty) unless $\tau \to \infty$. It is widely regarded that physical processes are real. However, although the total energy E represented by the operator H_α is automatically conserved, there is nothing in quantum or classical theory that requires a physical process to conserve only part of this energy, such as the part represented by h. This is required and occurs in the S matrix only because $H_\alpha = h$ at the beginning and the end of a scattering process. Yet, the limit of infinite times with adiabatic switching is an idealization, such that purely real processes cannot truly occur. In this sense the

term real is a misnomer. Further still, only when a process is real (i.e., it is a scattering process) can the gauge relativity of the subsystems necessarily be ignored. In other words, scattering theory is gauge nonrelativistic.

All predictions are fundamentally gauge invariant in the sense of Eq. (132). Thus, both sides of inequality (133) are gauge-invariant predictions, but beyond scattering theory, i.e., over finite times, we must recognize that they are different gauge-invariant predictions. We are then confronted with the task of determining which (if either) is more relevant.

B. Partitioning the Hamiltonian

Although the S matrix is gauge nonrelative in the previously defined sense, it can be defined only relative to a partition of the Hamiltonian into unperturbed and interacting parts as $H = h + V$, which is nonunique. Naively, one might attempt to define V and h as the components that, respectively, do and do not depend on a "coupling" parameter, of which the only obvious choice is the electric charge q. According to this definition, h would consist of the free photonic Hamiltonian $H_{\mathrm{ph}} = \int d^3k \sum_\lambda \omega[a_\lambda^\dagger(\mathbf{k})a_\lambda(\mathbf{k}) + 1/2]$ together with particle energies $H_m = \sum_{\mathrm{charges}} \mathbf{p}^2/(2m)$. The unperturbed vectors would therefore be incapable of representing bound material states. This definition would be of little use in applications of QED at low energies whereby a separation of near-field interactions is advantageous in allowing bound charge systems to emerge as the constituents of "unperturbed" stable matter. In particular, the most commonly used definition of h in nonrelativistic QED, namely, the definition suggested by the Coulomb gauge that reads

$$h = \sum_{\mathrm{charges}} \frac{\mathbf{p}^2}{2m} + V_{\mathrm{Coul}} + H_{\mathrm{ph}}, \tag{137}$$

where $V_{\mathrm{Coul}} = \int d^3x \mathbf{E}_{\mathrm{L}}^2/2$ is the Coulomb energy, would be ruled out because V_{Coul} depends on q.

A different criterion to define V would be that it must not include any terms that act exclusively within the matter Hilbert space or exclusively within the photonic Hilbert space. In particular, V must not include any "self-interaction" terms, which although dependent on q are of the form $O_m \otimes I_{\mathrm{ph}}$ or $I_m \otimes O_{\mathrm{ph}}$. In the Coulomb gauge this criterion does indeed lead to the commonly used definition of h given in Eq. (137), and therefore to the familiar Coulomb-gauge interaction Hamiltonian of the form $-q\mathbf{p}\cdot\mathbf{A}_{\mathrm{T}}(\mathbf{r})/m + q^2\mathbf{A}_{\mathrm{T}}(\mathbf{r})^2/2m$ for each charge q.

However, this method does not in general yield the same definition of h when it is applied in other gauges. In the gauge g, the material Hamiltonian H_m would include the total polarization energy $\int d^3x \mathbf{P}_g^2/2$, which in addition to V_{Coul} includes the transverse polarization "self-term" $\int d^3x \mathbf{P}_{g\mathrm{T}}^2/2$. In the multipolar gauge this additional term is divergent. It cannot contribute to processes in which the number of photons change, and otherwise it is often ignored until such a point that its contributions can be "renormalized out" of final predictions. This is how on-energy-shell T-matrix elements for bound-state level shifts are typically calculated using the multipolar gauge (Craig and Thirunamachandran, 1998). Predictions obtained in this way are identical to those found using the Coulomb gauge because they result from having employed the same definition of h.

If the multipolar transverse polarization is instead regularized, as described in Secs. II.D.2 and II.E, then $\int d^3x \mathbf{P}_{g\mathrm{T}}^2/2$ is finite but its relative magnitude depends on the cutoff k_M. It can be considered a weak perturbation of V_{Coul} provided that k_M is chosen appropriately (Vukics, Grießer, and Domokos, 2015). In this case, and more generally whenever H_m includes terms other than V_{Coul}, a different S matrix is obtained than when h is used in Eq. (137).

One might also consider relative magnitudes to be a guide in determining appropriate definitions of h and V. In order for weak-coupling methods to be applicable, the interaction V must be a weak perturbation of h. For example, when considering multiple systems of interest within a common reservoir, if direct intersystem interactions are sufficiently strong, then they should be included within h rather than within the system-reservoir interaction V (Santos and Semião, 2014; Stokes and Nazir, 2018). Subsequently applying weak-coupling theory yields a reduced description in the form of a Lindblad master equation whose coefficients are S-matrix elements. An example of this is given in Sec. V.C.2, but it should be noted that the S matrix obtained is specific to whatever definition of h is adopted. Similarly, when one deals with strong system-reservoir couplings analytic methods such as polaron transformations (Pollock *et al.*, 2013; Nazir and McCutcheon, 2016) and Hamiltonian mapping techniques (Iles-Smith, Lambert, and Nazir, 2014; Strasberg *et al.*, 2016) work by redrawing the system-reservoir boundary so as to obtain a weak perturbation V.

Physically, when subsystem interactions are strong it is unclear to what extent the subsystems should be considered operationally accessible. A given experiment may (or may not) be capable of granting access to only a dressed composite rather than the individual subsystems that compose it. The balance between localization and dressing within the context of nonrelativistic QED is discussed throughout Sec. VI. In the context of open quantum systems theory, this topic is closely related to the distinction between local and global approaches to deriving reduced descriptions, which is discussed in Sec. VII.C.

In conclusion, we note that while the S matrix is gauge nonrelative in the sense defined in Sec. V.A, this property does not necessarily circumvent the challenge of determining a gauge relative to which one is to obtain physical predictions, even within scattering theory itself. Indeed, the myriad existing scattering-theoretic predictions of nonrelativistic QED found using low-order perturbation theory rely on the specific definition of h given in Eq. (137). The prospect of deriving alternative QED S matrices that result from different definitions of h, for example, those that include a "weak" self-term $\int d^3x \mathbf{P}_{g\mathrm{T}}^2/2$, warrants further study.

C. Quantum optical approximations: Mimicking the S matrix

We now directly show that subsystem gauge relativity can be eliminated after a sufficient number of weak-coupling approximations are performed.

1. Toy model: Material oscillator and a single mode

We begin by again considering a simple toy model consisting of a material harmonic oscillator and a single radiation mode, such that Eqs. (14) and (9) become

$$P_{\mathrm{T}\alpha} = \frac{\alpha q x}{v}, \tag{138}$$

$$A_\alpha = (1-\alpha)A, \tag{139}$$

where v is the cavity volume. The cavity canonical operators are $A = (a^\dagger + a)/\sqrt{2\omega v}$ and $\Pi = i\sqrt{\omega/2v}(a^\dagger - a)$, such that $[A,\Pi] = i/v$. We assume that the material oscillator points in the same direction as the mode. The theory is gauge invariant because the gauge-fixing transformations remain unitary ($R_{\alpha\alpha'} = e^{iq(\alpha-\alpha')rA}$).

The α-gauge Hamiltonian in Eq. (22) can be written as $H^\alpha = h + V^\alpha$, where $h = \omega(a^\dagger a + 1/2) + \omega_m(b^\dagger b + 1/2)$ and

$$V^\alpha = \frac{\eta^2\omega}{4}[(1-\alpha)^2(a^\dagger + a)^2 + \delta\alpha^2(b^\dagger + b)^2] + iu_\alpha^-(ab^\dagger - a^\dagger b) + iu_\alpha^+(a^\dagger b^\dagger - ab), \tag{140}$$

with $\eta = -q/\omega\sqrt{mv}$ a dimensionless coupling parameter, $\delta = \omega/\omega_m$, and

$$u_\alpha^\pm = \frac{\eta\omega_m}{2}\sqrt{\delta}[(1-\alpha) \mp \delta\alpha]. \tag{141}$$

The value of α, which determines the physical definitions of the two oscillator subsystems, can have a profound effect on the form of V^α. This is completely eliminated, however, if we assume weakly coupled nearly resonant oscillators. We can then let $\omega_m = \omega$ and neglect terms quadratic in η. We can also perform the rotating-wave approximation by setting $u_\alpha^+ = 0$. The final result is the α-independent Hamiltonian $H = h + V$, where $h = \omega(a^\dagger a + b^\dagger b + 1)$ and

$$V^\alpha = V = \frac{i}{2}\omega\eta(ab^\dagger - a^\dagger b). \tag{142}$$

This Hamiltonian satisfies bare-energy conservation,

$$[h, H] = 0, \tag{143}$$

which we saw, in the context of the S matrix, was crucial in eliminating subsystem gauge relativity. We have obtained the same result here in a direct manner. We can now pretend that the two oscillators represent unique physical subsystems.

Outside of the regime of validity of weak-coupling approximations, it is typically thought that one cannot let $u_\alpha^+ \approx 0$. In general, this is true, by which we mean that one can use this approximation independent of the value of α only in the weak-coupling regime. However, whether V^α includes counterrotating terms depends on the value of α, so there is a range of values for which the rotating-wave approximation will remain valid well into the ultrastrong-coupling regime. For a specific choice of α the rotating-wave approximation is exact (Drummond, 1987; Stokes *et al.*, 2012; Stokes and Nazir, 2018, 2019, 2021b). Specifically, by choosing

$$\alpha(\omega) = \alpha_{\mathrm{JC}}(\omega) \coloneqq \frac{\omega_m}{\omega_m + \omega} \tag{144}$$

we obtain $u_\alpha^+ \equiv 0$, so the counterrotating terms in the bilinear component of V^α in Eq. (140) are automatically eliminated. As before, by performing nonmixing Bogoliubov transformations within the separate light$_{\mathrm{JC}}$ and matter$_{\mathrm{JC}}$ Hilbert spaces, we can eliminate terms quadratic in η via modes c and d such that

$$\frac{p^2}{2m} + \frac{m\omega_m^2}{2}x^2 + \frac{q^2}{2v}\alpha_{\mathrm{JC}}^2 r^2 = \tilde{\omega}_m\left(d^\dagger d + \frac{1}{2}\right), \tag{145}$$

$$\frac{v}{2}(\Pi^2 + \omega^2 A^2) + \frac{q^2}{2m}(1-\alpha_{\mathrm{JC}})^2 A^2 = \tilde{\omega}\left(c^\dagger c + \frac{1}{2}\right), \tag{146}$$

where $\tilde{\omega}_m^2 = \omega_m^2\mu$ and $\tilde{\omega}^2 = \omega^2\mu$, in which $\mu = 1 + [\eta\omega/(\omega_m + \omega)]^2$. In the single-mode case this elimination of self-energy terms is exact. It follows that α_{JC} can be written as $\alpha_{\mathrm{JC}} = \tilde{\omega}_m/(\tilde{\omega} + \tilde{\omega}_m)$. The corresponding Hamiltonian is

$$H_{\mathrm{JC}} = \tilde{\omega}_m(d^\dagger d + \tfrac{1}{2}) + \tilde{\omega}(c^\dagger c + \tfrac{1}{2}) - iq\sqrt{\frac{\omega\omega_m}{mv}}\frac{1}{\omega_m + \omega}(d^\dagger c - dc^\dagger). \tag{147}$$

The ground state is represented by the vacuum of the c and d modes ($|G_{\mathrm{JC}}\rangle = |0_d, 0_c\rangle$). We emphasize that at no point have we made use of any approximations or assumptions that ruin the gauge invariance of the theory. Nor, however, have we performed a diagonalizing transformation of the Hamiltonian. We have simply considered a definition of the subsystems specified by a value α_{JC} between the commonly chosen values $\alpha = 0$ and 1, and within this gauge we have only performed nonmixing Bogoliubov transformations of the form $U_m \otimes U_{\mathrm{ph}}$. Whether or not the latter transformations are employed, counterrotating terms in the linear interaction are absent because $u_{\mathrm{JC}}^+ \equiv 0$. Thus, we make the following statement.

- It is premature to define the paradigm of extreme-coupling light-matter physics through properties such as high levels of ground-state light-matter entanglement and photon population, which result from terms appearing in commonly chosen gauges but which are not necessarily present.

There are no ground-state virtual excitations in the modes c and d when they are defined relative to the gauge α_{JC}. We see in Sec. VI.C [see also note XIII of the Supplemental Material (245)] that as a result matter cannot be fully localized in this gauge. Finally, we remark that although in this example a projection $P = |0_d\rangle\langle 0_d| + |1_d\rangle\langle 1_d|$ onto the first two levels of the material oscillator is as ill justified as it can ever be as an approximation (because the matter system is harmonic), such a projection nevertheless yields the exact ground state ($P|G_{\mathrm{JC}}\rangle = |G_{\mathrm{JC}}\rangle$). This fact is relevant to our discussion of material truncation in Sec. IV.

2. Quantum optical master equation

We now turn our attention to a more realistic setting by deriving the quantum optical master equation for the dipole$_\alpha$, which can be viewed as a detector for the corresponding α-gauge radiation field. We show that the weak-coupling approximations comprising the traditional quantum-optics paradigm have the effect of mimicking the S matrix, and they thereby cause all α dependence to drop out of the final result. More precisely, they ensure that all master equation coefficients are well-known second-order QED matrix elements. A similar demonstration was given for a pair of two-level dipoles by Stokes and Nazir (2018). Here we consider only one dipole (the detector), but we do not restrict our attention to only two dipolar energy levels. The Hamiltonian reads

$$H = h + V_1^\alpha + V_2^\alpha, \tag{148}$$

$$h = \sum_n \epsilon^n |\epsilon^n\rangle\langle\epsilon^n| + \int d^3k \sum_\lambda \omega \left(a_\lambda^\dagger(\mathbf{k}) a_\lambda(\mathbf{k}) + \frac{1}{2} \right), \tag{149}$$

$$V_1^\alpha = -(1-\alpha)\frac{q}{m}\mathbf{p}\cdot\mathbf{A}_{\mathrm{T}}(\mathbf{0}) + \alpha q \mathbf{r}\cdot\mathbf{\Pi}(\mathbf{0}), \tag{150}$$

$$V_2^\alpha = (1-\alpha)^2 \frac{q^2}{2m}\mathbf{A}_{\mathrm{T}}(\mathbf{0})^2 + \frac{\alpha^2 q^2}{2}\mathbf{r}\cdot\delta^{\mathrm{T}}(\mathbf{0})\cdot\mathbf{r}, \tag{151}$$

where h, V_1^α, and V_2^α are zeroth, first, and second order, respectively, in q.

We make the following weak-coupling approximations concerning the state of the detector$_\alpha$ represented by the density operator $\rho(t)$ in a suitable interaction picture:

(1) *Born approximation*.—The dipole and reservoir are uncorrelated over the relevant timescale.
(2) *Second-order perturbation theory*.—The coupling is much smaller than the unperturbed energies.
(3) *Markov approximation A*.—The system dynamics are memoryless. $\rho(s) \approx \rho(t)$ for all $s \in [0, t]$.
(4) *Markov approximation B*.—The temporal limit of the integrated von Neumann equation is $t \approx \infty$.
(5) *Secular (rotating-wave) approximation*.—Rapidly oscillating contributions are negligible.

The Markov approximations mimic the adiabatic switching condition of the S matrix, and together with the secular approximation they enforce bare-energy conservation.

The derivation of the quantum optical master equation is well known (Breuer and Petruccione, 2007), but we repeat it in note XI of the Supplemental Material (245) using an arbitrary gauge α in order to show how approximations (1)–(5) cause all α dependence to drop out. Specifically, approximation (1) ensures that the master equation coefficients can be calculated using the photonic vacuum at any time t. Approximation (2) ensures that they are second order in q. Approximation (3) ensures that they can be calculated independent of ρ. Approximation (4) ensures that the expected energy denominators are obtained as in the T matrix, and approximation (5) ensures that they are evaluated on energy shell. By reducing all master equation coefficients to well-known QED matrix elements, approximations (1)–(5) ensure α independence.

In the Schrödinger picture the final result reads

$$\dot\rho = i[\rho, \bar H_m] + \sum_{\substack{n,m \\ n>m}} \Gamma_{nm}\left(L_{nm}\rho L_{nm}^\dagger - \frac{1}{2}\{L_{nm}^\dagger L_{nm}, \rho\} \right), \tag{152}$$

where $\bar H_m = H_m + \Delta$ and where Δ and Γ_{nm} are α-independent QED matrix elements, namely, the Lamb shift and the Fermi-golden-rule spontaneous emission rate, respectively; see note XI of the Supplemental Material (245). The Lindblad operators are $L_{nm} = |\epsilon^m\rangle\langle\epsilon^n|$. The master equation (152) is readily extended to a finite temperature reservoir (Breuer and Petruccione, 2007). The following is evident.

- The reduced description of the detector$_\alpha$ is α independent within approximations (1)–(5), such that "detector" becomes an ostensibly unique theoretical concept.

The stationary state ρ_0 of this detector is the bare ground state $\rho_0 = |\epsilon^0\rangle\langle\epsilon^0|$, according to which the probability of excitation of the detector initially in the ground state is $\mathcal{P}_{\mathrm{d},0}(t) = 0$ for all t. Within the approximations made, photon emission requires a downward dipolar transition and absorption requires an upward one. Furthermore, the energies of any photons involved must be exactly equal to the energies of the corresponding dipolar transitions involved. The processes captured by the master equation (152) are precisely those captured by the S matrix where h is strictly conserved.

Outside of approximations (1)–(5) emission and absorption can occur without preserving the number of bare quanta, but evidently such virtual processes are not perfectly bare energy conserving and they are nonsecular and/or non-Markovian inasmuch as they are eliminated only when both Markov and secular approximations are performed. These processes are allowed (not only as intermediates) by the more general matrix $S^{(\tau)}$ defined in Eq. (136) and, although they are viewed as unphysical in scattering theory (except as intermediates), in open quantum systems theory the opposite is true: they are allowed unless they have been suppressed by approximations whose avoidance must provide a more accurate description. Moreover, these approximations have a relatively narrow regime of validity (Breuer and Petruccione, 2007). There is presently considerable interest within open quantum systems theory in understanding strong-coupling and non-Markovian effects using both numerical and analytical methods (Ishizaki *et al.*, 2010; Breuer *et al.*, 2016; Nazir and McCutcheon, 2016; de Vega and Alonso, 2017; Nazir and Schaller, 2018). From this perspective, when approximations (1)–(5) break down, the idealizations used to define the S matrix must be interpreted as no longer realistic.

D. Time-dependent interactions and ground-state photons

We now turn our attention to nonadiabatic interaction switching whereby the gauge relativity of subsystems cannot be ignored. It is sometimes argued that the Coulomb gauge must be used to describe the residual photon population left after a sufficiently fast interaction switch off (Stefano *et al.*, 2019; Settineri *et al.*, 2021). In fact, the correct description depends on the experimental context (Stokes and Nazir, 2021b), as later discussed in detail.

The ground state of a light-matter system is gauge invariant, but its representation using a vector differs between gauges; see Sec. III.B. This gives rise to different photon number predictions, all of which are physical. The differing predictions within one and the same physical state correspond to different gauge-invariant definitions of a photon. The task remains to determine which prediction is most relevant in a given situation. For our purposes it is sufficient to consider the simple α-gauge framework, but it should be borne in mind that the gauge function is completely arbitrary and the following considerations apply generally.

For each fixed α the Hamiltonian operator H_α represents the same total energy observable E. The total energy eigenvectors are defined by $H_\alpha|E_\alpha^n\rangle = E^n|E_\alpha^n\rangle$, where the eigenvalues E^n are manifestly α independent (unitary transformations are isospectral). According to the postulates of quantum theory, the vector $|E_\alpha^n\rangle$ represents, within the gauge α, the unique physical state in which the system definitely possesses the energy E^n. Consider now the average

$$N_\alpha = \langle G_\alpha|\sum_{\mathbf{k}\lambda} a_\lambda^\dagger(\mathbf{k})a_\lambda(\mathbf{k})|G_\alpha\rangle = \langle G_\alpha|n|G_\alpha\rangle, \tag{153}$$

where the vector $|G_\alpha\rangle = |E_\alpha^0\rangle$ represents the ground state in the gauge α and $a_\lambda(\mathbf{k})$ is as defined in Eq. (71). At first glance it seems that the predicted photon number N_α is fundamentally gauge noninvariant, and that this is because $|G_\alpha\rangle$ depends on α, but this is not the case. Rather, the operator n represents the gauge-invariant number of photons defined relative to the gauge α. In the gauge α' the same observable is represented by $n' = R_{\alpha\alpha'}nR_{\alpha\alpha'}^\dagger$ and the physical ground state is represented by the vector $|G_{\alpha'}\rangle = R_{\alpha\alpha'}|G_\alpha\rangle$. Thus, N_α is gauge invariant ($N_\alpha = \langle G_\alpha|n|G_\alpha\rangle = \langle G_{\alpha'}|n'|G_{\alpha'}\rangle$). For each different fixed value of α the average N_α is that of a different physical observable, and it is therefore a different gauge-invariant prediction. The subscript α labels which gauge-invariant definition of photon is being considered. A special case is the number of $\mathbf{E}_\mathrm{T}$-type photons given by $N_0 =: N_{\mathbf{E}_\mathrm{T}}$ because $\tilde{\mathbf{\Pi}}(\mathbf{k}) = -\tilde{\mathbf{E}}_\mathrm{T}(\mathbf{k})$ when $\alpha = 0$. Another special case is the number of $\mathbf{D}_\mathrm{T}$-type photons, which is given by $N_1 =: N_{\mathbf{D}_\mathrm{T}}$, because $\tilde{\mathbf{\Pi}}(\mathbf{k}) = -\tilde{\mathbf{D}}_\mathrm{T}(\mathbf{k})$ when $\alpha = 1$.

We now consider a system prepared in the ground state before we suddenly switch off the interaction. When the interaction vanishes, photons are defined as in Eq. (71), but this definition is now unique because the noninteracting canonical momentum is unique ($\mathbf{\Pi} = -\mathbf{D}_\mathrm{T} = -\mathbf{E}_\mathrm{T} = -\mathbf{E}$). We can therefore ask how many of these unique photons are present for times $t > t_f$ if the interaction is suddenly switched off at $t = t_f$. Modeling this situation using a time-dependent coupling in the gauge α gives the Hamiltonian

$$H_\alpha(t) = H_m + H_\mathrm{ph} + \theta(t - t_f)V^\alpha(\eta), \tag{154}$$

where θ is the Heaviside step function and η is a coupling parameter such that $V^\alpha(0) = 0$. These $H_\alpha(t)$ are not equivalent to each other for different α (Stokes and Nazir, 2021b). This is unsurprising because for $\alpha \neq \alpha'$, $H_\alpha(t)$ and $H_{\alpha'}(t)$ model two different experiments in which V^α and $V^{\alpha'}$ are suddenly switched off, respectively. For each α the evolution generated by $H_\alpha(t)$ from time $t = 0$ consists of sequential evolutions $[U_\alpha(t) = e^{-i(H_m+H_\mathrm{ph})(t-t_f)}e^{-iH_\alpha t_f}]$. It follows that the gauge-invariant physical prediction $N_\alpha =: N_{\mathbf{E}_\mathrm{T}+\alpha\mathbf{P}_\mathrm{T}}$ gives the number of photons left over in an experiment realizing a sudden switch off of the α-gauge interaction. Settineri *et al.* (2021) noted that the prediction $N_0 = N_{\mathbf{E}_\mathrm{T}}$ is gauge invariant, but as we have shown more generally the same is true for any of the predictions N_α.

There is a famous set of experiments for which it is well known that the sudden switching condition appears to be ill justified in the Coulomb gauge compared to the multipolar gauge, namely, the early experiments of Lamb (Lamb, 1952; Power and Zienau, 1959; Fried, 1973; Davidovich and Nussenzveig, 1980; Milonni, Cook, and Ackerhalt, 1989; Woolley, 2000; Stokes, 2013). The natural line shape prediction can be obtained by assuming the atom to initially be in a bare excited state with no photons. This amounts to a sudden switch on of the interaction (Milonni, Cook, and Ackerhalt, 1989). Within the multipolar gauge the prediction is sufficiently close to the experimental result to rule out the corresponding Coulomb-gauge prediction (Power and Zienau, 1959; Fried, 1973; Davidovich and Nussenzveig, 1980; Milonni, Cook, and Ackerhalt, 1989; Woolley, 2000; Stokes, 2013). In other words, the multipolar-gauge subsystems are more relevant for the description of this experiment. The natural line shape of spontaneous emission is discussed in detail in Sec. VI.B.

One can consider more general time-dependent interactions and the same considerations will apply. The generalization can be achieved by letting

$$H_\alpha(t) = H_m + H_\mathrm{ph} + V^\alpha(\eta\mu(t)) \tag{155}$$

where $\mu(t)$ is an arbitrary coupling envelope that vanishes smoothly after some time t_f. We assume, as Stokes and Nazir (2021b) did, that $\mu(t)$ vanishes before some time t_i, so the system can be prepared at $t = 0 < t_i$ in the ground state represented by eigenvector $|g\rangle$ of $h = H_m + H_\mathrm{ph}$. The total number of photons at a time $t > t_f$ is

$$N_\alpha(t) = \langle g|U_\alpha(t)nU_\alpha(t)^\dagger|g\rangle, \tag{156}$$

where $U_\alpha(t)$ is the evolution operator generated by $H_\alpha(t)$. To prove the gauge invariance of $N_\alpha(t)$, one must take into account that gauge transformations are now time dependent because they depend on the coupling parameter ($R_{\alpha\alpha'}[\eta\mu(t)] \equiv R_{\alpha\alpha'}(t)$). The vector $|g_\alpha(t)\rangle = U_\alpha(t)|g\rangle$ represents the Schrödinger-picture state at the time t in the gauge α. The same physical state is represented in the gauge α' by the vector $|g_\alpha^{\alpha'}(t)\rangle = R_{\alpha\alpha'}(t)|g_\alpha(t)\rangle$. The physical observable represented by n in the Schrödinger picture in the gauge α is represented by $n^{\alpha'}(t) = R_{\alpha\alpha'}(t)nR_{\alpha\alpha'}(t)^\dagger$ in the gauge α'. We see therefore that $N_\alpha(t)$ is a gauge-invariant prediction.

The two different vector representations $|g_\alpha(t)\rangle$ and $|g_\alpha^{\alpha'}(t)\rangle$ of the state at t satisfy the Schrödinger equations $id|g_\alpha(t)\rangle/dt = H_\alpha(t)|g_\alpha(t)\rangle$ and $id|g_\alpha^{\alpha'}(t)\rangle/dt = H_\alpha^{\alpha'}(t)|g_\alpha^{\alpha'}(t)\rangle$. The Hamiltonians $H_\alpha^{\alpha'}(t)$ and $H_\alpha(t)$ are easily related via direct differentiation of the expression $|g_\alpha^{\alpha'}(t)\rangle = R_{\alpha\alpha'}(t)|g_\alpha(t)\rangle$, which implies that

$$H_{\alpha}^{\alpha'}(t) = R_{\alpha\alpha'}(t)H_{\alpha}(t)R_{\alpha\alpha'}(t)^{\dagger} + i\dot{R}_{\alpha\alpha'}(t)R_{\alpha\alpha'}(t)^{\dagger}. \quad (157)$$

It is a trivial matter to generate an equivalent model to any one of the $H_{\alpha}(t)$ by properly accounting for the time dependence of gauge transformations.

The Hamiltonian $H_{\alpha}^{\alpha'}(t)$ depends on two parameters α and α' that have different roles. The parameter α selects the gauge within which the time-dependent coupling assumption $e \to e(t)$ has been made, whereas the parameter α' selects the choice of gauge used for calculations after this assumption has been made. The nonequivalence of the $H_{\alpha}(t)$ for different α shows that $e \to e(t)$ constitutes a different physical assumption in different gauges. In other words, gauge ambiguities arise because each $H_{\alpha}(t)$ generates its own equivalence class $\mathcal{S}_{\alpha} = \{H_{\alpha}^{\alpha'}(t) : \alpha' \in \mathbb{R}\}$, and distinct classes describe different experiments. The prediction $N_{\alpha}(t)$ is relevant if the experimental protocol modeled happens to realize a switch on and switch off of the interaction V^{α}. For example, if the experimental arrangement considered is somehow capable of effectively manipulating the gauge-invariant bare dipole moment $q\mathbf{r}$, then the multipolar-gauge interaction might be controlled.

These points were directly demonstrated by Stokes and Nazir (2021b), who considered the concrete setup of a dipole uniformly moving in and out of a Gaussian cavity mode, as depicted in Fig. 10. This situation can be modeled using a Gaussian envelope $\mu(t)$. In addition to the nonequivalent models $H_{\alpha}(t)$, a more complete description $\tilde{H}_{\alpha}(t)$ is provided by retaining an explicit model for the control system, which in this example is the center-of-mass motion of the dipole. Unlike $H_{\alpha}(t)$, the more complete descriptions $\tilde{H}_{\alpha}(t)$ are equivalent to each other for different α. In this way, the procedure of using a time-dependent coupling $\eta\mu(t)$ can be viewed as an approximation. The value of α such that $\tilde{H}_{\alpha}(t) = H_{\alpha}(t)$ is then the correct value to choose when describing the experiment using the result of this approximation $H_{\alpha}(t)$.

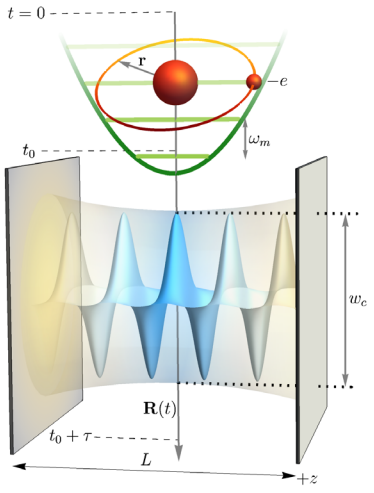

FIG. 10. Cavity of length L supporting standing waves in the z direction and a Gaussian perpendicular mode profile with waist w_c, along with a dipole $-er$ oscillating with the frequency ω_m. At $t = 0$ the cavity and dipole are noninteracting. The dipole follows a classical trajectory $\mathbf{R}(t)$ through the cavity, entering the cavity at t_0 and exiting at $t_0 + \tau$.

Stokes and Nazir (2021b) showed that if there is a value α for which $\tilde{H}_{\alpha}(t) = H_{\alpha}(t)$, then the value strongly depends on the experimental protocol. The prediction $N_{\alpha}(t)$ obtained using $H_{\alpha}(t)$ is correct if and only if the dipole moment is aligned with the mode polarization and these vectors make an angle θ with the direction of the center-of-mass motion such that $\cos^2\theta = \alpha$. The result demonstrates that in general which prediction $N_{\alpha}(t)$ is the correct (relevant) one strongly depends on the experimental context. It is not the case that the Coulomb-gauge prediction $N_0(t)$ is always correct. This result further illustrates why there are indeed gauge ambiguities. To find which of the predictions $N_{\alpha}(t)$ may be relevant for describing a concrete setup and experimental protocol, Stokes and Nazir (2021b) resorted to invoking an explicit model of the control system. The result obtained could not be anticipated without such a description, yet such descriptions are available in only the simplest of cases where the control system accommodates tractable modeling.

VI. MEASUREMENTS AND VIRTUAL PHOTONS

We now turn to the topic of subsystem measurements. Their description when dealing with virtual processes within the weak-coupling regime was considered some time ago via simple models (Compagno, Passante, and Persico, 1988a, 1988b, 1990, 1991, 1995). The topic was recently addressed during discussions of ultrastrong interactions (Di Stefano *et al.*, 2018; Settineri *et al.*, 2021). We focus on a system consisting of a source and/or a detector within a single photonic environment. This situation is distinguished from the case of a source and a detector occupying different environments that are modeled separately, such as a source within a cavity with a detector external to the cavity. The outlook for the latter situation is discussed beginning in Sec. VII.C.

The natural starting point for our considerations is Glauber's photodetection theory (Glauber, 1963, 2007). We review aspects of photodetection that are important beyond the standard quantum-optics paradigm, including how photodetection divergences are related to virtual excitations. We consider the gauge relativity of the predicted natural line shape of spontaneous emission and determine the relation between subsystem gauge relativity, locality, and dressing.

The main conclusion of this section is that, outside of conventional weak-coupling and Markovian regimes, there is necessarily a trade-off between defining material systems as localized objects and avoiding virtual vacuum excitations. In the multipolar gauge material systems are the most localized. We see in Sec. VI.C that if such a detector is deemed accessible and therefore prepared in its lowest energetic state, then under the influence of the interaction it will necessarily become excited even within the corresponding photonic vacuum because this state is not the ground state of the interacting composite. These virtual excitations are not encountered if one instead defines physical subsystem excitations relative to the true ground state. This, however, constitutes a definition of the physical subsystems relative to an unconventional gauge (neither Coulomb nor multipolar). Material systems defined in this way are necessarily delocalized to some extent. Thus, while in practice a detection process necessarily possesses a finite extent in space and time,

theoretically some degree of spatial localization of a detector must be sacrificed if one wants to eliminate the prediction of its virtual excitation.

A. Conventional photodetection theory and its limitations

Glauber photodetection theory (Glauber, 1963, 2007) has been a major workhorse in weak-coupling quantum optics and constitutes a natural starting point. Here we review this theory and its limitations. Photodetection in the context of ultrastrongly coupled light-matter systems is discussed in Secs. VII.C and VII.D.

1. Real excitations

Typical photodetectors work by amplifying photon ionization to produce a macroscopic current. As such they are substantial objects consisting of photoconductive electrons over a cross-sectional area S that is correlated with detection efficiency. In addition to being large, such detectors are also typically slow to respond, at least compared to the correlation times of the photonic reservoirs that they monitor. Thus, actual photon measurements are not restricted to individual points in spacetime, and this fact is certainly relevant outside of weak-coupling regimes. However, as a model for dealing with weakly coupled detectors, we may consider a localized detector dipole $\mathbf{d}=q\mathbf{r}$ fixed at the origin 0. The charge q is a suitable perturbation parameter (with the fine-structure constant being $q^2/4\pi$).

In each gauge the unperturbed eigenvectors of $h=H_\alpha-V^\alpha$ represent different physical states. Photons are by definition quanta of the light subsystem and a detector is a material subsystem. A photodetection process therefore involves an energetic change of the material system, usually accompanied by a change in the number of photons; i.e., it is a process between unperturbed states. In general these states do not coincide with well-defined states of energy of the light-matter composite, and thus they are not stationary. An examination of photodetection probabilities in a gauge α provides insight into the physical natures of the light and matter subsystems defined relative to the gauge α.

In conventional treatments (Glauber, 2007) a linear dipolar form of coupling is adopted, as occurs in the multipolar gauge. This is often written as either $V^1=-\mathbf{d}\cdot\mathbf{E}_\mathrm{T}(\mathbf{0})$ or $V^1=-\mathbf{d}\cdot\mathbf{E}(\mathbf{0})$. Neither expression is correct. As noted in Sec. II.F, the correct linear part of the multipolar interaction in the EDA is $V^1=-\mathbf{d}\cdot\mathbf{D}_\mathrm{T}(\mathbf{0})$. Two further common misconceptions are that the Coulomb gauge defines photons using the electric field, and that this is the basic field that first enters into Glauber's photodetection theory (Settineri *et al.*, 2021). In fact, the Coulomb gauge defines photons using $\mathbf{E}_\mathrm{T}\neq\mathbf{E}$ and, at the dipole's position $\mathbf{0}$, the relevant field $\mathbf{D}_\mathrm{T}$ is also infinitely different from $\mathbf{E}(\mathbf{0})=\mathbf{D}_\mathrm{T}(\mathbf{0})-q\mathbf{r}\delta(\mathbf{0})$. However, the infinite term $\mathbf{P}_1(\mathbf{x})=q\mathbf{r}\delta(\mathbf{0})$ is a difference in the source components of the two fields, which are at least first order in q. Since the detector's dipole moment is first order in q, only the free component of $\mathbf{D}_\mathrm{T}(\mathbf{0})$ contributes to detection probabilities to second order in q for an initially unperturbed state, and it may be taken to coincide with the free component of $\mathbf{E}(\mathbf{0})$.

We begin by following conventional treatments, which define the subsystems photons$_\alpha$ and detector$_\alpha$ relative to the multipolar gauge $\alpha=1$, then employ perturbation theory to second order in q. The probability of finding the detector$_1$ excited into the nth level at time t, given the initial detector-phonon state $|\epsilon^m,\psi_\mathrm{ph}\rangle$ with a fixed number of photons$_1$ and with $m<n$, is

$$\mathcal{P}_\mathrm{d}^{nm}(t)=d_{nm,i}d_{mn,j}\int_0^t ds\int_0^t ds' e^{i\omega_{nm}(s'-s)}G_{ij}(s,s'),\tag{158}$$

where repeated indices are summed and

$$G_{ij}(s,s')=\langle\psi_\mathrm{ph}|E_{\mathrm{vac},i}(s,\mathbf{0})E_{\mathrm{vac},j}(s',\mathbf{0})|\psi_\mathrm{ph}\rangle,\tag{159}$$

in which

$$\mathbf{E}_\mathrm{vac}(t,\mathbf{x})=-i\int d^3k\sum_\lambda\sqrt{\frac{\omega}{2(2\pi)^3}}\mathbf{e}_\lambda(\mathbf{k})\times[a_\lambda^\dagger(0,\mathbf{k})e^{i\omega t-i\mathbf{k}\cdot\mathbf{x}}-a_\lambda(0,\mathbf{k})e^{-i\omega t+i\mathbf{k}\cdot\mathbf{x}}]\tag{160}$$

denotes the free component of $\mathbf{D}_\mathrm{T}(t,\mathbf{x})$. Since $\omega_{nm}>0$, the antinormally ordered contribution in Eq. (158) is taken to be rapidly oscillating and is neglected in a RWA such that we may let

$$G_{ij}(s,s')=\langle\psi_\mathrm{ph}|E^{(-)}_{\mathrm{vac},i}(s,\mathbf{0})E^{(+)}_{\mathrm{vac},j}(s',\mathbf{0})|\psi_\mathrm{ph}\rangle,\tag{161}$$

where

$$\mathbf{E}^{(+)}_\mathrm{vac}(t,\mathbf{0})=i\int d^3k\sum_\lambda\sqrt{\frac{\omega}{2(2\pi)^3}}\mathbf{e}_\lambda(\mathbf{k})a_\lambda(0,\mathbf{k})e^{-i\omega t},\qquad\mathbf{E}^{(-)}=(\mathbf{E}^{(+)})^\dagger.\tag{162}$$

We see that normal ordering occurs as an approximation based on the detector$_1$ excitation process having a supposedly dominant contribution coming from photon$_1$ absorption. The neglected contribution is virtual, i.e., number nonconserving, and corresponds to detector$_1$ excitation with emission of a photon$_1$.

The detector$_1$ level n typically belongs to the ionization continuum, and after excitation a number of physical processes must occur for a detection event to actually be registered. The description of these processes is subsumed into a classical epistemic probability $\mathcal{D}_n$ for a detection event given excitation to the level n. The total probability of detection is therefore

$$\mathcal{P}_\mathrm{d}^m(t)=\sum_n\mathcal{D}_n\mathcal{P}_\mathrm{d}^{nm}(t),\tag{163}$$

where formally the summation over n is understood to include integration over continuum levels. Defining the spectral density (sensitivity)

$$\mathcal{S}_{ij}(\omega)=2\pi\sum_n d_{nm,i}d_{mn,j}\mathcal{D}_n\delta(\omega-\omega_{nm})\tag{164}$$

enables one to model different detection schemes by assuming different forms of $\mathcal{S}_{ij}(\omega)$. The photon$_1$ counting rate is

$$\frac{d\mathcal{P}_{\mathrm{d}}^{nm}}{dt} = 2\mathrm{Re}\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}\mathcal{S}_{ij}(\omega)\mathcal{G}_{ij}(\omega,t), \tag{165}$$

where

$$\mathcal{G}_{ij}(\omega,t) = \int_0^t ds\, e^{i\omega(t-s)}G_{ij}(s,t), \tag{166}$$

whose Fourier transform is

$$\begin{aligned}\mathcal{G}_{ij}(s,t) &= \int\frac{d\omega}{2\pi}e^{i\omega s}\mathcal{G}_{ij}(\omega,t)\\ &= \theta(s)\theta(t-s)G_{ij}(t-s,t),\end{aligned} \tag{167}$$

which vanishes unless $0 \le s \le t$. Since photodetectors are slow, the measurement time t is typically much longer than the reservoir correlation time $T_c = 1/\Delta\omega_G$, where $\Delta\omega_G$ is the bandwidth of the correlation function G_{ij}. Therefore, the s width of $\mathcal{G}_{ij}(s,t)$ is approximately T_c.

Glauber (2007) defined an ideal broadband detector as one with a flat spectral density $\mathcal{S}_{ij}(\omega) = \mathcal{S}_{ij}$. This requires that the width of the sensitivity function must be much larger than $\Delta\omega_G = 1/T_c$, such that $\mathcal{G}_{ij}(\omega,t)$ is sharply peaked as a function of ω when compared to $\mathcal{S}_{ij}(\omega)$. The photon counting rate is then simply $S_{ij}\mathcal{G}_{ij}(t,t)$, such that if $S_{ij}\sim\delta_{ij}$, then the rate is proportional to the Glauber intensity

$$I_G(t) = \langle \mathbf{E}_{\mathrm{vac}}^{(-)}(t,\mathbf{0})\cdot\mathbf{E}_{\mathrm{vac}}^{(+)}(t,\mathbf{0})\rangle. \tag{168}$$

2. Virtual excitations

As in the textbook by Mandel and Wolf (1995), the actual field involved in photodetection theory can be left open by defining

$$\mathbf{F}^{(+)}(t,\mathbf{x}) = \int\frac{d^3k}{\sqrt{2(2\pi)^3}}\sum_\lambda \mathbf{e}_\lambda(\mathbf{k})\beta(\omega)a_\lambda(t,\mathbf{k})e^{i\mathbf{k}\cdot\mathbf{x}}, \tag{169}$$

where a number of noteworthy choices of $\beta(\omega)$ can be made. For example, if $\beta(\omega) = i\sqrt{\omega}$, then $\mathbf{F} = \mathbf{D}_{\mathrm{T}}$. If $\beta(\omega) = 1/\sqrt{\omega}$, then $\mathbf{F} = \mathbf{A}_{\mathrm{T}}$. If $\beta(\omega) = 1$, then $\mathbf{F}^{(+)}$ defines a direct inverse Fourier transform of $\sum_\lambda \mathbf{e}_\lambda(\mathbf{k})a_\lambda(\mathbf{k})/\sqrt{2}$. This last choice of β is noteworthy for the reason that, although it is impossible to define a local number operator for relativistic quanta (Fulling, 1989; Mandel and Wolf, 1995; Haag, 1996), the operator $\mathbf{F}^{(-)}(\mathbf{x})\cdot\mathbf{F}^{(+)}(\mathbf{x})$ can be interpreted as a real-space number density of photons that is approximately localized on a scale much larger than the corresponding wavelengths (Mandel and Wolf, 1995); see also note XII of the Supplemental Material (245). As it is local in $\mathbf{k}$ space, the relation between fields corresponding to different $\beta(\omega)$ in Eq. (169) is highly nonlocal in spacetime. This point is relevant to understanding the interplay between electromagnetic dressing and localization and is discussed further in note XIII of the Supplemental Material (245).

To understand the limitations of conventional photodetection theory, we return to Eq. (158). If we assume the vacuum state $|\psi_{\mathrm{ph}}\rangle = |0\rangle$ and we allow the levels m and n to be arbitrary, then evaluating the polarization summation and angular integrals gives

$$\mathcal{P}_{\mathrm{d,vac}}^{nm}(t) = \frac{|\mathbf{d}_{nm}|^2}{3\pi}\int_0^\infty d\omega\,\omega^3\frac{\sin^2[(\omega_{mn}-\omega)t/2]}{\pi(\omega_{mn}-\omega)^2/2}. \tag{170}$$

If $m > n$, the process described is spontaneous emission. If $n > m$, then the process described is virtual. The dominant peak of the integrand then lies outside of the domain of integration and is oscillatory for positive frequencies. The amplitude of the oscillations in the integrand grows with ω due to the prefactor of ω^3. This behavior is bounded only by an ultraviolet cutoff ω_M, and the integral is in fact quadratically divergent with ω_M. The divergence is relatively severe, such that $\mathcal{P}_{d,\mathrm{vac}}^{nm}(t)$ is non-negligible even for realistic yet modest values of ω_M that are consistent with the EDA and the nonrelativistic treatment (Drummond, 1987; Stokes *et al.*, 2012).

If we repeat the derivation of the detector excitation rate for a detector$_0$, i.e., for a detector defined relative to the Coulomb gauge, then the field entering into the theory is now $\mathbf{A}_{\mathrm{T}}(\mathbf{0})$, which amounts to letting $\beta(\omega) = 1/\sqrt{\omega}$ in Eq. (169). In place of Eq. (170), we obtain

$$\mathcal{P}_{d,\mathrm{vac}}^{nm}(t) = \frac{|\mathbf{d}_{nm}|^2}{3\pi}\int_0^\infty d\omega\,\omega\omega_{mn}^2\frac{\sin^2[(\omega_{mn}-\omega)t/2]}{\pi(\omega_{mn}-\omega)^2/2}. \tag{171}$$

When $n > m$, the probability is in this case only logarithmically divergent. This is a direct consequence of the $\mathbf{k}$-space normalization of the field $\mathbf{A}_{\mathrm{T}}$, which varies as $1/\sqrt{\omega}$.

The probability $\mathcal{P}_{d,\mathrm{vac}}^{nm}(t)$ is generally nonzero because the initial unperturbed state consisting of no photons and m excitations of the detector is not an eigenstate of the Hamiltonian and, in particular, it is not the ground state even if m is the lowest dipolar level. If this final result is deemed unphysical, then we must conclude that the assumed physical states are not operationally relevant in the description of photodetection. In particular, if the physical detector is not the localized detector$_1$, then it must be delocalized to some extent. The interplay between localization and dressing is discussed beginning in Sec. VI.C.

The virtual detection probability $\mathcal{P}_{d,\mathrm{vac}}^{nm}(t)$, with $n > m$, was removed in the progression from Eq. (158) to Eq. (165) using the RWA. The counting rate $d\mathcal{P}_{d,\mathrm{vac}}^{nm}/dt$ without the RWA can be found by direct differentiation of Eq. (170) and can again be reduced to the gauge nonrelative Fermi-golden-rule rate

$$\frac{d\mathcal{P}_{\mathrm{d,vac}}^{nm}}{dt} = \begin{cases}\omega_{mn}^3|\mathbf{d}_{nm}|^2/3\pi =: \Gamma_{mn}, & n < m,\\ 0, & n > m,\end{cases} \tag{172}$$

in three different ways, all of which amount to imposing strict bare-energy conservation as in the S matrix.

(1) Differentiation of Eq. (170) yields the frequency integrand $\omega^3\sin[(\omega_{mn}-\omega)t]/(\omega_{mn}-\omega)$, which expresses a bare-energy-time uncertainty constraint.

Taking the infinite-time limit $\lim_{t\to\infty}\sin(\omega t)/\pi\omega = \delta(\omega)$ gives Eq. (172).

(2) Defining the counting rate as the difference quotient $[\mathcal{P}^{nm}_{\mathrm{d,vac}}(t)-\mathcal{P}^{nm}_{\mathrm{d,vac}}(0)]/t=\mathcal{P}^{nm}_{\mathrm{d,vac}}(t)/t$ yields via Eq. (170) the frequency integrand $\omega^3\sin^2[(\omega_{mn}-\omega)t]/([\omega_{mn}-\omega]^2t/2)$. In the limit $t\to\infty$ one obtains the right-hand side of Eq. (172) using $\lim_{t\to\infty}\sin^2(\omega t/2)/(\pi\omega^2 t/2)=\delta(\omega)$. Meanwhile, the derivative $d\mathcal{P}^{nm}_{\mathrm{d,vac}}/dt$ on the left-hand side of Eq. (172) is recovered in the limit $t\to 0$. This shows that the procedure for obtaining Eq. (172) constitutes a form of Markov approximation that requires a clear separation of timescales as specified by the *Markovian regime* $1/\omega_{mn}\ll t\ll 1/\Gamma_{mn}$. The final result is valid provided that matrix elements of the interaction Hamiltonian between initial and final unperturbed states are sufficiently small and slowly varying, as demonstrated in method (3).

(3) Evaluating the prefactor ω^3 in Eq. (170) at resonance ($\omega=\omega_{mn}$) is valid if it can be considered sufficiently slowly varying compared to the peak in $\sin^2[(\omega_{mn}-\omega)t]/([\omega_{mn}-\omega]^2t/2)$ near ω_{mn}. One may then extend the lower integration limit to $-\infty$ by supposing that the integrand is dominated by this peak for sufficiently long times ($\omega_M t\gg 1$). This again yields Eq. (172).

It is not clear that any of the procedures (1), (2), or (3) can be justified for virtual excitation with $n>m$, because as already noted the dominant peak in $\sin^2[(\omega_{mn}-\omega)t]/([\omega_{mn}-\omega]^2t/2)$ then lies outside of the range of integration and the integral diverges quadratically with ω_M. In this sense virtual contributions are non-Markovian.

Both of the predictions in Eqs. (170) and (171) are gauge invariant in the sense of Eq. (132), but without use of the Markovian approximation they are clearly different. This is an example of the gauge relativity expressed by inequality (133), which as noted in Sec. V.A becomes important outside of Markovian regimes. We note that, in any gauge, if the RWA is avoided and the broadband limit is taken, then the photon counting rate is $S_{ij}G_{ij}(t,t)$ with $G_{ij}(t,t)$ given by Eq. (161) rather than Eq. (166). Thus, a generally large virtual contribution occurs. However, the broadband limit is inapplicable to this contribution because the vacuum has infinite bandwidth. Thus, the significance of such contributions is in general dependent on the measurement schemes available.

In comparing the different predictions given by Eqs. (170) and (171), Power and Thirunamachandran (1999a, 1999b) noted that which one is the more accurate will depend on which set of distinct physical states represented by the same unperturbed vectors within the two gauges is closer to the states actually realized in the considered experiment. Power and Thirunamachandran also noted that experiments could be used to determine which descriptions are most appropriate. Spectroscopic experimental signatures, in particular, are discussed in Secs. VI.B and VII.D.

The elimination of divergent contributions requires "renormalization" of the bare dipole by defining the "physical" dipole relative to the appropriate gauge, as recognized some time ago by Drummond (1987). One can use the elimination of virtual excitations as a criterion by which to select the most operationally relevant subsystem definitions, that is, to select the most appropriate gauge relative to which the dipole is to be defined in the context of photodetection. To this end we consider a one-dimensional dipole harmonically quantized in the direction $\hat{\mathbf{u}}$ with the canonical operators $\mathbf{r}=\hat{\mathbf{u}}(b^\dagger+b)/\sqrt{2m\omega_m}$ and $\mathbf{p}=i\hat{\mathbf{u}}(b^\dagger-b)\sqrt{m\omega_m/2}$. From early on purely bosonic models of this kind have been relevant to ultrastrong coupling in polaritonic systems with quantum wells and microcavities (Ciuti, Bastard, and Carusotto, 2005; Ciuti and Carusotto, 2006; Todorov *et al.*, 2010; Bamba and Ogawa, 2012).

We consider gauges of the form specified in Eq. (53) while assuming that $\alpha(\mathbf{k})=\alpha(\omega)$ is real and depends only on the magnitude of $\mathbf{k}$. We discretize the Fourier modes within a volume v and combine wave vector and polarization indices into a single-mode label, writing $\alpha(\omega)=\alpha_k$. The polarization self-energy term $\int d^3x\mathbf{P}^2_{\mathrm{T}g}/2$ can be absorbed via new material modes such that $\mathbf{r}=\hat{\mathbf{u}}(d^\dagger+d)/\sqrt{2m\tilde{\omega}_m}$ and $\mathbf{p}=i\hat{\mathbf{u}}\sqrt{m\tilde{\omega}_m}(d^\dagger-d)/\sqrt{2}$, where

$$\tilde{\omega}_m^2=\omega_m^2+\frac{q^2}{mv}\sum_k(\mathbf{e}_k\cdot\hat{\mathbf{u}})^2\alpha_k^2. \tag{173}$$

Similarly, to second order in q the field self-energy term $q^2(\mathbf{A}^{\mathrm{EDA}}_g)^2/2m$ can be absorbed via radiative mode operators c_k such that

$$\begin{aligned}a_k&=\sum_j([\cosh\theta]_{kj}c_j+[\sinh\theta]_{kj}c_j^\dagger)\\&\approx c_k+\sum_j\theta_{kj}c_j^\dagger,\end{aligned} \tag{174}$$

where the approximate equality holds to second order in q and

$$\theta_{kj}=-\frac{q^2}{2mv}\frac{\mathbf{e}_k\cdot\mathbf{e}_j(1-\alpha_k)(1-\alpha_j)}{\sqrt{\omega_k\omega_j}(\omega_k+\omega_j)}. \tag{175}$$

The arbitrary-gauge Hamiltonian can now be written correct to second order in q as

$$\begin{aligned}H_g=&\tilde{\omega}_m\left(d^\dagger d+\frac{1}{2}\right)+\sum_{k,j}\omega_{kj}\left(c_k^\dagger c_j+\frac{\delta_{kj}}{2}\right)\\&-\frac{q}{m}\mathbf{p}\cdot\tilde{\mathbf{A}}_g(\mathbf{0})+q\mathbf{r}\cdot\tilde{\mathbf{\Pi}}_g(\mathbf{0}),\end{aligned} \tag{176}$$

where $\omega_{kj}=\omega_k\delta_{kj}+(\omega_k+\omega_j)\theta_{kj}$ and

$$\tilde{\mathbf{A}}_g(\mathbf{0})\coloneqq\sum_{k,j}\frac{\mathbf{e}_k}{\sqrt{2\omega_k v}}(1-\alpha_k)[e^\theta]_{kj}(c_j^\dagger+c_j), \tag{177}$$

$$\tilde{\mathbf{\Pi}}_g(\mathbf{0})\coloneqq i\sum_{k,j}\mathbf{e}_k\sqrt{\frac{\omega_k}{2v}}\alpha_k[e^{-\theta}]_{kj}(c_j^\dagger-c_j). \tag{178}$$

Since the linear interaction components in Eq. (176) contain a prefactor of q, we may let $[e^\theta]_{kj}=\delta_{kj}$ in the mode expansions (177) and (178) to obtain results correct to second order in q. This amounts to making the straightforward replacement $a_k\to c_k$ within the interaction Hamiltonian. Similarly, when it is used within the interaction Hamiltonian, we may let

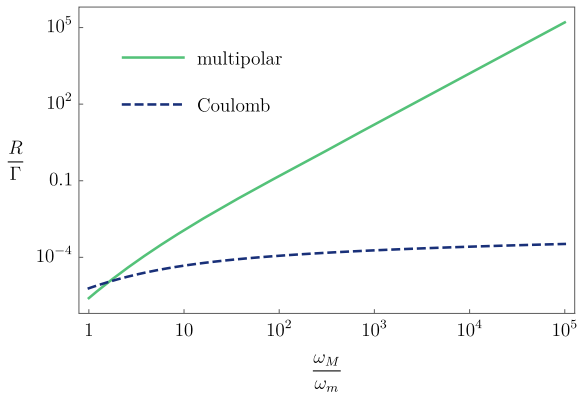

FIG. 11. The time-averaged detector excitation rate R is plotted as a function of the cutoff ω_M/ω_m in the Coulomb gauge and the multipolar gauge, assuming that $\omega_m T = 10^4$. The multipolar rate, in particular, is severely divergent with ω_M, whereas the Coulomb-gauge rate is logarithmically divergent.

$\tilde{\omega}_m = \omega_m$ within the expressions for $\mathbf{r}$ and $\mathbf{p}$ in terms of the material ladder operators, amounting to the replacement $b \to d$. The renormalization of self-terms is consistent with an interpretation in which bare frequencies are not viewed as physical. The renormalization does not affect the choice of gauge or the subsystem partition.

Assuming the initial state $|0_d, 0_c\rangle$ with no photons and no initial detector excitation, we calculate the average detector population as

$$\langle d^\dagger(t)d(t)\rangle_{0_d,0_c} = \frac{2\Gamma}{\pi}\int_0^\infty d\omega \left[\frac{\omega u^+(\omega)\sin[(\omega_m+\omega)t]}{\omega_m(\omega_m+\omega)}\right]^2, \tag{179}$$

where $\Gamma = q^2\omega_m^2/6m\pi$ is the total oscillator spontaneous emission rate into the ground state and

$$u^+(\omega) = \sqrt{\frac{\omega_m}{\omega}}\left([1-\alpha(\omega)] - \frac{\omega}{\omega_m}\alpha(\omega)\right). \tag{180}$$

The multipolar- and Coulomb-gauge results are obtained by letting $\alpha(\omega) = 1$ and 0, respectively, and are consistent with Eqs. (170) and (171), respectively. The rate $d\langle d^\dagger(t)d(t)\rangle_{0_d,0_c}/dt$ is highly oscillatory. These oscillations can be removed by taking the time average over an interval $T \gg 1/\omega_m$ defined by

$$R = \frac{1}{T}\int_0^T dt \frac{d}{dt}\langle d^\dagger(t)d(t)\rangle_{0_d,0_c} = \frac{1}{T}\langle d^\dagger(t)d(t)\rangle_{0_d,0_c} = \frac{\Gamma}{\pi T}\int_0^\infty d\omega\left[\frac{\omega u^+(\omega)}{\omega_m(\omega_m+\omega)}\right]^2, \tag{181}$$

where we have replaced $\sin^2[(\omega_m+\omega)T/2]$ in Eq. (179) with its average $1/2$ for $\omega_m T \gg 1$. The Coulomb- and multipolar-gauge time-averaged rates are plotted in Fig. 11. The multipolar rate, in particular, is quadratically divergent with ω_M and is unphysical for values of ω_M that are consistent with the EDA. However, if we choose $\alpha = \omega_m/(\omega_m+\omega_k)$ [cf. Eq. (144)], then we obtain the Hamiltonian [cf. Eq. (147)]

$$H_{\rm JC} = \tilde{\omega}_m\left(d^\dagger d + \frac{1}{2}\right) + \sum_{k,j}\omega_{kj}\left(c_k^\dagger c_j + \frac{\delta_{kj}}{2}\right) - iq\sum_k\sqrt{\frac{\omega_k\omega_m}{mv}}\frac{1}{\omega_m+\omega_k}(d^\dagger c_k - d c_k^\dagger). \tag{182}$$

In this gauge the ground state is represented by the vector $|0_d, 0_c\rangle$ annihilated by d and c_k. It is easy to verify that the ground energy eigenvalue of $H_{\rm JC}$ produces the expected order q^2 ground-state Lamb shift (Drummond, 1987). In this gauge the detector excitation rate is identically zero because $u^+(\omega) \equiv 0$.

3. Dressing transformation for an arbitrary multilevel dipole

The idea of the JC gauge can be extended beyond the previously considered simple systems through a systematic approach to defining and understanding the concept of dressing. The task was undertaken relatively early on by van Hove (1955), who understood dressing in terms of the Hamiltonian resolvent $G(z) = 1/(z-H)$, $z \in \mathbb{C}$. Let $L(z)$ be the part of $G(z)$ that is diagonal in the eigenstates of $h = H - V$, and let the subscript i refer to any state represented by an eigenvector of h. We express the eigenvalues of $L(z)$ in the following form:

$$L_i(z) = \frac{1}{z - \omega_i - \Delta_i(z) + (i/2)\Gamma_i(z)}, \tag{183}$$

where $\Delta_i(z)$ and $\Gamma_i(z)$ are real. One can characterize states in terms of these quantities (van Hove, 1955; Davidovich and Nussenzveig, 1980; Cohen-Tannoudji, Dupont-Roc, and Grynberg, 2010).

Following Davidovich and Nussenzveig (1980), we assume that the equation $\omega - \omega_i - \Delta_i(\omega) = 0$ has only one real root. There are then three further possible cases.

(1) $\Gamma_i(\omega_i) \neq 0$.
(2) $\Gamma_i(\omega) = 0 \ \ \forall \ \ \omega \in \mathbb{R}$.
(3) $\Gamma_i(\omega_i) = 0$, but $\Gamma_i(\omega) \neq 0$ for some $\omega \in \mathbb{R}$.

In case (1), ω_i lies on a cut of $L_i(z)$, and the state i is said to be *dissipative* because it will typically decay in the presence of the interaction. The quantities $\Gamma_i(\omega)$ and $\Delta_i(\omega)$ are the associated linewidth and level shift, respectively; see Sec. VI.B. An example is the state represented by the eigenvector $|\epsilon^e, 0\rangle$ of h in Eq. (148), in which the dipole has been excited to a level e and there are no photons (this state will be considered as an initial state in Sec. VI.B). In case (2), $L_i(\omega)$ has a simple pole at ω_i. The state i is said to be *asymptotically stationary* because asymptotically it is unaffected by the interaction V. The interaction produces only transient effects, and the S-matrix elements between such states are given by Eq. (135) without the need to invoke the condition of adiabatic interaction switching. Case (3) lies between cases (1) and (2). The state i is not asymptotically stationary, but it is also distinguished from a dissipative state. In this case the interaction is said to give rise to *persistent perturbation effects*. Physically, this can be thought of as dressing by a virtual "cloud" of quanta. The ground state of h represented by the vector $|\epsilon^0, 0\rangle$ is an example.

The JC gauge can be defined as a representation in which the ground state of H is represented by the ground eigenvector of h, removing the effects of persistent perturbations. Physically, this means absorbing virtual dressing excitations, such that subsystem excitations are defined relative to the true ground state of the composite. To show how such a representation can be derived systematically, Davidovich and Nussenzveig (1980) let $H = h + qV$ and $H' = e^{iS}[h + qV]e^{-iS} = h + qV'$, where q is a small parameter. Writing $S = \sum_{n=1}^{\infty} q^n S_n$ and $V' = \sum_{n=1}^{\infty} q^n V'_n$ and equating coefficients in powers of q, we have

$$V'_1 = V + i[S, h], \tag{184}$$

$$V'_2 = i[S_1, V_1] + i[S_2, h] - \tfrac{1}{2}[S_1, [S_1, h]],$$
$$V'_3 = \cdots. \tag{185}$$

One now chooses S such that in V' the component of V that is responsible for persistent perturbation effects is canceled out to the required order in q.

We consider the example of the dipole-field Hamiltonian H_α in Eq. (148). To illustrate the procedure, we eliminate the cause of persistent perturbations up to first order in q, which is often sufficient for applications within the weak-coupling regime. We begin in the Coulomb gauge $\alpha = 0$. The order q part of the interaction Hamiltonian can be partitioned into rotating-wave and counterrotating parts as $V_1^0 = V_{1,\mathrm{rot}}^0 + V_{1,\mathrm{counter}}^0$, where $V_{1,\mathrm{rot}}^0 \coloneqq \sum_{n>p}^{n,p} r_{np} + \mathrm{H.c.}$ and $V_{1,\mathrm{counter}}^0 \coloneqq \sum_{n<p}^{n,p} r_{np} + \mathrm{H.c.}$, with $r_{np} \coloneqq -\sum_{\mathbf{k}\lambda} i\omega_{np} g_k [\mathbf{d}_{np} \cdot \mathbf{e}_{\mathbf{k}\lambda}] a_{\mathbf{k}\lambda} |\epsilon^n\rangle\langle\epsilon^p|$. Here H.c. stands for Hermitian conjugate. The term $V_{1,\mathrm{rot}}^0$ satisfies $V_{1,\mathrm{rot}}^0 |\epsilon^0, 0\rangle = 0$, whereas the term $V_{1,\mathrm{counter}}^0$ is responsible for persistent perturbation effects. We define the generalized gauge-fixing transformation as (Stokes, 2013)

$$R_{0\{\alpha\}} \coloneqq e^{iS_{\{\alpha\}}}, \tag{186}$$

$$S_{\{\alpha\}} \coloneqq -\sum_{\mathbf{k}\lambda} \sum_{n,p} g_k (\mathbf{e}_{\mathbf{k}\lambda} \cdot \mathbf{d}_{np}) \alpha_{k,np} |\epsilon^n\rangle\langle\epsilon^p| (a_{\mathbf{k}\lambda}^\dagger + a_{\mathbf{k}\lambda}), \tag{187}$$

which reduces to $R_{0\alpha}$ if $\alpha_{k,np} = \alpha$. It is easily verified that if

$$\alpha_{k,np} = \begin{cases} \frac{\omega_{np}}{\omega_{np} + \omega_k}, & n > p \\ \frac{\omega_{np}}{\omega_{np} - \omega_k}, & n < p \end{cases} = \frac{|\omega_{np}|}{|\omega_{np}| + \omega_k}, \tag{188}$$

then the interaction within the transformed representation $V^{\{\alpha\}} = R_{0\{\alpha\}} H_0 R_{0\{\alpha\}}^\dagger - h$ satisfies $V_1^{\{\alpha\}} |\epsilon^0, 0\rangle = 0$ and thus contains no persistent perturbation contributions of first order in q (Stokes, 2013).

This choice of $\alpha_{k,np}$ generalizes the JC gauge defined by Eq. (144), which applies to a two-level or harmonic dipole. Bear in mind, however, that in general, i.e., for an arbitrary anharmonic multilevel dipole, the choice in Eq. (188) results in the sought after cancellation only up to first order in q. The resulting interaction Hamiltonian correct to first order in q is (Stokes, 2013)

$$V_1^{\{\alpha_{\mathrm{JC}}\}} = -i \sum_{\mathbf{k}\lambda} \sum_{\substack{n,p \\ n>p}} \sqrt{\frac{\omega_k}{2v}} \frac{2\omega_{np}}{\omega_{np} + \omega_k} \mathbf{d}_{np} \cdot \mathbf{e}_{\mathbf{k}\lambda} |\epsilon^n\rangle\langle\epsilon^p| a_{\mathbf{k}\lambda} + \mathrm{H.c.}, \tag{189}$$

which allows photon annihilation (creation) if and only if the dipole transitions to a higher (lower) level. In particular, $H_{\{\alpha\}} = h + V_1^{\{\alpha\}}$ possesses the same ground state as h and the model possesses additional symmetry that allows the Hilbert space to be split into sectors. These are the prototypical properties of the JC model.

B. Natural line shape

We now discuss the natural line shape of spontaneous emission, which can be calculated using techniques similar to those reviewed previously. Lamb noted in 1952 that two different expressions can be obtained for the natural line shape, depending on whether Coulomb-gauge coupling or dipolar coupling is assumed. The prediction is a simple example of an experimentally testable signature of subsystem gauge relativity.

1. Gauge relativity of the prediction

Excited atoms decay via spontaneous emission. The line shape is defined as the frequency distribution (spectrum) of the emitted photons. Let the initial state of a dipole-field system be represented by $|\epsilon^e, 0\rangle$, where $|\epsilon^e\rangle$ represents an excited dipolar level and $|0\rangle$ denotes the photonic vacuum. The average number of photons $\mathbf{k}\lambda$ at time t is given by

$$N_\alpha(\mathbf{k}\lambda, t) = \langle a_{\mathbf{k}\lambda}^\dagger(t) a_{\mathbf{k}\lambda}(t) \rangle_{e0} = \sum_m \sum_{n_{\mathbf{k}\lambda}} n_{\mathbf{k}\lambda} |\langle \epsilon^m, n_{\mathbf{k}\lambda} | U_\alpha(t, 0) | \epsilon^e, 0 \rangle|^2, \tag{190}$$

where $|n_{\mathbf{k}\lambda}\rangle$ denotes the $n_{\mathbf{k}\lambda}$-photon Fock state. For each different α, the quantity $N_\alpha(\mathbf{k}\lambda, t)$ is gauge invariant and gauge relative; cf. Secs. III.C and V.D. The distinction between gauge invariance and gauge relativity [Eqs. (132) and (133)] is important, but there appears to have been a lack of recognition of this distinction in the literature on the natural line shape, as further discussed in Sec. VI.B.3.

The line shape may be defined in the mode continuum limit $\omega_k \to \omega$ by

$$S_\alpha(\omega) = \frac{v}{(2\pi)^3} \rho(\omega) \int d\Omega \sum_\lambda \lim_{t\to\infty} N_\alpha(\mathbf{k}\lambda, t), \tag{191}$$

where $\omega = |\mathbf{k}|$ and $\rho(\omega)$ is the density of modes [$\rho(\omega) = \omega^2$ in free space], the summation is over the polarizations $\lambda = 1, 2$, and the integration is over all directions for $\mathbf{k}$. We have assumed photonic modes confined to a volume v.

Power and Thirunamachandran (1999a) calculated Coulomb- and multipolar-gauge photon number averages

and atomic populations up to second order in the dipole moment, such that the gauge relativity of these quantities can be seen explicitly; see also Sec. VI.A for a discussion of the case of atomic populations. Damping is then described by adding explicit exponential temporal decay of the dipole moment operator, such that the gauge relativity of the spectrum is also confirmed through the attainment of different results for the $\alpha = 0$ and 1 cases. Power and Thirunamachandran (1999a) remarked that in principle such differences should be possible to test experimentally.

The multipolar prediction, which when one ignores details of the dipole's excitation is $S_1(\omega)$, appeared to be in better agreement with the experiments of Lamb (1952) than the Coulomb-gauge prediction (Power and Zienau, 1959; Fried, 1973; Milonni, Cook, and Ackerhalt, 1989; Woolley, 2000; Stokes, 2013). Power and Zienau explained this using what is now known as the PZW transformation to remove "static precursor" contributions that occur in the Coulomb gauge (Power and Zienau, 1959). In other words, passage to the multipolar gauge removes the electrostatic field that is implicit in the definition of the Coulomb-gauge dipole [cf. Secs. II.E and VI.C.1], which Power and Zienau deemed to be unphysical, at least within the context of the natural line shape prediction. This amounts to the stipulation that the multipolar subsystems are more operationally relevant than the corresponding Coulomb-gauge ones, as appears to have been borne out by the experiments.

The prediction $S_1(\omega)$ is gauge invariant and can therefore be calculated in any gauge; cf. Eq. (132). Milonni, Cook, and Ackerhalt (1989) provided a derivation of the line shape $S_1(\omega)$ using the Coulomb gauge. This works by neglecting the difference between the source components of the Coulomb-gauge and multipolar-gauge photonic operators as follows. For a dipole at the origin, the integrated equation of motion for the α-gauge annihilation operator is found using Eq. (148) and possesses the source term

$$\begin{aligned} a_{\mathbf{k}\lambda,\mathrm{s}}(t) &= i\int_0^t dt' \frac{e^{-i\omega_k(t-t')}}{\sqrt{2\omega_k v}}\mathbf{e}_{\mathbf{k}\lambda}\cdot[i(1-\alpha)\dot{\mathbf{d}}(t') + \alpha\omega_k \mathbf{d}(t')] \\ &= \frac{i(1-\alpha)}{\sqrt{2\omega_k v}}\mathbf{e}_{\mathbf{k}\lambda}\cdot[\mathbf{d}(0)e^{-i\omega_k t} - \mathbf{d}(t)] \\ &\quad + \int_0^t dt' \sqrt{\frac{\omega_k}{2v}}\mathbf{e}_{\mathbf{k}\lambda}\cdot\mathbf{d}(t')e^{-i\omega_k(t-t')}, \end{aligned} \tag{192}$$

where the second equality follows from an integration by parts. Neglecting the boundary term $\sim\mathbf{d}(0)e^{-i\omega_k t} - \mathbf{d}(t)$ gives the source part of the integrated equation of motion for the multipolar-gauge annihilation operator, which in turn yields the spectrum $S_1(\omega)$. Milonni, Cook, and Ackerhalt (1989) argued that ignoring this term can be justified based on a sensible choice of boundary conditions. Specifically, exponential decay implies that the contribution from $\mathbf{d}(t)$ will vanish in the longtime limit $t \gg 1/\Gamma_e, \Gamma_e = \sum_{m<e}\Gamma_{em}$, while the term depending on $\mathbf{d}(0)$ may be set to zero provided that the motion of the bare mechanical dipole (as defined relative to the multipolar gauge) is assumed to start after $t = 0$. This is another way to understand the procedure of removing static precursor contributions found in the Coulomb gauge, but more generally the argument can be applied for any $\alpha \neq 1$. Equivalently, it can be understood as a method of implementing the sudden switch on of the multipolar-gauge interaction within the α gauge. Again, these arguments essentially amount to the submission that the subsystems defined relative to the multipolar gauge are more physically relevant.

2. Radiation damping

There are different methods available to move beyond a second-order phenomenological calculation. These include Hamiltonian resolvent and projection operator techniques (Davidovich and Nussenzveig, 1980; Cohen-Tannoudji, Dupont-Roc, and Grynberg, 2010). An exact derivation of the line shape is found using the formal theory of radiation damping, which goes back to the early work of Heitler (2003). Details are given in note XV of the Supplemental Material (245). To calculate the line shape one assumes a dipole initially in an excited state with no photons present and then calculates the longtime probability $[|b_{n\mathbf{k}\lambda,e0}(\infty)|^2]$ that a transition has occurred into a state with the dipole in level n and with one photon $\mathbf{k}\lambda$ present. The frequency spectrum is defined by

$$S(\omega) = \frac{v}{(2\pi)^3}\omega^2\int d\Omega\sum_\lambda \lim_{t\to\infty}|b_{n\mathbf{k}\lambda,e0}(t)|^2. \tag{193}$$

Note that one could assume that the dipole is excited adiabatically starting in the distant past, and that the interaction is switched off adiabatically such that $b_{n\mathbf{k}\lambda,e0}(\infty)$ becomes an S-matrix element and therefore is no longer gauge relative; see Sec. V.A and the discussion in Sec. VI.B.3. This assumption may or may not be realistic when one models an experiment.

For a Hamiltonian $H = h + V$, the longtime probability for the transition from an initial ($t = 0$) state i represented by an eigenvector of h to a final state f represented by a different eigenvector of h is given by

$$|b_f(\infty)|^2 = \frac{|R_{fi}(\omega_f)|^2}{[\omega_{fi} - \Delta_i(\omega_f)]^2 + [\Gamma_i(\omega_f)/2]^2}, \tag{194}$$

where

$$\Gamma_i(\omega) = 2\pi\sum_{m\neq i}|R_{mi}(\omega)|^2\delta(\omega-\omega_m), \tag{195}$$

$$\Delta_i(\omega) = V_{ii} + \mathcal{P}\sum_{m\neq i}\frac{|R_{mi}(\omega_m)|^2}{\omega-\omega_m}, \tag{196}$$

and

$$R_{ni}(\omega) = V_{ni} + \sum_{m\neq i}V_{nm}R_{mi}(\omega)\zeta(\omega-\omega_m), \quad n\neq i, \tag{197}$$

in which

$$\zeta(x) \coloneqq \mathcal{P}\frac{1}{x} - i\pi\delta(x). \tag{198}$$

Note that $b_f(\infty)$ can be written as $R_{fi}(\omega_f)L_i(\omega_f)$, where $L_i(z)$ is as defined in Eq. (183).

Now consider the case in which $V = V^\alpha = V_1^\alpha + V_2^\alpha$ is the α-gauge interaction Hamiltonian for a dipole-field system in Eq. (148), while $|i\rangle = |\epsilon^e, 0\rangle$ and $|f\rangle = |\epsilon^n, \mathbf{k}\lambda\rangle$. The matrix elements $R_{mi}(\omega_m)$ are in general gauge relative (α dependent), as is $|b_f(\infty)|^2$. This gauge relativity can, however, be eliminated by invoking gauge nonrelativistic approximations. Specifically, if bare-energy conservation $\omega_f = \omega_i$ is imposed (from outside the theory), then the quantities in Eq. (194) are evaluated at ω_i (on energy shell) and are then α independent (Woolley, 2000). This exemplifies the general result discussed in Sec. V.A, namely, that strict bare-energy conservation is required to eliminate subsystem gauge relativity within probability amplitudes connecting bare states.

To make contact with the Markovian approximations used in Sec. VI.A, we now look more closely at the quantities in Eq. (194). To lowest order in V, Eq. (197) gives $R_{mi}(\omega) = V_{mi}$, which we can use to find $\Gamma_i(\omega_f)$ and $\Delta_i(f)$ as

$$\Gamma_i(\omega_f) = 2\pi \sum_m \sum_{\mathbf{k}'\lambda'} |[V_1^\alpha]_{m\mathbf{k}'\lambda',e0}|^2 \delta(\omega_k + \omega_{nm} - \omega_{k'}), \quad (199)$$

$$\Delta_i(\omega_f) = [V_2^\alpha]_{e0,e0} + \mathcal{P} \sum_m \sum_{\mathbf{k}'\lambda'} \frac{|[V_1^\alpha]_{m\mathbf{k}'\lambda',e0}|^2}{\omega_k + \omega_{nm} - \omega_{k'}}. \quad (200)$$

If we evaluate $[V_1^\alpha]_{m\mathbf{k}\lambda,e0}$ at $\omega_f = \omega_i$, that is, at $\omega_k = \omega_{en}$, then $\Gamma_i(\omega_f) = \Gamma_e$ and $\Delta_i(\omega_f) = \Delta_e$, where $\Gamma_e = \sum_{m<e} \Gamma_{em}$ and Δ_e are the total spontaneous emission rate and on-energy-shell Lamb shift associated with the dipole level e, as calculated using Fermi's golden rule and second-order perturbation theory, respectively. Both quantities are α independent.

An on-energy-shell evaluation may be justified within the quantity $L_i(\omega)$ defined in Eq. (183) and is known as the pole approximation, which is commonly employed in the calculation of Lorentzian spectra (Barnett, Radmore, and Barnett, 1997). Specifically, it is justified in the Markovian regime ($\Gamma_{em} t \gg 1$) provided that $\Gamma_i(\omega)$ and $\Delta_i(\omega)$ are sufficiently slowly varying near ω_i because then $L_i(\omega)$ has a pole near ω_i such that it may with sufficient accuracy be approximated by $L_i(\omega_i + i\eta)$ with $\eta \to 0^+$. In a similar fashion, Markovian approximations were used in Sec. VI.A to derive the gauge-nonrelative rate Γ from either of the gauge-relative expressions (170) or (171). Applying the pole approximation to $L_i(\omega_f)$ within $b_f(\infty) = R_{fi}(\omega_f)L_i(\omega_f)$ yields the final result

$$S_\alpha(\omega) = \frac{\Gamma_{en}}{2\pi} \frac{(\omega/\omega_{en}^3)[(1-\alpha)\omega_{en} + \alpha\omega]^2}{(\omega - \tilde{\omega}_{en})^2 + (\Gamma_e/2)^2}, \quad (201)$$

where $\tilde{\omega}_{en} = \omega_{en} + \Delta_e$. Further evaluating the numerator on energy shell implies that all remaining α dependence drops out and we obtain the pure Lorentzian

$$S(\omega) = \frac{\Gamma_{en}}{2\pi} \frac{1}{(\omega - \tilde{\omega}_{en})^2 + (\Gamma_{en}/2)^2}. \quad (202)$$

Note that away from resonance ($\omega = \omega_{en}$) this is significantly different from the line shape $S_1(\omega)$, but it should also be noted that a description of the dipole's excitation has not been included. This is important when describing the Lamb experiments. Nevertheless, even if an on-energy-shell evaluation of the line shape denominator can be justified, the same procedure applied to the numerator may be difficult to justify because the numerator may not be sufficiently slowly varying compared with the denominator.

3. The $2s_{1/2} \to 1s_{1/2}$ transition in hydrogen

We now more closely consider the experiments of Lamb (1952), which probed the $2s_{1/2} \to 1s_{1/2}$ transition in hydrogen. The atoms start in the metastable state $2s_{1/2}$. They are irradiated with a microwave frequency ω close to the frequency ω_0 of the $2s_{1/2} \to 2p_{1/2}$ transition. The microwave resonance $\omega - \omega_0$ is detected by photons spontaneously emitted in the $2p_{1/2} \to 1s_{1/2}$ transition.

A main goal of previous studies (Fried, 1973; Bassani, Forney, and Quattropani, 1977; Davidovich and Nussenzveig, 1980; Cohen-Tannoudji, Dupont-Roc, and Grynberg, 1989; Woolley, 2000) was to eliminate the α dependence of the line shape prediction, which has been viewed as a paradoxical property (gauge noninvariance). However, we have provided a precise mathematical definition of gauge invariance [Eq. (70)] according to which the α dependence of the line shape prediction does not constitute gauge noninvariance. Equation (194), in particular, is exact, but for each different value of α the labels i and f therein refer to different physical states. The result is therefore gauge relative, which is not paradoxical and is simply an example of the expected inequality (133). Each of the predictions is certainly gauge invariant in the sense of Eq. (132).

While the prediction is fundamentally gauge relative, use of the S matrix will circumvent this relativity; see Sec. V.A. However, subsequent approximations of the S matrix may in turn eliminate this special property. We must distinguish the simplifying assumptions that eliminate α dependence by defining the S matrix from subsequent approximations of the S matrix that may then eliminate its α independence. Previous studies have identified which approximations of relevant S-matrix elements must be avoided in order for them to be α independent (gauge nonrelative). In particular, full sets of intermediate states must be retained in calculations (Bassani, Forney, and Quattropani, 1977; Cohen-Tannoudji, Dupont-Roc, and Grynberg, 1989), despite the apparent dominance of the intermediate state $2p_{1/2}$ (Power and Zienau, 1959). As we saw in Sec. IV, the significance of higher dipole levels is greater in the Coulomb gauge, such that summation over these levels converges much more quickly in the multipolar gauge. In the context of calculating two-photon transition matrix elements, this has been understood for some time (Bassani, Forney, and Quattropani, 1977; Cohen-Tannoudji, Dupont-Roc, and Grynberg, 1989).

The S matrix is perturbative, essentially by definition [the T matrix in Eq. (135) is expanded iteratively in powers of V], making it difficult to obtain an expression for the line shape with a finite width corresponding to exponential decay. However, damping can be included by alteration of the two-photon on-energy-shell (Kramers-Heisenberg) transition

matrix element describing the process (Power and Zienau, 1959; Fried, 1973; Davidovich and Nussenzveig, 1980; Stokes, 2013). Fried showed using a semiclassical treatment (Fried, 1973) that when damping is included in this way, and "nonresonant background" terms that are present within the modified matrix element are not ignored, the Coulomb-gauge and multipolar-gauge predictions can be brought into significantly closer agreement. The situation in which excitation occurs via a tunable microwave field and an alternative situation of a fixed microwave field with an applied magnetic field are both considered. This method was extended to a full quantum treatment by Davidovich and Nussenzveig (1980).

Nevertheless, if an exact radiation damping treatment is adopted [Eq. (194)], then the prediction will be α dependent (gauge relative). As a somewhat extreme position, one might reject outright the validity of treating eigenvectors of h as physically meaningful outside of scattering theory. Either one would be confined to the use of scattering theory or alternative physical states would have to be identified. Over finite times without adiabatic switching, the eigenvectors of $H_\alpha = h + V^\alpha$ uniquely represent fully dressed physical states. However, these states are stationary, such that a rejection of initial and final states that are not eigenstates of H appears to preclude the possibility of studying nontrivial dynamics. One could instead consider the ground state of H_α as the initial state that is then subjected to a time-dependent external perturbation, but in this case a microscopic description would entail identifying a gauge relative to which the time-dependent interaction is to be defined, such that the prediction again becomes gauge relative; cf. Sec. V.D.

A criterion by which the most meaningful physical states can be identified was suggested in Sec. VI. Specifically, one may attempt to define physical light and matter excitations relative to the ground state of H_α. The virtual admixtures otherwise present in the ground state have therefore been absorbed into the subsystem definitions. This "JC gauge" for an arbitrary multilevel dipole was discussed in Sec. VI.A.3 and was discussed in the context of the line shape by Davidovich and Nussenzveig (1980) and Stokes (2013). Stokes (2013) adopted a radiation damping treatment under the assumption that excitation of the dipole to the state $2s_{1/2}$ occurs through absorption of photons with a spectrum much sharper than the emitted spectrum. Excitation via continuous laser irradiation prior to emission is also considered. The fluorescence rates found using different gauges have Lorentzian forms with $L_i(\omega_f)$ evaluated on energy shell, but with differing numerators. The associated line shapes are compared for the Coulomb gauge, multipolar gauge, and JC gauge. The multipolar prediction is closest to a bare Lorentzian curve. As expected, the JC gauge curve interpolates between this curve and the Coulomb-gauge result. Differences are increasingly conspicuous farther away from resonance ($\omega_f = \omega_i$). We note finally that in the weak-coupling regime differences between gauge-relative predictions such as emission line shapes will typically be small. Spectroscopic experimental signatures outside of this regime are discussed in Sec. VII.D.

C. Localization and causality

1. Electromagnetic source fields in an arbitrary gauge

To understand the balance between localization and dressing, it is necessary to determine the electromagnetic fields generated by a source in an arbitrary gauge. In particular, if we consider a system consisting of both a source s and a detector d, then the total electric field is a superposition of the vacuum, source, and detector fields,

$$\mathbf{E} = \mathbf{E}_{\mathrm{vac}} + \mathbf{E}_{\mathrm{s}} + \mathbf{E}_{\mathrm{d}}. \tag{203}$$

A full description of the source-detector-field system is given in Sec. VI.C.2. We first note that due to subsystem gauge relativity the partitioning of a gauge-invariant field into vacuum, source, and detector components is gauge relative (Power and Thirunamachandran, 1999a, 1999b). In other words, while the left-hand side of Eq. (203) is unique, the individual components on the right-hand side represent different physical fields in different gauges. We therefore start by considering only one material system: a point dipole fixed at $\mathbf{0}$ and with dipole moment $q\mathbf{r}$. For simplicity, we again restrict our attention to the one-parameter α-gauge framework.

We now consider the canonical field $\mathbf{\Pi}$ at an arbitrary point $\mathbf{x} \neq \mathbf{0}$, which can be partitioned as

$$\begin{aligned}\mathbf{\Pi}(t,\mathbf{x}) &= -\mathbf{E}_{\mathrm{T}}(t,\mathbf{x}) - \alpha\mathbf{E}_{\mathrm{L}}(t,\mathbf{x}) \\ &= \mathbf{\Pi}^\alpha_{\mathrm{vac}}(t,\mathbf{x}) + \mathbf{\Pi}^\alpha_{\mathrm{s}}(t,\mathbf{x}).\end{aligned} \tag{204}$$

In the gauge α the vacuum and source components $\mathbf{\Pi}^\alpha_{\mathrm{vac}}(t,\mathbf{x})$ and $\mathbf{\Pi}^\alpha_{\mathrm{s}}(t,\mathbf{x})$ are defined as the components whose dynamics are generated by H_{ph} and V^α, respectively. The vacuum field is defined by the right-hand side of Eq. (160). Since the photons defined by $a_\lambda(0,\mathbf{k})$ are physically distinct for each α, the vacuum field depends on α. The source field also depends on α, and the dynamics generated by $H_\alpha = H_m + H_{\mathrm{ph}} + V^\alpha$ yields

$$\begin{aligned}\mathbf{\Pi}^\alpha_{\mathrm{s}}(t,\mathbf{x}) &= -\theta(t_r)\mathbf{X}_{\mathrm{T}}(t_r,\mathbf{x}) \\ &\quad + (1-\alpha)[\mathbf{P}_{\mathrm{T}}(t,\mathbf{x}) - \theta(-t_r)\mathbf{P}_{\mathrm{T}}(0,\mathbf{x})],\end{aligned} \tag{205}$$

where $t_r = t - x$ is the retarded time (in units with $c = 1$) in which $x = |\mathbf{x}|$ is the distance from the dipole source at $\mathbf{0}$ and where, for $\mathbf{x} \neq \mathbf{0}$,

$$X_{\mathrm{T},i}(t,\mathbf{x}) = (-\partial^2\delta_{ij} + \partial_i\partial_j)\frac{qr_j(t_r)}{4\pi x}. \tag{206}$$

Note that the derivative operators in Eq. (206) act on t_r as well as on $1/x$. Only the top line on the right-hand side of Eq. (205) is causal, by which we mean vanishing for $t_r < 0$, and the second line vanishes only for $\alpha = 1$.

Using the fact that the $a_\lambda(0,\mathbf{k})$ of different gauges are related by $R_{\alpha\alpha'}$, one finds that the different vacuum components $\mathbf{\Pi}^\alpha_{\mathrm{vac}}$ are related by

$$\mathbf{\Pi}^\alpha_{\mathrm{vac}}(t,\mathbf{x}) = \mathbf{\Pi}^{\alpha'}_{\mathrm{vac}}(t,\mathbf{x}) - (\alpha-\alpha')\theta(-t_r)\mathbf{P}_{\mathrm{T}}(0,\mathbf{x}). \tag{207}$$

It follows that the combination $\mathbf{\Pi}^{\alpha}_{\mathrm{vac}}(t,\mathbf{x})+\alpha\theta(-t_r)\mathbf{P}_{\mathrm{T}}(0,\mathbf{x})$ is actually α independent. We see also that for different α the vacuum components $\mathbf{\Pi}^{\alpha}_{\mathrm{vac}}$ differ by an $\alpha-\alpha'$ weighted factor of $\mathbf{P}_{\mathrm{T}}=\mathbf{E}_{\mathrm{L}}$ evaluated at $t=0$, and that this contribution is restricted to the complement of the interior light cone of the origin $(0,\mathbf{0})$ of the dipole's rest frame.

It is instructive to consider some specific physical fields. For example, $\mathbf{E}_{\mathrm{T}}=-\mathbf{\Pi}|_{\alpha=0}$, for which $\mathbf{E}^{\alpha}_{\mathrm{T,s}}=-\mathbf{\Pi}^{\alpha}_{\mathrm{s}}-\alpha\mathbf{P}_{\mathrm{T}}$ and $\mathbf{E}^{\alpha}_{\mathrm{T,vac}}=-\mathbf{\Pi}^{\alpha}_{\mathrm{vac}}$. Although the free and source components are different in different gauges, their sum is

$$\mathbf{E}_{\mathrm{T}}(t,\mathbf{x})=\theta(t_r)\mathbf{X}_{\mathrm{T}}(t,\mathbf{x})+\theta(-t_r)\mathbf{P}_{\mathrm{T}}(0,\mathbf{x})-\mathbf{P}_{\mathrm{T}}(t,\mathbf{x})-\mathbf{\Pi}^{\alpha}_{\mathrm{vac}}(t,\mathbf{x})-\alpha\theta(-t_r)\mathbf{P}_{\mathrm{T}}(0,\mathbf{x}),\tag{208}$$

which upon taking into account Eq. (207) is seen to be unique (α independent), as required. The total electric field is for $\mathbf{x}\neq\mathbf{0}$ given by $\mathbf{E}=\mathbf{D}_{\mathrm{T}}=-\mathbf{\Pi}|_{\alpha=1}=\mathbf{E}_{\mathrm{T}}+\mathbf{P}_{\mathrm{T}}$, which can be read off immediately from Eq. (208) as

$$\mathbf{E}(t,\mathbf{x})=\mathbf{D}_{\mathrm{T}}(t,\mathbf{x})=\theta(t_r)\mathbf{X}_{\mathrm{T}}(t,\mathbf{x})+\theta(-t_r)\mathbf{P}_{\mathrm{T}}(0,\mathbf{x})-\mathbf{\Pi}^{\alpha}_{\mathrm{vac}}(t,\mathbf{x})-\alpha\theta(-t_r)\mathbf{P}_{\mathrm{T}}(0,\mathbf{x}).\tag{209}$$

As in Sec. II.E, the previous results demonstrate that what differs for different choices of α is the localization properties of the source. For $t_r>0$, we have $\mathbf{E}_{\mathrm{s}}(t,\mathbf{x})=\mathbf{D}_{\mathrm{T,s}}(t,\mathbf{x})=\mathbf{X}_{\mathrm{T}}(t,\mathbf{x})$ and $\mathbf{E}_{\mathrm{T,s}}(t,\mathbf{x})=\mathbf{X}_{\mathrm{T}}(t,\mathbf{x})-\mathbf{P}_{\mathrm{T}}(t,\mathbf{x})$ for all α. In words, at all points $\mathbf{x}$ that can be connected to the source's center by a light signal emitted a time x earlier, each physical field's source component is independent of the source's definition. In contrast, for $t_r<0$ the source-vacuum partitioning of a given physical field differs among different gauges α.

As explained in Sec. II.E, within the EDA the gauge controls the extent to which the instantaneous field $\mathbf{E}_{\mathrm{L}}(\mathbf{x})=\mathbf{P}_{\mathrm{T}}(\mathbf{x})$ (where $\mathbf{x}\neq\mathbf{0}$) is included within the source's definition. The gauges $\alpha=0$ and $\alpha=1$ are extremal cases where $\mathbf{E}_{\mathrm{L}}$ is fully included and completely absent, respectively. For this reason, the source component of the field $\mathbf{\Pi}=-\mathbf{E}_{\mathrm{T}}-\alpha'\mathbf{P}_{\mathrm{T}}$, when partitioned according to the gauge α to give $\mathbf{\Pi}^{\alpha}_{\mathrm{s}}=-\mathbf{E}^{\alpha}_{\mathrm{T,s}}-\alpha'\mathbf{P}_{\mathrm{T}}$, is causal (meaning vanishing for $t_r<0$) if and only if $\alpha=1$ and $\alpha'=1$. The latter equality specifies that the physical field under consideration is $\mathbf{E}$, which is a local field, and the former equality ($\alpha=1$) specifies that the source producing this field is defined relative to the multipolar gauge, and is therefore itself also local. It is easy to show that, unlike $\mathbf{E}$, the magnetic field $\mathbf{B}=\mathbf{B}_{\mathrm{vac}}+\mathbf{B}_{\mathrm{s}}$ has unique vacuum and source components and that $\mathbf{B}_{\mathrm{s}}$ is causal (Power and Thirunamachandran, 1999a).

These results generalize those of Power and Thirunamachandran (1999a) by giving vacuum-source partitions of the physically arbitrary field $\mathbf{\Pi}$, using an arbitrary gauge α. For any given physical field the relative magnitude of the nonlocal contributions occurring for $t_r<0$ vary with α and provide a measure of the delocalization of the source, as elaborated on later and in note XIII of the Supplemental Material (245).

2. Source-detector-field system

We now consider the tripartite source-detector-field system. If we require the detector dipole to be fully localized at $\mathbf{x}$ and a source dipole to be fully localized at $\mathbf{0}$, then matter must be defined relative to the multipolar gauge. From the results of Sec. VI.C.1 it is also clear that the response of the detector$_1$ to the source$_1$ is causal, as required (Cohen-Tannoudji, Dupont-Roc, and Grynberg, 1989; Biswas *et al.*, 1990; Milonni, James, and Fearn, 1995; Power and Thirunamachandran, 1997, 1999a, 1999b; Sabín *et al.*, 2011). In any other gauge where $\alpha\neq1$, matter is dressed by $\alpha\mathbf{E}_{\mathrm{L}}$ and thus is not fully localized. However, questions regarding the causal nature of an interaction are well posed only for separated localized objects. The instantaneous response of a delocalized detector to a delocalized source will vary with α and can be taken as a measure of the overlap of the source and detector, and hence as a measure of the delocalization of matter as defined within the gauge α.

To make these statements concrete, we consider a system of two identical dipoles labeled s (source) and d (detector) at positions $\mathbf{R}_{\mathrm{s}}=\mathbf{0}$ and $\mathbf{R}_{\mathrm{d}}$, respectively. To quantify the response of the detector to the source it suffices to consider the rate of change of the detector's energy. Excitation probabilities such as those considered in Sec. VI.A are determined from the spectral projections of the detector's energy. The multipolar Hamiltonian can be partitioned as

$$H_1=\tilde{H}_{\mathrm{d}}+\tilde{H}_{\mathrm{s}}+\tilde{V}_{\mathrm{d}}+\tilde{V}_{\mathrm{s}}+H_{\mathrm{ph}},\tag{210}$$

where $\mu=\mathrm{s},\mathrm{d}$,

$$\tilde{H}_{\mu}=\frac{\mathbf{p}_{\mu}^2}{2m}+V(\mathbf{r}_{\mu})+S_{\mu},\qquad\tilde{V}_{\mu}=q\mathbf{r}_{\mu}\cdot\mathbf{\Pi}(\mathbf{R}_{\mu}),\tag{211}$$

in which the term $S_{\mu}:=(1/2)\int d^3x\mathbf{P}^2_{\mathrm{T},\mu}$, with $\mathbf{P}_{\mathrm{T},\mu}(\mathbf{x}):=q\mathbf{r}_{\mu}\cdot\delta^{\mathrm{T}}(\mathbf{x}-\mathbf{R}_{\mu})$, has not been placed in the interaction Hamiltonian. The rate of change of $\tilde{H}_{\mathrm{d}}$ is

$$\dot{\tilde{H}}_{\mathrm{d}}(t)=-q\dot{\mathbf{r}}_{\mathrm{d}}(t)\cdot\mathbf{\Pi}(t,\mathbf{R}_{\mathrm{d}})=q\dot{\mathbf{r}}_{\mathrm{d}}(t)\cdot\mathbf{D}_{\mathrm{T}}(t,\mathbf{R}_{\mathrm{d}}).\tag{212}$$

If one instead considers $H_{\mathrm{d}}=\tilde{H}_{\mathrm{d}}-S_{\mathrm{d}}$, then the rate of change includes an additional self-term that depends only on the detector, which does not affect its response to the source. The total displacement field at $\mathbf{R}_{\mathrm{d}}$ can be partitioned as in Eq. (203). We therefore obtain an expression of Poynting's theorem for the detector$_1$ in the presence of the external field $\mathbf{E}^1_{\mathrm{s}}(t,\mathbf{R}_{\mathrm{d}})$. Specifically, the rate at which work is done by $\mathbf{E}^1_{\mathrm{s}}$ on the detector$_1$ in the volume $\mathcal{V}$ is (Jackson, 1998; Griffiths, 2017)

$$\int_{\mathcal{V}}d^3x\mathbf{J}_{\mathrm{d}}(t,\mathbf{x})\cdot\mathbf{E}^1_{\mathrm{s}}(t,\mathbf{x})=q\dot{\mathbf{r}}_{\mathrm{d}}(t)\cdot\mathbf{E}^1_{\mathrm{s}}(t,\mathbf{R}_{\mathrm{d}})=:\dot{\tilde{H}}_{\mathrm{d,s}}(t),\tag{213}$$

where $\mathbf{J}_{\mathrm{d}}(t,\mathbf{x})=q\dot{\mathbf{r}}_{\mathrm{d}}(t)\delta(\mathbf{x}-\mathbf{R}_{\mathrm{d}})$ is the detector current in the EDA. The detector$_1$ response rate $\dot{\tilde{H}}_{\mathrm{d}}(t)$ can be decomposed in its eigenbasis as $\dot{\tilde{H}}_{\mathrm{d}}(t)=\sum_n\epsilon^n_{\mathrm{d}}\dot{\mathsf{P}}^n_{\mathrm{d}}(t)$, where $\mathsf{P}^n_{\mathrm{d}}(t)$ is

the projection onto the nth level at time t. For a two-level detector$_1$, as is typically considered (Fermi, 1932; Biswas *et al.*, 1990; Milonni, James, and Fearn, 1995; Power and Thirunamachandran, 1997; Sabín *et al.*, 2011; Stokes, 2012), the rate of excitation into the excited state $\dot{\mathsf{P}}^1_{\rm d}(t)$ is easily found to be $\dot{\mathsf{P}}^1_{\rm d}(t) = \dot{\tilde{H}}_{\rm d}(t)/\omega_m$, where $\omega_m = \epsilon^1_{\rm d} - \epsilon^0_{\rm d}$ is the two-level detector$_1$ transition frequency. The source-dependent component is therefore $\dot{\mathsf{P}}^1_{\rm d,s}(t) = \dot{\tilde{H}}_{\rm d,s}(t)/\omega_m$.

3. Discussion on localization and dressing

For fully localized and hence bare multipolar dipoles the detector's response to the source is causal because $\mathbf{E}^1_{\rm s}(t, \mathbf{R}_{\rm d}) = \mathbf{0}$ for $t < t_r$, where $t_r = t - R_{\rm d}$. It follows that each of the spectral projections $\mathsf{P}^n_{\rm d}(t)$ must also depend causally on s, and therefore that the probability of finding the bare detector$_1$ in an excited state causally depends on s. There is also a nonzero component of $\dot{\tilde{H}}_{\rm d}(t)$ that is independent of the source$_1$, namely, $\dot{H}_{\rm d,0}(t) = \dot{\tilde{H}}_{\rm d}(t) - \dot{\tilde{H}}_{\rm d,s}(t)$. In fact, such a contribution must exist if the response of the detector$_1$ to the source$_1$ is to be causal. This follows from Hegerfeldt's theorem, which is a general mathematical result that assumes that (i) the energy is bounded from below, (ii) the source and detector are initially localized in disjoint regions, and (iii) the initial state consists of the source excited and the detector in its ground state with no photons present (Hegerfeldt, 1994). Hegerfeldt showed that under these assumptions the total probability of excitation of the detector $[\mathcal{P}^e_{\rm d}(t) = \mathcal{P}^e_{\rm d,0}(t) + \mathcal{P}^e_{\rm d,s}(t)]$ is necessarily either nonzero for times $t_r < 0$ or identically zero for all times. It follows that, for an initial state represented by the vector $|\epsilon^n_{\rm s}, \epsilon^0_{\rm d}, 0\rangle$ in the multipolar gauge, if $\mathcal{P}^e_{\rm d,0}(t)$ were to vanish, then $\mathcal{P}^e_{\rm d,s}(t)$ would be nonzero for $t_r < 0$, and this would violate Einstein causality because the multipolar-gauge dipoles are localized and spacelike separated.

Hegerfeldt (1994) concluded that the two-atom system (source and detector) engenders a conflict with Einstein causality modulo some ways out that he listed. The claimed violation was contested by Buchholz and Yngvason (1994), Milonni, James, and Fearn (1995), and Power and Thirunamachandran (1997), with the recognition that the possible ways out listed by Hegerfeldt are not mere technicalities and have to be taken seriously. In particular, removing the virtual excitations of a localized material system means absorbing the cloud of virtual particles around it (Buchholz and Yngvason, 1994; Hegerfeldt, 1994) such that states in which there are no such excitations are not ones in which the atoms are strictly localized. States in which the atoms are localized in disjoint regions (and which therefore permit well-posed questions regarding signal propagation) will contain virtual photons (Buchholz and Yngvason, 1994; Milonni, James, and Fearn, 1995).

By assuming the initial state $|\epsilon^n_{\rm s}, \epsilon^0_{\rm d}, 0\rangle$ in the multipolar gauge, one is assuming that the bare multipolar-gauge dipoles are those that are operationally relevant at the preparation stage, but since $|\epsilon^0_{\rm d}, 0\rangle$ is not the ground state of the detector-field system, this leads to the immediate virtual excitation of the detector for $t > 0$. We have seen that this virtual excitation is actually necessary to preserve Einstein causality. However, like a violation of Einstein causality, such virtual spontaneous excitations are themselves conceptually problematic and are essentially what one seeks to eliminate within a successfully renormalized theory. Indeed, we saw in Sec. VI.A that the multipolar dipole's virtual excitation was particularly unphysical and we identified a different gauge within which such excitations were eliminated. In any such theory the detector responds to the source for times $t_r < 0$. To avoid a conflict with Einstein causality, one must interpret the renormalized source and detector as objects that are delocalized around their centers at $\mathbf{0}$ and $\mathbf{R}_{\rm d}$, respectively.

The representation in which virtual excitations are removed is one in which the initial state of the detector and field subsystems coincides with the detector-field ground state, which might be considered a more realistic initial state (see Sec. VI.A.3), but this state is not one that specifies definite energy of a localized detector. Since preparation and measurement procedures necessarily possess finite extents in spacetime, there is a balance to be struck between dressing and localization. The parameter α affects this balance by controlling the extent to which bare matter is dressed by $\mathbf{E}_{\rm L}$, which in turn affects the value of $\dot{\mathcal{P}}_{\rm d,0}$ resulting from the ground-state virtual photons surrounding the bare detector$_1$. It is therefore sensible to conclude that the value of α that specifies the most relevant subsystems will depend on coupling strengths, as well as on the experimental protocols for preparation and measurement, including their spatial and temporal properties.

These questions can essentially be ignored within the traditional quantum optical regime because, as shown in Sec. V.C.2, the reduced description of the detector is independent of the gauge relative to which it is defined and its stationary state is $|\epsilon^0_{\rm d}\rangle$. This is also the regime in which the fields $\mathbf{E}^{1\,(\pm)}_{\rm s}$ are approximately causal (Milonni, James, and Fearn, 1995; Stokes, 2018). Thus, in this regime it is possible to define the detector dipole as a localized system while also retaining a fully causal response to the source, but without spontaneous vacuum excitation. This combination of properties is forbidden by Hegerfeldt's theorem and must therefore be the culmination of weak-coupling approximations. In sufficiently strong-coupling regimes one or more of these properties must be sacrificed. The gauge α relative to which the detector is defined will affect which properties of its weak-coupling counterpart it continues to possess. The multipolar gauge continues to define localized dipoles with causal interactions, but with $\mathcal{P}_{\rm d,0}(t) \not\approx 0$. On the other hand, values $\alpha \neq 1$ define dipoles that are delocalized to some extent, but which may retain the property $\mathcal{P}_{\rm d,0}(t) \approx 0$ even outside of the weak-coupling regime. In note XIII of the Supplemental Material (245), we review concrete demonstrations of this by considering the average electromagnetic energy-momentum in the vicinity of a dipole.

VII. MEASUREMENTS AND CAVITY QED BEYOND WEAK-COUPLING APPROXIMATIONS

We now turn our attention to understanding photonic fields confined to a cavity where weak-coupling theory is generally inapplicable and subsystem gauge relativity is expected to be

important. We first provide a simple but arbitrary-gauge description of the field inside a cavity containing a two-level dipole. This extends the results of Sánchez Muñoz, Nori, and De Liberato (2018), which identify the field bound to the dipole$_1$, as distinguished from the propagating field. The results can also be thought of as a simplified extension of the results for a dipole in free space presented in note XIII of the Supplemental Material (245). An early attempt to relate the dressing of a two-level dipole to weak measurement protocols through the explicit modeling of a pointer system is detailed in note XVI of the Supplemental Material (245). We discuss the topic of ground-state photon condensation in cavity QED systems, which is of considerable current interest but also strongly gauge relative. Finally, we discuss extracavity fields, including a review of simple models describing associated measurement signals.

A. Simple model of intracavity fields

We first consider a simple analysis of intracavity fields produced by a dipole at the cavity center. This closely mirrors the analysis in note XIII of the Supplemental Material (245) for free space. An early step toward evaluating the Glauber intensity within a cavity in the ultrastrong-coupling regime was given by Sánchez Muñoz, Nori, and De Liberato (2018). They placed emphasis upon the need for a multimode theory in accommodating the requisite spatiotemporal structure to elicit signal propagation. We consider a similar analysis in an arbitrary gauge.

We model the cavity as a one-dimensional field in the x direction with periodic boundary conditions at $x=\pm L/2$, where L is the cavity length. The allowed wave numbers are $k=2\pi n/L$, $n\in\mathbb{Z}$. The canonical fields are assumed to point in the z direction and have bosonic mode expansions

$$A(t,\mathbf{x})=\sum_k \frac{1}{\sqrt{2\omega_k v}}[a_k^\dagger(t)e^{-ikx}+a_k(t)e^{ikx}], \tag{214}$$

$$\Pi(t,\mathbf{x})=i\sum_k \sqrt{\frac{\omega_k}{2v}}[a_k^\dagger(t)e^{-ikx}-a_k(t)e^{ikx}], \tag{215}$$

where v is the cavity volume. The cross-sectional area is therefore v/L. As usual, we have $[a_k,a_{k'}^\dagger]=\delta_{kk'}$ and $\omega_k=|k|$. To be consistent with the assumed expressions for $\mathbf{A}_{\mathrm{T}}$ and $\mathbf{\Pi}$, we assume that the transverse polarization $\mathbf{P}_{\mathrm{T}\alpha}$ points in the z direction.

We assume that the dipole within the cavity is sufficiently anharmonic so that we can expect a two-level truncation in the multipolar gauge to generally be robust; see Fig. 6 in Sec. IV.F. Bear in mind that, for a less anharmonic dipole, truncation may remain accurate for predicting the low-energy properties that we consider later, but the optimal gauge for truncation may no longer be the multipolar gauge. It is also important to note that while the procedure of two-level truncation is performed in the multipolar gauge, this does not restrict our attention to subsystems defined relative to the multipolar gauge. It is straightforward to identify the observables that define the α-gauge subsystems within the multipolar gauge, where the truncation of these observables may then be performed. In particular, we are free to consider the canonical field Π defined relative to an arbitrary gauge α. The physical observable represented by the momentum Π in the gauge α is denoted O_α. The notation Π is reserved for the multipolar-gauge canonical momentum $-D_{\mathrm{T}}$; therefore, $O_\alpha=\Pi+P_{\mathrm{T}1}-P_{\mathrm{T}\alpha}$. Here the α-gauge polarization is within the multipolar-gauge truncation given by

$$P_{\mathrm{T}\alpha}(t,x)=\sum_k \frac{d}{v}\sigma^x(t)\alpha\cos[kx], \tag{216}$$

where $d=\hat{\mathbf{z}}\cdot\mathbf{d}$ is the two-level transition dipole moment in the z direction. In fact, since $\mathbf{P}_{\mathrm{T}\alpha}$ commutes with gauge-fixing transformations, Eq. (216) is an example of a truncated expression that is actually independent of the gauge within which truncation is performed.

To obtain the Hamiltonian, truncation within the multipolar gauge gives the following multipolar-gauge multimode QRM:

$$H_1^2=\tilde{\omega}_m\sigma^+\sigma^-+\sum_k\omega_k\left(a_k^\dagger a_k+\frac{1}{2}\right)+i\sum_k g_k(a_k^\dagger-a_k)\sigma^x, \tag{217}$$

where $g_k=d\sqrt{\omega_k/2v}$ and we have absorbed the multipolar-gauge polarization self-energy term into a renormalization of the two-level transition frequency denoted $\tilde{\omega}_m$. Sánchez Muñoz, Nori, and De Liberato (2018) [see also Casanova *et al.* (2010)] demonstrated via a comparison with numerical results utilizing matrix product states that, for sufficiently large coupling strengths and numbers of modes, the two-level system frequency $\tilde{\omega}_m$ may be neglected in Eq. (217), resulting in the following independent-boson model:

$$H_1^2\approx\sum_k\omega_k\left(a_k^\dagger a_k+\frac{1}{2}\right)+i\sum_k g_k(a_k^\dagger-a_k)\sigma^x. \tag{218}$$

Since σ^x is now a symmetry, the Hamiltonian is easily diagonalized using a polaron transformation,

$$\mathcal{T}_{10}=\exp\left[i\sum_k\frac{g_k}{\omega_k}(a_k^\dagger+a_k)\sigma^x\right]. \tag{219}$$

Equation (219) is the type of transformation that was encountered in Sec. IV. Although it is not in general a gauge transformation [as defined by Eqs. (96) and (97)], we noted in Sec. IV.E that when acting on the canonical momentum Π this transformation has the same effect as the projected PZW gauge-fixing transformation PR_{10}.

The dynamics of the observables O_α closely mirror those found for free space in Sec. VI.C.1. Using Eq. (218), we obtain

$$a_k(t)=a_k e^{-i\omega_k t}+g_k\int_0^t ds\, e^{-i\omega_k(t-s)}\sigma^x(s)\equiv a_{k,\mathrm{vac}}(t)+a_{k,s}(t). \tag{220}$$

Note that here the vacuum-source partitioning is that given by the multipolar gauge. This is the most convenient partitioning

if we want to determine averages when assuming an initial bare state in the multipolar gauge, which corresponds to assuming a well-defined state of energy of a fully localized dipole. The operator $\sigma^x(s) = \sigma^x(0) = \sigma^x$ is time independent since the two-level dipole energy has been neglected. As a result the temporal integral in Eq. (220) can be evaluated immediately and, since σ^x is stationary, so too is the electrostatic field $P_{\mathrm{T}\alpha}(t,x) = P_{\mathrm{T}\alpha}(0,x)$ defined in Eq. (216). The negative frequency fields are found to be

$$\begin{aligned} O_\alpha^{(-)}(t,x) &= \Pi_{\mathrm{vac}}^{(-)}(t,x) + O_{\alpha,\mathrm{s}}^{(-)}(t,x), \\ O_{\alpha,\mathrm{s}}^{(-)}(t,x) &= \Pi_{\mathrm{s}}^{(-)}(t,x) + P_{\mathrm{T1}}^{(-)}(t,x) - P_{\mathrm{T}\alpha}^{(-)}(t,x) \\ &= \frac{d}{2v}\sigma^x(1-\alpha) + \sum_{k>0}^{N} \frac{d}{v}\sigma^x(e^{i\omega_k t} - \alpha)\cos[kx], \\ P_{\mathrm{T}\alpha}^{(-)} &:= \sum_k \frac{d}{2v}\sigma^x \alpha e^{-ikx} = \frac{1}{2}P_{\mathrm{T}\alpha}, \end{aligned} \tag{221}$$

where the integer N sets the total number of modes retained within the model. Positive frequency components are obtained by Hermitian conjugation, and the sum of positive and negative frequency parts of a field gives the total field. By construction, these expressions yield $O_\alpha = O_\alpha^{(-)} + O_\alpha^{(+)}$ for any α. Choosing $\alpha = 1$ gives the case $O_1 = \Pi = -D_{\mathrm{T}} = -E_{\mathrm{T}} - P_{\mathrm{T1}}$.

It is now possible to evaluate the average of arbitrary functions of O_α, $O_\alpha^{(-)}$, and $O_\alpha^{(+)}$ using any initial state. We use both the initial multipolar bare state $|\epsilon^1, 0\rangle$ and the ground state, which is represented by the vector $|\epsilon^0, 0\rangle$ in the polaron frame. Since we have neglected the dipole energy and the polaron transformation coincides with the projected PZW transformation when acting on Π (though not more generally), for the purpose of finding the dynamics of O_α the polaron frame is simply the Coulomb gauge. Specifically, we have $\mathcal{T}_{10}\Pi\mathcal{T}_{10}^\dagger = \Pi - P_{\mathrm{T1}}$ and $\mathcal{T}_{10}O_\alpha\mathcal{T}_{10}^\dagger = \Pi - P_{\mathrm{T}\alpha}$. Since the operator $\mathcal{T}_{10}\Pi\mathcal{T}_{10}^\dagger$ represents the observable $-D_{\mathrm{T}}$ in the polaron frame, the operator Π represents the observable $-D_{\mathrm{T}} + P_{\mathrm{T1}} = -E_{\mathrm{T}}$, as in the Coulomb gauge. In this gauge the electrostatic field is absorbed into the definition of the dipole. Further still, within the approximations made the Coulomb gauge coincides with the JC gauge [$\alpha_{\mathrm{JC}} = \tilde{\omega}_m/(\tilde{\omega}_m + \omega_k) \approx 0$]. Thus, the simple treatment in which the free dipole Hamiltonian has been neglected is unable to distinguish between electrostatic and virtual-photonic bound fields. Note XIII of the Supplemental Material (245) indicates that this distinction is also obscured when considering the near-field limit of the ground-state energy density in free space, where the total electric energy density becomes approximately purely electrostatic, as shown in Eq. (213) in note XIV of the Supplemental Material (245). We emphasize that the coincidence of the Coulomb gauge, the JC gauge, and the polaron frame for calculating averages of functions of Π does not occur without the simplifications made. In general, these representations are distinct.

We now calculate various quadratic energy densities as in note XIII of the Supplemental Material (245). For the initial state $|\epsilon^1, 0\rangle$ we obtain

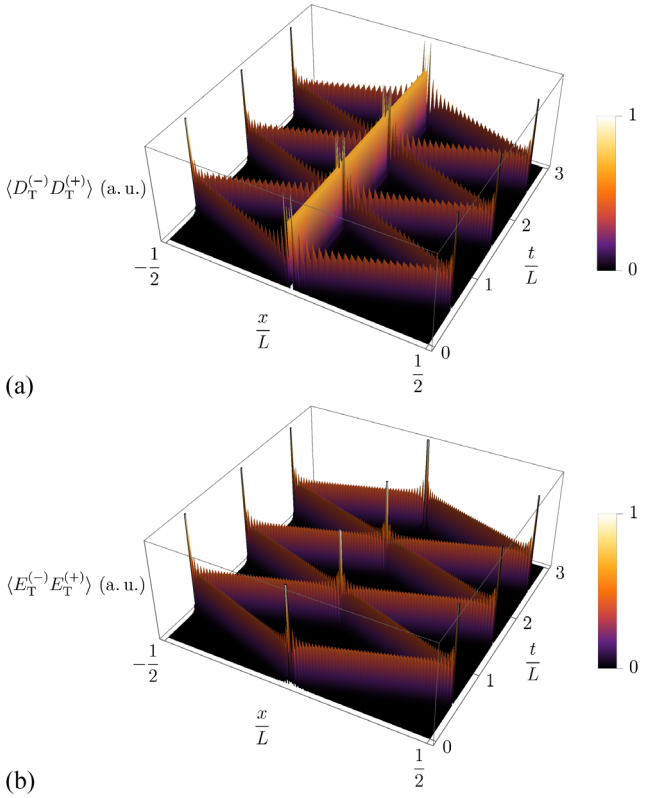

FIG. 12. Averages (a) $\langle D_{\mathrm{T}}^{(-)}(t,x) D_{\mathrm{T}}^{(+)}(t,x)\rangle$ and (b) $\langle E_{\mathrm{T}}^{(-)}(t,x) E_{\mathrm{T}}^{(+)}(t,x)\rangle$ plotted with space and time, showing the presence and absence of a bound field around the multipolar- and Coulomb-gauge dipoles, respectively. Essentially the same propagating field is obtained in both cases. We have assumed $N = 50$ and normalized both densities via the maximum value attained when the propagating field is coincident with the dipole [$(t,x) = (nL, 0)$, $n \in \mathbb{Z}$].

$$\langle O_\alpha(t,x)^2\rangle - E_{\mathrm{vac}} = \left[\sum_k \frac{d}{v}(\cos[kx - \omega_k t] - \alpha\cos[kx])\right]^2, \tag{222}$$

$$\langle O_\alpha^{(-)}(t,x) O_\alpha^{(+)}(t,x)\rangle = \left|\frac{d}{2v}(1-\alpha) + \sum_{k>0}^{N}\frac{d}{v}(e^{i\omega_k t} - \alpha)\cos[kx]\right|^2, \tag{223}$$

where $E_{\mathrm{vac}} = \sum_k \omega_k/2v$ is an energy density of the vacuum. For $\alpha = 1$ (multipolar gauge), Eq. (223) reduces to the result obtained by Sánchez Muñoz, Nori, and De Liberato (2018). Ground-state averages are obtained using the polaron frame and are found to be $\langle O_\alpha(t,x)^2\rangle_G - E_{\mathrm{vac}} = \langle P_{\mathrm{T}\alpha}(t,x)^2\rangle_G$, and $\langle O_\alpha^{(-)}(t,x) O_\alpha^{(+)}(t,x)\rangle_G = (1/4)\langle P_{\mathrm{T}\alpha}(t,x)^2\rangle_G$, where

$$\langle P_{\mathrm{T}\alpha}(t,x)^2\rangle_G = \left[\sum_{k\neq 0}\frac{d}{v}\alpha\cos[kx]\right]^2. \tag{224}$$

Equation (224) confirms that, within the approximations made, the bound field tied to the α-gauge dipole is simply the electrostatic field $P_{\mathrm{T}\alpha}$. In the Coulomb gauge this field is fully included within the definition of the dipole, so $\langle O_0(t,x)^2\rangle_G - E_{\mathrm{vac}} = 0$. Figure 12 shows

$\langle O_\alpha^{(-)}(t,x)O_\alpha^{(+)}(t,x)\rangle$ given in Eq. (223) for the Coulomb and multipolar gauges $\alpha = 0$ and 1, respectively. It can be seen that all gauges possess essentially the same propagating fields. In contrast the ground-state bound-field energy has the weight α^2 within the gauge α and is evidently highly localized at the position of the dipole within the one-dimensional model employed.

Sánchez Muñoz, Nori, and De Liberato (2018) proposed that the initial multipolar bare state $|\epsilon^1, 0\rangle$ could be prepared by controlling the interaction. However, given the level of localization of the bound field, it is far from clear that the latter could ever be separated from the dipole allowing the corresponding interaction to be controlled. A possible exception may be to quickly move the dipole in and out of the cavity. As described in Sec. V.D, in this case the relevant gauge for modeling the interaction using a time-dependent coupling will strongly depend on the microscopic details of the system.

The treatment of this section is highly idealized. The cavity is taken as one dimensional, the two-level truncation has also been made, and the dipole moment dynamics have been taken to be approximately stationary. The extension of these results using more realistic treatments warrants further investigation, including a more physical model for the cavity and a more sophisticated method of solution, for example, via a variational polaron *Ansatz* (Díaz-Camacho, Bermudez, and García-Ripoll, 2016).

Evidently, the physical nature of the internal cavity field strongly depends on the gauge relative to which it is defined. As we have emphasized, gauge ambiguities arise because it is not always clear which subsystems should be considered operationally addressable. In particular, the interaction between the system of interest and the apparatus used in preparation and measurement must be defined relative to the choice of gauge. Simple gedanken experiments for the weak measurement of intracavity subsystems within the weak-coupling regime were introduced some time ago by Compagno, Passante, and Persico, 1988a, 1988b, 1990, 1991, 1995). We discuss these models in note XVI of the Supplemental Material (245).

B. Ground-state superradiance

Here we exemplify the importance of the preceding discussions concerning intracavity fields and subsystem gauge relativity by reviewing the phenomenon of ground-state superradiance [also called photon condensation by Andolina *et al.* (2019, 2020)].

1. Dicke models

Ground-state superradiance was first predicted in the Dicke model (Dicke, 1954; Hepp and Lieb, 1973; Wang and Hioe, 1973). There is now an extensive literature on this topic including extended Dicke models (Carmichael, Gardiner, and Walls, 1973; Hioe, 1973; Pimentel and Zimerman, 1975; Emeljanov and Klimontovich, 1976; Sung and Bowden, 1979), connections with quantum chaos (Holstein and Primakoff, 1940; Emary and Brandes, 2003a, 2003b), driven and open systems (Grimsmo and Parkins, 2013; Klinder *et al.*, 2015; Gegg *et al.*, 2018; Kirton and Keeling, 2018; Peng *et al.*, 2019), and artificial systems (Yamanoi, 1979; Haug and Koch, 1994; Lee and Johnson, 2004; Nataf and Ciuti, 2010; Viehmann, von Delft, and Marquardt, 2011; Todorov and Sirtori, 2012; Bamba and Ogawa, 2014b; Leib and Hartmann, 2014; Bamba, Inomata, and Nakamura, 2016; Jaako *et al.*, 2016; Bamba and Imoto, 2017; De Bernardis, Jaako, and Rabl, 2018). The topic has received renewed interest in light of rapid progress in magnonic systems and in controlling correlated electron systems inside cavities (Andolina *et al.*, 2019, 2020; Mazza and Georges, 2019; Nataf *et al.*, 2019; Guerci, Simon, and Mora, 2020; Bamba *et al.*, 2022).

Despite this, whether or not a phase transition does indeed occur and its precise nature have remained open questions. This is due to the existence of so-called no-go theorems, which prohibit a superradiant phase and which have been proven in the Coulomb gauge (Rzazewski, Wódkiewicz, and Zakowicz, 1975). Further variants of this theorem have subsequently been both refuted and confirmed (Kudenko, Slivinsky, and Zaslavsky, 1975; Emeljanov and Klimontovich, 1976; Rzazewski, Wódkiewicz, and Zakowicz, 1976; Knight, Aharonov, and Hsieh, 1978; Bialynicki-Birula and Rzazewski, 1979; Sung and Bowden, 1979; Yamanoi, 1979; Rzazewski and Wódkiewicz, 1991; Keeling, 2007; Nataf and Ciuti, 2010; Vukics and Domokos, 2012; Bamba and Ogawa, 2014b; Vukics, Grießer, and Domokos, 2014; Tufarelli *et al.*, 2015; Grießer, Vukics, and Domokos, 2016; Bamba and Imoto, 2017; Andolina *et al.*, 2019, 2020).

Keeling (2007) noted that, since the radiation modes are distinct in the Coulomb and multipolar gauges, a ground-state phase transition may possess different characterizations and showed that a ferroelectric phase transition occurs within the Coulomb gauge at the same point in parameter space as the superradiant phase transition of the conventional Dicke model. More recently, Stokes and Nazir (2020b) showed that a unique (gauge-invariant) phase transition can be supported within cavity QED systems using the one-parameter α-gauge framework. It was further shown that the macroscopic manifestation is gauge invariant, but that the classification of the phase transition depends on the gauge relative to which the quantum subsystems are defined.

For a cavity containing N dipoles labeled by $\mu = 1, \ldots, N$, with dipole moments $\mathbf{d}_\mu$ and fixed positions $\mathbf{R}_\mu$, the α-dependent canonical momenta are found to be (Stokes and Nazir, 2020b)

$$\mathbf{p}_\mu = m\dot{\mathbf{r}}_\mu - e(1-\alpha)\mathbf{A}(\mathbf{R}_\mu), \tag{225}$$

$$\mathbf{\Pi}(\mathbf{x}) = -\mathbf{E}_{\mathrm{T}}(\mathbf{x}) - \mathbf{P}_{\mathrm{T}\alpha}(\mathbf{x}). \tag{226}$$

The Hamiltonian is the total energy (Stokes and Nazir, 2020b) wherein the total electrostatic energy can be split into an atomic binding energy for each dipole V and an interdipole electrostatic coupling V_{dip} (dipole-dipole interaction).

Assuming that the dipole moments $d = \mathbf{d}\cdot\mathbf{e}$ point in the direction of the cavity polarization $\mathbf{e}$, the single-mode approximation is performed in such a way as to preserve gauge invariance; see Secs. II.B and III.F.2. This eliminates the need to regularize $\mathbf{P}_{\mathrm{T}}$ (Vukics, Grießer, and Domokos, 2015) and ensures that the transverse commutation relation for

the canonical fields is preserved. The fundamental kinematic relations given by Eqs. (225) and (226) are therefore also preserved. To obtain a Dicke Hamiltonian the limit of closely spaced dipoles around the origin ($\mathbf{R}_\mu \approx \mathbf{0}$) is taken, and the dipoles are approximated as two-level systems. The following collective operators are then introduced: $J^i_\alpha = \sum_{\mu=1}^N \sigma^i_{\mu\alpha}$, $i = \pm, z$, where $\sigma^\pm_{\mu\alpha}$ are the raising and lowering operators of the μth two-level dipole and $\sigma^z_{\mu\alpha} = [\sigma^+_{\mu\alpha}, \sigma^-_{\mu\alpha}]/2$.

Although the nontruncated Hamiltonian H is unique, we now have a continuous infinity of Dicke Hamiltonians $H^{\alpha,2}$ such that $H^{\alpha,2}$ and $H^{\alpha',2}$ are not equal when $\alpha \neq \alpha'$ (De Bernardis *et al.*, 2018; Roth, Hassler, and DiVincenzo, 2019; Stefano *et al.*, 2019; Stokes and Nazir, 2019). The breaking of gauge invariance due to truncation turns out not to be a barrier in eliminating all ambiguities regarding the occurrence and nature of a quantum phase transition.

The thermodynamic limit is defined by $N \to \infty$, $V \to \infty$, with $\rho = N/V$ constant. In this limit the Holstein-Primakoff map defined by $J^z_\alpha = b^\dagger_\alpha b_\alpha - N/2$, $J^+_\alpha = b^\dagger_\alpha \sqrt{N - b^\dagger_\alpha b_\alpha}$, and $J^-_\alpha = (J^+_\alpha)^\dagger$, where $[b_\alpha, b^\dagger_\alpha] = 1$, is used (Holstein and Primakoff, 1940; Emary and Brandes, 2003a, 2003b). The Hamiltonian obtained by substituting these expressions into $H^{\alpha,2}$ is denoted as $H^{\alpha,2}_{\rm th}$.

The Hamiltonian is found to support two distinct phases and reads (Stokes and Nazir, 2020b)

$$H^{\alpha,2,{\rm i}}_{\rm th} = E^{\rm i}_{\alpha+} f^{{\rm i}\,\dagger}_\alpha f^{\rm i}_\alpha + E^{\rm i}_{\alpha-} c^{{\rm i}\,\dagger}_\alpha c^{\rm i}_\alpha + \tfrac{1}{2}(E^{\rm i}_{\alpha+} + E^{\rm i}_{\alpha-}) + C^{\rm i}, \tag{227}$$

where the superscript i is either i = n for normal phase or i = a for abnormal phase. The polariton operators $f^{\rm i}_\alpha, c^{\rm i}_\alpha$ are bosonic ones satisfying $[f^{\rm i}_\alpha, f^{{\rm i}\,\dagger}_\alpha] = 1 = [c^{\rm i}_\alpha, c^{{\rm i}\,\dagger}_\alpha]$, with all other commutators vanishing. The polariton energies $E^{\rm i}_{\alpha\pm}$ and constant $C^{\rm i}$ are known functions of the couplings and frequencies appearing in the Hamiltonian $H^{\alpha,2}$. It can be shown that the lower polariton energy $E^{\rm n}_-$ is real provided that

$$\tau := \frac{\omega_m}{2\rho d^2} \geq 1, \tag{228}$$

while the lower polariton energy $E^{\rm a}_-$ is real provided that

$$\tau \leq 1. \tag{229}$$

It can also be shown that $H^{\alpha,2,{\rm n}}_{\rm th} = H^{\alpha,2,{\rm a}}_{\rm th}$ for $\tau = 1$. As ρd^2 is increased, a unique phase transition is predicted to occur at the critical point $\tau = 1$ in parameter space, beyond which the normal-phase Hamiltonian $H^{\alpha,2,{\rm n}}_{\rm th}$ breaks down and the abnormal-phase Hamiltonian $H^{\alpha,2,{\rm a}}_{\rm th}$ takes over. This prediction is gauge invariant.

It remains only to determine the nature of the unique phase transition predicted. To demonstrate equivalence between all gauges, the α-gauge transverse polarization $P_{{\rm T}\alpha} = \alpha \mathbf{e} \cdot \mathbf{P}_{\rm T} = \alpha(\Pi_0 - \Pi_1)$ is calculated. In the normal phase the thermodynamic limit of this quantity, denoted as $P_{{\rm T}\alpha,{\rm th}}$, vanishes, whereas in the abnormal phase it is found to be $P^{\rm a}_{{\rm T}\alpha,{\rm th}} = -\alpha\rho d\sqrt{1-\tau^2}$. It can be further shown that in the thermodynamic limit one obtains $\Pi^{\rm a}_{\rm th} = -P^{\rm a}_{{\rm T}\alpha,{\rm th}}$, such that choosing $\alpha = 0$ we have $-E^{\rm a}_{{\rm T},{\rm th}} = \Pi^{\rm a}_{\rm th} = 0$, thereby verifying the fundamental kinematic relation (226). This establishes consistency between all gauges. The onset of the abnormal phase manifests in the form of a macroscopic value of the gauge-invariant field $\mathbf{P}_{\rm T}$;

$$P^{\rm a}_{{\rm T},{\rm th}} = P^{\rm a}_{{\rm T}1,{\rm th}} = -\rho d\sqrt{1-\tau^2}. \tag{230}$$

Previous no-go and counter–no-go results can be reconciled by noting that radiation is gauge relative. In the Coulomb gauge radiation is defined by $\mathbf{\Pi} = -\mathbf{E}_{\rm T}$ such that the phase transition does not appear superradiant in character and only the material subsystem acquires a macroscopic population. This constitutes a "no-go theorem" for superradiance defined relative to the Coulomb gauge. In the multipolar gauge radiation is defined by $\mathbf{\Pi} = -\mathbf{E}_{\rm T} - \mathbf{P}_{\rm T}$ such that both the material and radiative subsystems acquire a macroscopic population in the abnormal phase. This constitutes a "counter–no-go theorem" for superradiance defined relative to the multipolar gauge. These results are not in contradiction, because they are referring to different definitions of radiation. Indeed, the results demonstrate that they are in fact equivalent (Stokes and Nazir, 2020b). More generally, since $\mathbf{\Pi} = -\mathbf{E}_{\rm T} - \mathbf{P}_{{\rm T}\alpha}$, the degree to which the unique phase transition is classed as superradiant is directly determined by the value of α.

As we have seen, α controls the balance between localization and dressing in defining the quantum subsystem called matter. In Sec. VII.A we observed that the field $\mathbf{P}_{\rm T}$ is highly localized at the position of the dipole within the approximations made and the one-dimensional model adopted. As discussed in Sec. VII.A and note XVI of the Supplemental Material (245), which predictions are most relevant depends on which observables are accessible via the available preparation and measurement protocols.

2. Condensed matter systems in the Coulomb gauge

The superradiant phase transition has been discussed predominantly in the context of Dicke-type models. The gauge invariance of the predicted instability and the gauge invariance of its manifestation are now established. However, there remains a question as to whether such simplified models can realistically describe actual physical systems; see Grießer, Vukics, and Domokos (2016) for a discussion. Recent work by Andolina *et al.* (2019, 2020), Mazza and Georges (2019), Nataf *et al.* (2019), Guerci, Simon, and Mora (2020), Bamba *et al.* (2022), and Rouse, Stokes, and Nazir (2022) moved beyond simplified Dicke model–type treatments. Strongly correlated electron systems of the type encountered in condensed matter theory are considered rather than the gas of dipoles occurring in the Dicke model.

Andolina *et al.* (2019) showed that ground-state photon condensation cannot occur in strongly correlated electron systems including an arbitrary electron-electron interaction potential, but considering only a single cavity mode and only photons defined relative to the Coulomb gauge. Andolina *et al.* (2020) advanced these findings by considering a three-dimensional electron system (3DES) in an inhomogeneous

cavity field, i.e., one that varies in space. Again, only photons defined relative to the Coulomb gauge are considered, but in this case it is found that photon condensation can occur if

$$\chi_{\mathrm{orb}}(k) > \frac{1}{4\pi}, \tag{231}$$

where $\chi_{\mathrm{orb}}(k)$ is the $\mathbf{k}$-space nonlocal orbital magnetic susceptibility of the 3DES (Giuliani and Vignale, 2005). If the model is extended to include the spin of electrons, this condition becomes $\chi_{\mathrm{orb}}(k)+\chi_{\mathrm{spin}}(k) > 1/4\pi$, where $\chi_{\mathrm{spin}}(k)$ is the spin magnetic susceptibility.

This transition to photon condensation possesses a simple interpretation as a magnetic instability (Andolina *et al.*, 2020). Specifically, Andolina *et al.* (2020) defined the magnetic energy of a material subject to a magnetic field $\mathbf{B}$ as $E_M = \int d^3x \mathbf{H}\cdot\mathbf{B}$, where $\mathbf{H} = \mathbf{B} - \mathbf{M}$ and $\mathbf{M}$ is the orbital magnetization of the material, which is traditionally interpreted as describing the response of the material to the applied field. Assuming linear response theory in which $\mathbf{M}$ is a linear functional of $\mathbf{B}$ and χ_{orb}, one then finds that E_M can be written (Andolina *et al.*, 2020)

$$E_M = -2\pi \int d^3x \int d^3x' \delta(\mathbf{x}-\mathbf{x}')\chi_{\mathrm{orb}}(|\mathbf{x}-\mathbf{x}'|)\mathbf{B}(\mathbf{x})\cdot\mathbf{B}(\mathbf{x}'). \tag{232}$$

An instability occurs if $E_M < 0$. Upon Fourier transforming E_M in Eq. (232), this inequality gives inequality (231), which is the condition for photon condensation. Note that relative to gauge α photons are defined by $\mathbf{\Pi} = -\mathbf{E}_{\mathrm{T}} - \alpha\mathbf{P}_{\mathrm{T}}$ and thus, upon noting the traditional interpretation of $\mathbf{P}$ as describing the response of a material to an electric field, one might expect condensation of photons$_\alpha$ to be related to electric instability for any $\alpha \neq 0$. This was confirmed by Rouse, Stokes, and Nazir (2022) for the case of a jellium source within a cavity.

Mazza and Georges (2019) considered strongly correlated electrons coupled to a single cavity mode in the Coulomb gauge and affirmed the no-go theorem for condensation of these photons. However, it was reported that the situation changes when electronic interactions and delocalization are taken into account. It was found that in a two-band model of interacting electrons a phase supporting condensation of excitons and photons can occur, even when one considers only one cavity mode.

Guerci, Simon, and Mora (2020) considered one- and two-dimensional strongly correlated electron systems coupled to a cavity field in the Coulomb gauge. The no-go theorem is again affirmed for the case of a single-mode homogeneous field, while photon condensation is found to be possible for a nonuniform field. Nataf *et al.* (2019) also considered an inhomogeneous cavity field coupled to a two-dimensional electron system in the Coulomb gauge, including spin-orbit coupling and a perpendicular applied magnetic field. It was found that a superradiant phase transition can occur. We conclude this section by remarking that the investigation of strongly correlated electron cavity QED systems beyond a restriction to the Coulomb gauge, as initially undertaken by Rouse, Stokes, and Nazir (2022), warrants further study.

C. Extracavity fields: Overview

The description of external coupling to the cavity has received considerable attention. We provide an overview here before discussing specific simple models in subsequent sections. We are again faced with two problems outside of traditional regimes. The first concerns the determination of which approximations might be applied and when, and the second concerns the determination of which physical states and observables are relevant in preparation and measurement.

Although the two problems are not unrelated, we consider the first problem first. For weakly coupled subsystems dissipation and decoherence can be modeled via separate loss mechanisms as though the subsystems were uncoupled. This constitutes the so-called local approach to deriving a master equation for the matter-cavity system. For example, the stationary state of a qubit in a cavity described by the local master equation

$$\dot{\rho} = -i[H,\rho] + \frac{\Gamma}{2}(2\sigma^-\rho\sigma^+ - \{\sigma^+\sigma^-,\rho\}) + \frac{\kappa}{2}(2a\rho a^\dagger - \{a^\dagger a,\rho\}) \tag{233}$$

is simply $|\epsilon^g,0\rangle$. In Eq. (233) $\sigma^+ = |\epsilon^e\rangle\langle\epsilon^g|$ is the qubit raising operator, $\sigma^- = (\sigma^+)^\dagger$, and a is the annihilation operator for the cavity. Dissipation is described via separate Lindblad tails corresponding to the qubit and mode. In the so-called global approach dissipation is instead described in the dressed basis of the light-matter system.

The difference between local and global approaches has been discussed extensively and in various contexts (Walls, 1970; Schwendimann, 1972; Carmichael and Walls, 1973; Scala, Militello, Messina, Maniscalco *et al.*, 2007; Scala, Militello, Messina, Piilo, and Maniscalco, 2007; Joshi *et al.*, 2014; Santos and Semião, 2014; Manrique *et al.*, 2015; Purkayastha, Dhar, and Kulkarni, 2016; Santos and Landi, 2016; Deçordi and Vidiella-Barranco, 2017; González *et al.*, 2017; Stockburger and Motz, 2017; Chiara *et al.*, 2018; Hamedani Raja *et al.*, 2018; Hewgill, Ferraro, and De Chiara, 2018; Mitchison and Plenio, 2018; Naseem, Xuereb, and Müstecaplıoğlu, 2018; Seah, Nimmrichter, and Scarani, 2018; Stokes and Nazir, 2018; Cattaneo *et al.*, 2019; Maguire, Iles-Smith, and Nazir, 2019). Cresser noted early on that the local master equation could apparently break down when describing a lossy Jaynes-Cummings model (Cresser, 1992). Hofer *et al.* (2017) found by comparison with exact predictions that the local equation may perform better in the weak-coupling regime, while the global master equation is generally better in the strong-coupling regime. However, the relative validity of the two approaches depends on the form of secular approximation used. Cattaneo *et al.* (2019) showed that the global master equation with partial secular approximation is always most accurate when Born-Markov approximations are also valid. The local approach is often claimed to fail (Santos and Semião, 2014; Manrique *et al.*, 2015; Deçordi and Vidiella-Barranco, 2017), but it has been shown to be thermodynamically consistent for fairly large ranges of coupling strengths (González *et al.*, 2017; Hofer *et al.*, 2017).

Here we note that, since the gauge parameter α selects the form of the interaction, one would not expect the relative

applicability of local versus global master equations to be independent of α. In general, the losses in a light-matter system will depend on how it couples to the external system or environment (Bamba and Ogawa, 2013, 2014a). For example, Ciuti and Carusotto (2006) applied input-output theory to quantum wells within a microcavity such that the cavity coupled to external photonic modes via a number-conserving interaction, while the electronic system similarly couples to another bosonic environment. With this treatment it is predicted that ground-state virtual cavity and electronic excitations cannot leak out of the cavity. In contrast, De Liberato *et al.* (2009) used a form of a non-Markovian master equation to describe a two-level system coupled to radiation while assuming fast modulation of the vacuum Rabi frequency. It was predicted that extra cavity quantum vacuum radiation would occur for state-of-the-art circuit cavity QED systems.

Predictions such as those by Ciuti and Carusotto (2006) and De Liberato *et al.* (2009) are in general specific to the forms of coupling adopted; i.e., they are specific to the physical subsystems considered. Indeed, as we have noted the second task that we are faced with is identifying which states and observables are relevant. If counterrotating terms are non-negligible in the interaction of a light-matter system, then the local master equation description of its losses will result in photon generation in the environmental vacuum (Werlang *et al.*, 2008). This would typically be taken as indicating the onset of the regime in which the bare states are no longer meaningful, such that one should switch to a global description in which dissipation is described holistically using the dressed states of the full light-matter Hamiltonian (Beaudoin, Gambetta, and Blais, 2011; Bamba and Ogawa, 2013, 2014a; Boité, 2020). Similarly, a coarse-grained projection onto the vacuum state, as in the Born approximation, will induce apparently paradoxical spontaneous excitations in polaritonic systems. The paradox is resolved by accounting for correlations between the dressed ground state of the system and the environmental vacuum within the reservoir correlation functions of the master equation (Bamba and Ogawa, 2012).

If we are interested in determining measurement signals from a source, then the generic problem consists of two multilevel systems, a source and a detector, coupled to a common reservoir (as was considered in Sec. VI.C.2). However, the multilevel source need not be elementary. In particular, it could be an ultrastrongly coupled light-matter composite. In a "global approach," the light-matter composite is diagonalized and then weakly coupled to whatever is external (Bamba and Ogawa, 2013, 2014a; Di Stefano *et al.*, 2018; Boité, 2020; Salmon *et al.*, 2022). In particular, Di Stefano *et al.* (2018) adopted precisely this strategy as a means by which to apply Glauber photodetection theory when dealing with an ultrastrongly coupled light-matter composite that is weakly coupled to a photon absorber. The same method was applied by Salmon *et al.* (2022) for an understanding of cavity leakage using a simple semiphenomenological approach, which is reviewed in Sec. VII.D. In this case all weak-coupling results for loss and detection are recovered, with the only difference being that the eigenstates of the source are the dressed states of a composite. As previously discussed, in this context there is a balance to be struck between electromagnetic dressing and localization in spacetime. This balance is affected by the choice of gauge.

In note XVII of the Supplemental Material (245), we review microscopic descriptions of cavity QED systems, including a perfect cavity containing matter and an imperfect empty cavity. The task of describing leakage from an imperfect cavity containing matter is more involved. A phenomenological approach consists of matter coupled linearly to the cavity, which in turn couples linearly to an environment, with reasonable coupling functions being chosen. This is the approach employed by Ciuti and Carusotto (2006) that was already discussed.

A promising means by which to provide a description from first principles is to use the theory of QED within absorbing and dispersing media, as reviewed in note VII of the Supplemental Material (245). Bamba and Ogawa (2013) [see also Bamba and Ogawa (2012)] used this theory in conjunction with Maxwell boundary conditions to describe dissipation from a cavity containing bosonic matter (a polaritonic system) while considering the good-cavity limit. It was found that external modes couple linearly to polaritonic raising and lowering operators via number-conserving form. Since these operators are linear combinations of the cavity and matter subsystem raising and lowering operators, we note that this global description differs from a phenomenological local description via a Gardiner-Collett model (Gardiner and Collett, 1985).

Bamba and Ogawa (2014a) considered the coupling to external modes of an ultrastrongly coupled light-matter system. Both cavity and circuit QED implementations are considered. It is again noted that the phenomenological Gardiner-Collett Hamiltonian will break down. We also emphasize that in this situation the form of the system-environment interaction Hamiltonian will become significant, as was noted in Sec. VII.C. Two forms of interaction Hamiltonian are considered. One in which the cavity couples to external modes via the position quadrature $\sim a^\dagger + a$ (this is referred to as magnetic or inductive coupling), and one in which the coupling instead occurs through the momentum quadrature $\sim i(a^\dagger - a)$ (this is referred to as electric or capacitive coupling). Both coupling forms can be derived from an underlying Lagrangian. Note that in the absence of a dissipative transmission line the inductive and capacitively coupled light-matter system Hamiltonians are unitarily equivalent, but this is no longer the case for the full Hamiltonians that include coupling of the system to a transmission line. This is similar to the situation encountered in Sec. VII.C, where coupling to external modes was defined relative to different gauges, which resulted in different reduced descriptions that corresponded to physically distinct reduced systems of interest. Bamba and Ogawa (2014a) noted that the difference in results obtained from different coupling forms can be ignored in sufficiently weak-coupling regimes, as well as in the good-cavity limit, which is effectively defined by the applicability of certain Markovian approximations.

Khanbekyan *et al.* (2005) also employed the theory of absorbing and dispersing dielectrics reviewed in note VII of the Supplemental Material (245). Choosing the multipolar gauge, they considered leakage from a one-dimensional

high-Q cavity consisting of one perfect and one imperfect mirror and containing a dipole, in both the weak- and strong-coupling regimes. It was found that on timescales that are large compared to the inverse separation of neighboring cavity resonances, the internal cavity field may be expressed in terms of internal bosonic mode operators that obey quantum Langevin equations. Radiative input-output coupling and absorption losses can then be viewed as independent, with each possessing a damping rate and corresponding Langevin noise force. Thus, in the regime considered the phenomenological Gardiner-Collett approach (Gardiner and Collett, 1985) is valid, inasmuch as the description of absorption losses requires only that the model is supplemented with bilinear interaction Hamiltonians between the cavity modes and appropriately chosen bosonic loss channels.

Franke *et al.* (2019) similarly used the dielectric theory reviewed in note VII of the Supplemental Material (245) applied to a single dipole within the multipolar-gauge and weak-coupling regime. Their approach was to approximate the Green's function defined by Eq. (92) in note VII.1 of the Supplemental Material (245) using an expansion in mode functions corresponding to only a few resonant modes that are assumed to be dominant: so-called quasinormal modes (QNMs). The internal field to which the dipole couples is expressed in terms of the QNM functions and global bosonic mode operators, while the external field is similarly described but with the QNM functions replaced by regularized counterparts. The use of only one or two QNMs has been found to be accurate within weak-coupling regimes (Kamandar Dezfouli, Gordon, and Hughes, 2017).

The extension of the descriptions given by Khanbekyan *et al.* (2005) and Franke *et al.* (2019) to ultrastrongly coupled light-matter systems within an arbitrary gauge warrants further study. Note, however, that a plausible physical model for the description of a lossy cavity containing atomic systems can already be proposed by combining insights from the case of a perfect cavity containing atomic systems [see note XVII.1 of the Supplemental Material (245)] with insights from the case of an imperfect but empty cavity [see note XVII.2 of the Supplemental Material (245)]. Specifically, in note XVII.2 of the Supplemental Material (245) it is shown that for a high-Q cavity a linear-coupling model between the cavity and external modes can be justified, while in note XVII.1 of the Supplemental Material (245) it is shown that a localized polarization that vanishes at the cavity boundary implies that the light-matter interaction is mediated entirely by the local cavity field evaluated at the positions of the atoms. Such local light-matter coupling away from the boundary should not affect the form of the coupling between the cavity and external modes at the boundary. Thus, within a gauge in which the atomic systems are highly localized, such as the multipolar gauge, a model in which atomic dipoles couple linearly to a cavity field that in turn couples linearly to external modes, would appear to be physically reasonable. In Sec. we review a spectroscopic signature of gauge relativity (Salmon *et al.*, 2022) that uses such a model and leads to the final master equation given in Eq. (239).

D. Spectroscopic signatures of gauge relativity via a simple model

An early attempt at modeling cavity leakage from an ultrastrongly coupled dipole-cavity system using different gauges was recently given by Salmon *et al.* (2022). They considered the simplest toy model system of a two-level dipole coupled to a single cavity mode with volume v and frequency ω in one spatial dimension, described by the multipolar-gauge QRM H_1^2 that is defined by the $\alpha = 1$ case of Eq. (105). Up to a constant this model reads

$$H_1^2 = \omega_m \sigma^+\sigma^- + \omega a^\dagger a + ig(a^\dagger - a)(\sigma^+ + \sigma^-), \tag{234}$$

where $\sigma^\pm$ are the raising and lowering operators for the two-level dipole with transition frequency ω_m, a and $a^\dagger$ are the cavity annihilation and creation operators for photons defined relative to the multipolar gauge, and $g = d\sqrt{\omega/2v}$ is the coupling strength. A dimensionless coupling strength is defined by $\eta = g/\omega$. Recall that the multipolar-gauge two-level truncation of the dipole is expected to be accurate for a sufficiently anharmonic dipole; see Sec. IV.F. Leakage at a rate κ to external environmental modes k described by bosonic operators $b_k, b_k^\dagger$ can be described in the gauge α using a linear weak-coupling Hamiltonian $V^\alpha_{\text{cav-ext}} = \pi \otimes \sum_k g_k(b_k + b_k^\dagger)$, where $\pi \coloneqq \sqrt{2v/\omega}\Pi$ is the cavity canonical momentum quadrature. This operator represents a different physical observable in each different gauge α.

Since the light-matter system is ultrastrongly coupled, Salmon *et al.* (2022) assumed a global approach in which dissipation is described using the dressed states of the light-matter composite. If one applies the standard derivation of the Lindblad master equation (cf. Sec. V.C.2) with the reduced system of interest being the dipole-cavity system described by the dressed states of the QRM H_1^2 and with the coupling to the bath $V^\alpha_{\text{cav-ext}}$, then one obtains

$$\dot\rho = i[\rho, H_1^2] + \mathcal{L}(\rho, x), \tag{235}$$

$$\mathcal{L}(\rho, x) = \kappa(x\rho x^\dagger - \tfrac{1}{2}\{x^\dagger x, \rho\}), \tag{236}$$

where ρ is the density operator describing the dipole-cavity system, H_1^2 is the multipolar-gauge QRM, and x is a Lindblad operator obtained by expressing the α-gauge canonical momentum quadrature π in the eigenbasis $\{|i\rangle\}$ of the QRM H_1^2 as

$$\pi = x + x^\dagger, \tag{237}$$

$$x = \sum_{\substack{i,j \\ i<j}} \langle i|\pi|j\rangle |i\rangle\langle j|. \tag{238}$$

Equation (235) constitutes a different physical description of cavity leakage for each different physical definition of $\pi = \sqrt{2v/\omega}\Pi$. In the multipolar gauge itself we have $\Pi = -D_\text{T} = i\sqrt{\omega/2v}(a^\dagger - a)$, where a and $a^\dagger$ are the same (multipolar-gauge) operators that appear in Eq. (234). Thus, for $\alpha = 1$ we have $\pi = \pi_1$, where $\pi_1 \coloneqq i(a^\dagger - a)$, yielding the corresponding master equation

$$\dot{\rho} = i[\rho, H_1^2] + \mathcal{L}_1(\rho, x), \tag{239}$$

$$\mathcal{L}_1(\rho, x) = \kappa(x\rho x^\dagger - \tfrac{1}{2}\{x^\dagger x, \rho\}), \tag{240}$$

$$x = \sum_{\substack{i,j \\ i<j}} \langle i|\pi_1|j\rangle |i\rangle\langle j|. \tag{241}$$

Salmon *et al.* (2022) referred to this result as the "dipole-gauge" master equation.

However, Salmon *et al.* (2022) assumed that the correct description is provided when the cavity couples to external modes via the transverse electric field E_T, which equals $-\Pi$ in the Coulomb gauge. The multipolar-gauge two-level truncation of the observable $\sqrt{2v/\omega}E_\mathrm{T}$ is $-\pi_1 - 2\eta\sigma^x$, where $\pi_1 \coloneqq i(a^\dagger - a)$. The resulting master equation is therefore

$$\dot{\rho} = i[\rho, H_1^2] + \mathcal{L}_0(\rho, x), \tag{242}$$

$$\mathcal{L}_0(\rho, x) = \kappa(x\rho x^\dagger - \tfrac{1}{2}\{x^\dagger x, \rho\}), \tag{243}$$

$$x = \sum_{\substack{i,j \\ i<j}} \langle i|(\pi_1 + 2\eta\sigma^x)|j\rangle |i\rangle\langle j|. \tag{244}$$

Salmon *et al.* (2022) referred to this master equation as the "dipole-gauge-fixed" master equation, which is clearly different from Eq. (239). Note that this master equation is obtained by assuming a coupling between the cavity and external modes using the Coulomb-gauge cavity canonical momentum E_T, but the two-level truncation of the dipole has been performed within the multipolar gauge, where, unlike in the Coulomb gauge, it is expected to be generally accurate for an anharmonic dipole; see Sec. IV.F. The observable E_T has therefore been expressed in terms of the multipolar-gauge operators σ^x and $a, a^\dagger$.

More generally, in the gauge α we have $\Pi = -E_\mathrm{T} - \alpha P_\mathrm{T}$, where $P_\mathrm{T} = d\sigma^x/v$. The multipolar-gauge two-level truncation of the observable $-\sqrt{2v/\omega}(E_\mathrm{T} + \alpha P_\mathrm{T})$ is represented by the operator $\pi_1 + 2(1-\alpha)\eta\sigma^x$, which results in the master equation

$$\dot{\rho} = i[\rho, H_1^2] + \mathcal{L}_\alpha(\rho, x), \tag{245}$$

$$\mathcal{L}_\alpha(\rho, x) = \kappa(x\rho x^\dagger - \tfrac{1}{2}\{x^\dagger x, \rho\}), \tag{246}$$

$$x = \sum_{\substack{i,j \\ i<j}} \langle i|[\pi_1 + 2(1-\alpha)\eta\sigma^x]|j\rangle |i\rangle\langle j|. \tag{247}$$

Equations (239) and (242) are the particular cases given by $\alpha = 1$ and 0, respectively. For each α the general master equation (245) constitutes a different physical model of cavity leakage in which the cavity is assumed to couple to external modes linearly through its canonical momentum Π, which represents the observable $-E_\mathrm{T} - \alpha P_\mathrm{T}$. In other words, cavity leakage is described relative to a choice of gauge.

The cavity emission spectrum is defined as the spectrum of the average external mode number operator $\langle b_k^\dagger b_k\rangle$ and is given using $V^\alpha_{\text{cav-ext}}$ as (Salmon *et al.*, 2022)

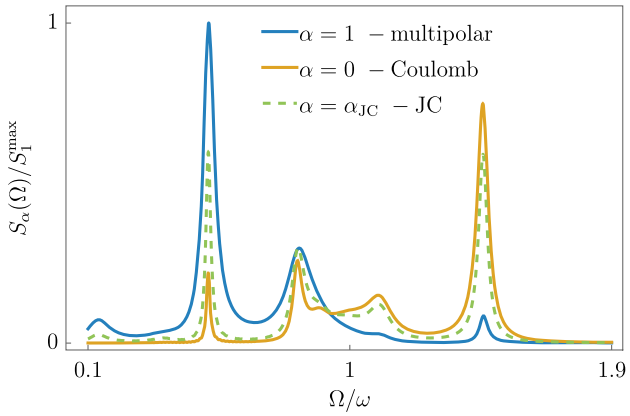

FIG. 13. Cavity emission spectra using the multipolar-gauge ($\alpha = 1$; darker curve), Coulomb-gauge ($\alpha = 0$; lighter curve), and Jaynes-Cummings-gauge ($\alpha = \alpha_\mathrm{JC} = 0.335\,115$; dashed curve) definitions of x for ultrastrong light-matter coupling $\eta = g/\omega = 0.5$ and weak incoherent pumping $P_\mathrm{inc} = 0.01g$. The spectra are normalized to the multipolar maximum and α_JC is determined for the same highly anharmonic double-well dipole as is considered in Sec. IV.F. Other parameters are $\kappa = 0.05g$ and $\delta = \omega/\omega_m = 1$.

$$S_\alpha(\Omega) \propto \mathrm{Re}\left[\int_0^\infty d\tau e^{i\Omega\tau}\langle x^\dagger(0)x(\tau)\rangle_{ss}\right], \tag{248}$$

where the conventional weak-coupling approximations have been applied in the dressed basis of the QRM and we consider the long-time limit. Like the master equation, the physical meaning of the spectrum is determined by the value of α, which specifies the physical observable in terms of which x is defined in Eq. (247). Salmon *et al.* (2022) considered the cases $\alpha = 0$ and 1, which define x in terms of E_T and D_T, respectively. Incoherent excitation of the dipole and coherent excitation under semiclassical driving are both considered. For incoherent driving Salmon *et al.* (2022) considered a phenomenological pump term, for which the master equation (245) becomes

$$\dot{\rho} = i[\rho, H_1^2] + \mathcal{L}_\alpha(\rho, x) + \mathcal{L}^\mathrm{inc}_\alpha(\rho, x), \tag{249}$$

$$\mathcal{L}^\mathrm{inc}_\alpha(\rho, x) = P_\mathrm{inc}(x^\dagger\rho x - \tfrac{1}{2}\{xx^\dagger, \rho\}). \tag{250}$$

Sample results for ultrastrong light-matter coupling and weak incoherent pumping are plotted in Fig. 13. They are markedly different for the two different gauges $\alpha = 0$ and 1 for ultrastrong light-matter coupling, as well as for the JC gauge, which lies between the two. Note that Salmon *et al.* (2022) considered a larger value of the cavity leakage rate ($\kappa = g/4$), for which one again sees clear qualitative differences between the spectra corresponding to different α, and one thus draws the same qualitative conclusions.

Salmon *et al.* (2022) assumed that the Coulomb-gauge model in Eq. (242) and the associated spectrum are "correct." Accordingly the multipolar-gauge model in Eq. (239) and the associated spectrum are deemed to "fail." Note that the Coulomb-gauge result can be transformed using the x_P-phase transformation $\mathcal{T}_{10}$ to give the following equivalent expression:

$$\dot{\rho} = i[\rho, h_1^2(0)] + \mathcal{L}_0(\rho, x), \tag{251}$$

$$\mathcal{L}_0(\rho, x) = \kappa(x\rho x^\dagger - \tfrac{1}{2}\{x^\dagger x, \rho\}), \tag{252}$$

$$x = \sum_{\substack{i,j \\ i<j}} \langle i|\pi_1|j\rangle |i\rangle\langle j|. \tag{253}$$

Here $h_1^2(0) = \mathcal{T}_{10} H_1^2 \mathcal{T}_{01}$ is the two-level model encountered in Sec. IV.C, the $|i\rangle$ denote its eigenvectors, and ρ denotes the density operator in the rotated frame. More generally, any two-level model unitary operator U^2 can be applied to any one of the master equations (245) corresponding to a fixed value of α, and this will result in an equivalent expression of the given master equation. Note that in all of these equations two-level truncation has been performed in the multipolar gauge and thus, provided that this truncation is accurate, each of the equations is an accurate approximation of a gauge-invariant equation. However, the master equations (245) corresponding to different α are not equivalent, because each one constitutes a different physical model of cavity leakage where the cavity couples to external modes via a different physical field.

The correct master equation can be determined only through the provision of a physical argument to prefer one of the results over another. As previously noted, Salmon *et al.* (2022) assumed that the $\alpha = 0$ result was correct based on the assumption that the cavity should couple to external modes via the transverse electric field E_T, which was referred to simply as the electric field by Salmon *et al.* (2022). The emission spectrum is then found using the same interaction Hamiltonian and is therefore given in terms of the same physical field. However, as described in Sec. VI, conventional photodetection theory uses the total electric field, which is equal to the field D_T at all points away from the source dipole itself. Moreover, boundary conditions defining an electromagnetic cavity are typically specified in terms of the local total electric field. A perfect conductor satisfies $\hat{\mathbf{n}} \times \mathbf{E}(\mathbf{x}) = \mathbf{0}$ for $\mathbf{x}$ on the boundary with unit normal vector $\hat{\mathbf{n}}$. Since $\mathbf{x}$ is not a point inside the source, we have $\mathbf{E}(\mathbf{x}) = \mathbf{D}_\mathrm{T}(\mathbf{x})$. Thus, in the simplified toy model discussed in this section, leakage to external modes through the field D_T seems to offer a more physically sensible description than leakage through E_T. According to these arguments, the specification by Salmon *et al.* (2022) of which result is correct and of which result fails should actually be reversed.

Regardless, these results demonstrate that the prediction of cavity leakage is strongly gauge relative because coupling of the cavity to external modes must be defined relative to a choice of gauge. The relativity becomes significant for sufficiently large values of the light-matter coupling strength even though the coupling $V^\alpha_{\text{cav-ext}}$ is weak.

VIII. CONCLUSIONS

In this review we have focused on the implications of gauge freedom for QED beyond conventional weak-coupling and Markovian regimes. We have shown that subsystems in QED are fundamentally gauge relative, meaning that in each gauge they are defined in terms of different physical observables. The fundamental condition known as gauge invariance states that the predictions for any physical observable must always be the same when found in different gauges. This is guaranteed by the unitarity of gauge-fixing transformations. However, if we compare predictions coming from different gauges of quantum subsystem properties such as photon number or light-matter entanglement, then we are comparing predictions for different physical observables, which are necessarily different. This is not a violation of gauge invariance. It is analogous to the fact that an interval in space or time between two events possesses a different value in different inertial frames even though the same labels "space" and "time" are used in every inertial frame.

Subsystem gauge relativity can be ignored within the idealized setting of scattering theory, beyond which it can be eliminated only when using various weak-coupling and Markovian approximations. It is therefore an important fundamental feature whenever such approximations cannot be employed, i.e., outside of gauge-nonrelativistic regimes.

We have provided descriptions of a number of simple systems, showing that subsystem gauge relativity is significant in the description of so-called virtual processes. It thereby affects the balance between localization and electromagnetic dressing. This has nontrivial implications for modeling controllable interactions, for photodetection theory, and for cavity QED. In all instances, the quantum subsystems, including reservoirs and measurement devices, must be defined relative to a choice of gauge. Beyond conventional weak-coupling and Markovian regimes, the physical predictions for subsystems defined relative to different gauges can be markedly different.

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See Supplemental Material at http://link.aps.org/supplemental/10.1103/RevModPhys.94.045003 for further details to complement the main text in the form of additional calculations, figures, and discussions of related topics.