Noncovariant gauge fixing in the quantum Dirac field theory of atoms and molecules

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Starting from the Weyl gauge formulation of quantum electrodynamics (QED), the formalism of quantum-mechanical gauge fixing is extended using techniques from nonrelativistic QED. This involves expressing the redundant gauge degrees of freedom through an arbitrary functional of the gauge-invariant transverse degrees of freedom. Particular choices of functional can be made to yield the Coulomb gauge and Poincaré gauge representations. The Hamiltonian we derive therefore serves as a good starting point for the description of atoms and molecules by means of a relativistic Dirac field. We discuss important implications for the ontology of noncovariant canonical QED due to the gauge freedom that remains present in our formulation.

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I. INTRODUCTION

Quantum electrodynamics (QED) in canonical form can be formulated in a number of different ways because the QED Lagrangian is independent of the velocity of the scalar potential. Formulations can be broadly categorized into two types. The first type comprises the manifestly covariant formulations within a Lorenz-type gauge such as the Feynman gauge. These require the addition of a gauge symmetry-breaking term to the Lagrangian after which the scalar potential can be taken as a legitimate canonical variable with a nonvanishing conjugate momentum. Quantization is carried out using an indefinite metric with the Lorenz subsidiary condition implemented as a constraint defining the physical space of states [1].

The second type of formulation consists of those in which the scalar potential is eliminated from the Lagrangian altogether. The vector potential is then the sole canonical coordinate in the Hamiltonian. Of course, the relative simplicity of such an approach comes at the price of sacrificing manifest covariance [2].

In the Weyl gauge the scalar potential is set identically equal to zero. In the Coulomb and Poincaré gauges it is eliminated in favor of the gauge-invariant degrees of freedom. How this is achieved, varies between the two gauges, so the resultant Lagrangians are distinct. In these cases the gauge is fixed at the classical level with a particular Lagrangian belonging to a particular gauge [2,3].

Conventional calculations in atomic physics and quantum optics most often start with a nonrelativistic Hamiltonian in the Coulomb or Poincaré gauge [2–6]. With regards to such calculations there are two main drawbacks. The first is their nonrelativistic description of the material degrees of freedom. The second is their ab initio restriction to a particular gauge, which makes them incapable of fully exploring the consequences of gauge freedom. Indicative of the potential benefit of a more general formulation is the fact that certain admixtures of the Coulomb and Poincaré gauge Hamiltonians have proven useful in quantum optics [7–9].

In this paper we focus on formulations of QED of the second (noncovariant) type in which the scalar potential is eliminated. Specifically, the aim of this paper is to address both of the issues raised above, first using a quantized Dirac field description for the material degrees of freedom and second keeping at the forefront the gauge freedom present in the theory.

The latter is achieved following Ref. [10] by reexpressing the gauge-dependent longitudinal degrees of freedom using the gradient of a linear functional of the gauge-invariant transverse degrees of freedom. The functional is defined in terms of an arbitrary c-number function $\boldsymbol{g}(\mathbf{x},\mathbf{x}')$, which then carries the gauge freedom of the theory.

As a description of the Maxwell field we use a Hilbert space of Schrödinger wave functionals of the vector potential in which an inner product can be defined through functional integration [11]. The material degrees of freedom can be defined similarly in terms of a Hilbert space of functionals of a Grassman field [11].

To obtain a Hamiltonian in an arbitrary noncovariant gauge (defined by $\boldsymbol{g}$), which is fully relativistic in the material degrees of freedom, we combine these elements with an adaptation of the quantum-mechanical gauge-fixing method presented in Ref. [12]. In this approach one initially adopts the Weyl gauge. From there the physical subspace of states (wave functionals) is defined as the subspace of states vanishing under the action of the Gauss law constraint. The Hamiltonian can be found in different noncovariant gauges through the use of unitary gauge-fixing transformations acting on the Weyl gauge Hamiltonian.

Particular choices of the function $\boldsymbol{g}$ can subsequently be made to render the Hamiltonian in a fixed gauge. Two particular choices yield the Coulomb gauge and Poincaré gauge Hamiltonians.

On the one hand our results are of importance for the relativistic theory of atoms and molecules and on the other they provide an interesting extension of the quantum-mechanical gauge-fixing formalism already employed in noncovariant relativistic QED. We will show that this extension has important implications with regards to the ontology of noncovariant QED in general because it allows for a thorough exploration of the gauge freedom of the theory in canonical (Hamiltonian) form. The requirement of gauge invariance of a result is translated into the requirement that it be independent of the choice of the arbitrary function $\boldsymbol{g}$.

There are four sections to this paper. In Sec. II we use the quantum-mechanical gauge-fixing formalism to obtain a Hamiltonian in an arbitrary noncovariant gauge. In Sec. III we discuss the implications of the gauge freedom still present in the formulation. We then define useful gauge-invariant operators and as an application address the problem of

causality in spatially separated material systems [13]. In Sec. IV we finish with a brief summary of our results.

II. THE QED HAMILTONIAN IN AN ARBITRARY NONCOVARIANT GAUGE

We will begin in Sec. II A by outlining how the general idea of gauge fixing we are going to employ works in the simple case of classical electrodynamics. The aim is then to extend this idea to noncovariant relativistic QED. To this end we start Sec. II B formally with the QED Lagrangian and obtain from it the Hamiltonian and canonical operators, which are supposed to satisfy canonical commutation relations. We go on to identify the Gauss law constraint, which defines the physical subspace of states, and review a particular class of transformations called residual gauge transformations [12].

In Sec. II C we identify the states of the system as Schrödinger wave functionals and determine the general form of a physical state using the coordinate representation for the canonical operators of the Maxwell field and the Gauss law constraint. From there we identify a general unitary gauge-fixing transformation U_g as a map from the physical space of states $\mathcal{H}_p$ to a space $\mathcal{H}_g$, which is the space of states for the gauge g. Next, in Sec. II D, we determine the effect of this transformation on the various operators of the theory and express the Hamiltonian in the arbitrary gauge g. We conclude in Sec. II E by using the Hamiltonian to calculate the Dirac equation in the gauge g.

A. Gauge in classical electrodynamics

In electrodynamics the electric and magnetic fields defined by

$$\begin{aligned} \mathbf{E} &= -\boldsymbol{\nabla}\phi - \frac{\partial \mathbf{A}}{\partial t}, \\ \mathbf{B} &= \boldsymbol{\nabla} \times \mathbf{A} \end{aligned} \tag{1}$$

are invariant under a change of gauge

$$\begin{aligned} \mathbf{A}' &= \mathbf{A} + \boldsymbol{\nabla} f, \\ \phi' &= \phi - \frac{\partial f}{\partial t} \end{aligned} \tag{2}$$

for an arbitrary function f. Clearly, the transverse vector potential $\mathbf{A}_\mathrm{T}$ is gauge invariant, while the (redundant) gauge-dependent degrees of freedom are longitudinal. For this reason the transverse vector potential serves as a convenient starting point relative to which vector potentials in other gauges can be defined by

$$\mathbf{A} = \mathbf{A}_\mathrm{T} + \boldsymbol{\nabla} f, \tag{3}$$

with the function f determining the gauge. The gauge-invariant transverse electric field is given by

$$\mathbf{E}_\mathrm{T} = -\frac{\partial \mathbf{A}_\mathrm{T}}{\partial t}, \tag{4}$$

while the Gauss law

$$\boldsymbol{\nabla} \cdot \mathbf{E} = \rho \tag{5}$$

involving the charge density ρ ensures that the longitudinal electric field is equal to minus the gradient of the (static) Coulomb potential $\mathbf{E}_\mathrm{L} = -\boldsymbol{\nabla} V$. Using this equality and Eq. (1) we see that the scalar potential accompanying the vector potential in Eq. (3) can be written

$$\phi = V - \frac{\partial f}{\partial t}. \tag{6}$$

The Coulomb gauge is defined by the choice $f \equiv 0$, but it is not the only way in which the longitudinal degrees of freedom can be eliminated. In nonrelativistic QED it has been shown [10] that a general gauge-fixing condition is given by a linear functional constraint satisfied by vector potentials of the form

$$\mathbf{A} = \mathbf{A}_\mathrm{T} + \boldsymbol{\nabla} \int d^3x' \boldsymbol{g}(\mathbf{x},\mathbf{x}') \cdot \mathbf{A}_\mathrm{T}(\mathbf{x}'), \tag{7}$$

where $\boldsymbol{g}(\mathbf{x},\mathbf{x}')$ is the Green's function for the divergence operator

$$\boldsymbol{\nabla} \cdot \boldsymbol{g}(\mathbf{x},\mathbf{x}') = \delta(\mathbf{x} - \mathbf{x}'). \tag{8}$$

In Eq. (7) the redundant degrees of freedom have been reexpressed through a functional of the gauge-invariant transverse degrees of freedom.

While the longitudinal component of the Green's function $\boldsymbol{g}$ is fixed according to Eq. (8) by

$$\boldsymbol{g}_\mathrm{L}(\mathbf{x},\mathbf{x}') = -\boldsymbol{\nabla} \frac{1}{4\pi |\mathbf{x} - \mathbf{x}'|}, \tag{9}$$

its transverse component is essentially arbitrary, meaning that the gauge is determined through a choice of $\boldsymbol{g}_\mathrm{T}$. This idea has been employed in nonrelativistic QED to obtain a Hamiltonian in an arbitrary gauge [10], but it has yet to be extended to the relativistic setting. Furthermore, the nature of gauge transformations and residual symmetries in such a framework has not been explored.

B. The QED Lagrangian and Hamiltonian

We start formally with the QED Lagrangian density

$$\mathscr{L} = i\gamma_0\gamma^\mu \psi^\dagger D_\mu \psi - (\gamma_0 m + e\phi_e)\psi^\dagger\psi - \tfrac{1}{4} F_{\mu\nu}F^{\mu\nu}, \tag{10}$$

where $D_\mu = \partial_\mu + ieA_\mu$ is the gauge-covariant derivative, $F_{\mu\nu} = \partial_\mu A_\nu - \partial_\nu A_\mu$ is the electromagnetic field strength tensor, and ϕ_e is an external potential due, for example, to nuclei. Since the Lagrangian is independent of the velocity of the scalar potential its conjugate momentum is identically zero. As a result, it is natural to quantize the theory within the Weyl gauge corresponding to the choice $\phi \equiv 0$. The remaining redundant degrees of freedom are eliminated by defining the physical subspace of states $\mathcal{H}_p$ consisting of those states, which vanish under the action of the Gauss law constraint $G|\varphi_p\rangle \equiv (\boldsymbol{\nabla} \cdot \mathbf{E} - \rho)|\varphi_p\rangle = 0$.

The Hamiltonian density is obtained from the Lagrangian density via a Legendre transformation

$$\begin{aligned} \mathscr{H} = &-i\psi^\dagger \boldsymbol{\alpha} \cdot (\boldsymbol{\nabla} - ie\mathbf{A})\psi + (\beta m + e\phi_e)\psi^\dagger\psi \\ &+ \tfrac{1}{2}[\boldsymbol{\Pi}^2 + (\boldsymbol{\nabla} \times \mathbf{A})^2]. \end{aligned} \tag{11}$$

Quantum mechanically ψ and its conjugate $\psi^\dagger$ are Dirac field operators satisfying the anticommutation relation

$$\{\psi(\mathbf{x}), \psi^\dagger(\mathbf{x}')\} = \delta(\mathbf{x} - \mathbf{x}'), \tag{12}$$

while $\mathbf{A}$ and $\mathbf{\Pi} = -\mathbf{E}$ are the canonical operators of the Maxwell field satisfying the commutation relation

$$[A_i(\mathbf{x}), \Pi_j(\mathbf{x}')] = i\delta_{ij}\delta(\mathbf{x}-\mathbf{x}'). \tag{13}$$

We employ the usual definitions of the charge and current densities

$$\begin{aligned} \rho &= e\psi^\dagger\psi, \\ \mathbf{j} &= e\psi^\dagger\boldsymbol{\alpha}\psi, \end{aligned} \tag{14}$$

in terms of which the (conserved) Noether four-current is $j^\mu = (\rho,\mathbf{j})$. Related to the charge density is the polarization field $\mathbf{P}_g$ defined by

$$\mathbf{P}_g(\mathbf{x}) = -\int d^3x'\, \boldsymbol{g}(\mathbf{x},\mathbf{x}')\rho(\mathbf{x}'), \tag{15}$$

whose longitudinal component satisfies

$$-\boldsymbol{\nabla}\cdot\mathbf{P}_g = \rho, \tag{16}$$

but whose transverse component is arbitrarily determined by $\boldsymbol{g}_\mathrm{T}$. Equation (16) bares a close resemblance to the Gauss law G, which can indeed be written

$$G = \boldsymbol{\nabla}\cdot\mathbf{\Pi} + \rho = \boldsymbol{\nabla}\cdot(\mathbf{\Pi} - \mathbf{P}_g). \tag{17}$$

As an operator G is a symmetry of the Hamiltonian $[G,H] = 0$ and is responsible for generating time-independent gauge transformations of the vector potential and Dirac field operators. Identifying a group $(\{\beta(\mathbf{x})\}, +)$ consisting of real-valued functions on $\mathbb{R}^3$ and group operation of addition, we define a group action Φ acting on the vector potential and Dirac fields by

$$\begin{aligned} \Phi[\psi,\beta] &= e^{-ie\beta}\psi, \\ \Phi[\mathbf{A},\beta] &= \mathbf{A} + \boldsymbol{\nabla}\beta. \end{aligned} \tag{18}$$

The action is implemented through unitary transformations Ω generated by G, viz., $\Omega\psi\Omega^{-1} = \Phi[\psi,\beta]$ and $\Omega\mathbf{A}\Omega^{-1} = \Phi[\mathbf{A},\beta]$, where

$$\Omega[\beta] = \exp\left(i\int d^3x(\mathbf{\Pi}\cdot\boldsymbol{\nabla} + \rho)\beta(\mathbf{x})\right). \tag{19}$$

These transformations are called *residual* gauge transformations, with the word residual intended to signify that the above time-independent symmetry is what remains of the local gauge symmetry present in the original formulation [12].

C. Unitary gauge-fixing transformations

We now turn our attention to the procedure of gauge fixing. In order to determine the form of a general gauge-fixing transformation we first need to identify the form of a physical state. To do this we take as a Hilbert space $\mathcal{H}$ for the composite system wave functionals $\varphi[\mathbf{A}]$ of the c-number vector potential $\mathbf{A}$, which take values in the Hilbert space of the Dirac field operators [11].

A realization of the algebra of the Maxwell field operators $\mathbf{A}$ and $\mathbf{\Pi}$ is given on $\mathcal{H}$ using the coordinate representation

$$\begin{aligned} (\hat{\mathbf{A}}\varphi)[\mathbf{A}] &= \mathbf{A}\varphi[\mathbf{A}], \\ (\hat{\mathbf{\Pi}}\varphi)[\mathbf{A}] &= -i\frac{\delta\varphi[\mathbf{A}]}{\delta\mathbf{A}}, \end{aligned} \tag{20}$$

where we have introduced carets to distinguish between operators and c-number vector fields. Defining a scalar function α by $\boldsymbol{\nabla}\alpha = \mathbf{A}_\mathrm{L}$, we can vary the wave functional φ with respect to α and make use of Eq. (20) to obtain

$$i\frac{\delta\varphi}{\delta\alpha} = -\boldsymbol{\nabla}\cdot\hat{\mathbf{\Pi}}\varphi. \tag{21}$$

Using the constraint G in Eq. (17), we get for a physical state φ_p

$$i\frac{\delta\varphi_p}{\delta\alpha} = \rho\varphi_p \tag{22}$$

and finally solving this equation gives the general form of a physical state

$$\begin{aligned} \varphi_p[\mathbf{A}] &= \varphi_p[\mathbf{A}_\mathrm{T} + \boldsymbol{\nabla}\alpha] \\ &= \exp\left(-i\int d^3x\,\alpha(\mathbf{x})\rho(\mathbf{x})\right)\varphi_p[\mathbf{A}_\mathrm{T}]. \end{aligned} \tag{23}$$

Having determined the form of a physical state, we can begin to define some unitary gauge-fixing transformations. In the original work of Lenz *et al.* [12] a unitary gauge-fixing transformation yielding the Coulomb gauge representation was given as

$$U \equiv \exp\left(i\int d^3x\,\hat{\alpha}(\mathbf{x})\rho(\mathbf{x})\right), \tag{24}$$

where $\hat{\alpha}$ is defined analogously to α by $\boldsymbol{\nabla}\hat{\alpha} = \hat{\mathbf{A}}_\mathrm{L}$. In the present context we see clearly that U eliminates the dependence of the physical state on $\mathbf{A}_\mathrm{L}$,

$$(U\varphi_p)[\mathbf{A}] = \varphi_p[\mathbf{A}_\mathrm{T}]. \tag{25}$$

Now, as in Eq. (7) of Sec. II A, we write the longitudinal vector potential as the gradient of a functional of the transverse vector potential

$$\mathbf{A}_\mathrm{L} = \boldsymbol{\nabla}\chi_g(\mathbf{x},[\mathbf{A}_\mathrm{T}]), \tag{26}$$

where

$$\chi_g(\mathbf{x},[\mathbf{A}_\mathrm{T}]) = \int d^3x'\,\boldsymbol{g}(\mathbf{x},\mathbf{x}')\cdot\mathbf{A}_\mathrm{T}(\mathbf{x}'). \tag{27}$$

We can then define a more general unitary gauge-fixing transformation U_g by

$$U_g \equiv \exp\left(i\int d^3x\{\hat{\alpha}(\mathbf{x}) - \chi_g(\mathbf{x},[\hat{\mathbf{A}}_\mathrm{T}])\}\rho(\mathbf{x})\right), \tag{28}$$

mapping from $\mathcal{H}_p$ to a space denoted $\mathcal{H}_g$, which is the space of states for the gauge g,

$$\begin{aligned} (U_g\varphi_p)[\mathbf{A}] &= \exp\left(-i\int d^3x\,\chi_g(\mathbf{x},[\mathbf{A}_\mathrm{T}])\rho(\mathbf{x})\right)\varphi_p[\mathbf{A}_\mathrm{T}] \\ &= \varphi_p[\mathbf{A}_\mathrm{T} + \boldsymbol{\nabla}\chi_g] \equiv \varphi_g[\mathbf{A}_\mathrm{T}] \in \mathcal{H}_g. \end{aligned} \tag{29}$$

The (transverse component of the) Green's function $\boldsymbol{g}$ is essentially arbitrary and determines the gauge. Two commonly used examples are the Coulomb gauge $\boldsymbol{g}_{\rm T} \equiv 0$ and the Poincaré gauge $g_{{\rm T},j}(\mathbf{x},\mathbf{x}') \equiv -\int_0^1 d\lambda x_i \delta^{\rm T}_{ij}(\mathbf{x}' - \lambda\mathbf{x})$ [2,4]. The vector potential operator in the gauge g is $\hat{\mathbf{A}}(\mathbf{x}) \equiv \hat{\mathbf{A}}_{\rm T}(\mathbf{x}) + \boldsymbol{\nabla}\chi_g(\mathbf{x},[\hat{\mathbf{A}}_{\rm T}])$ with action on $\mathcal{H}_g$ given by

$$(\hat{\mathbf{A}}\varphi_g)[\mathbf{A}_{\rm T}] = (\mathbf{A}_{\rm T} + \boldsymbol{\nabla}\chi_g)\varphi_g[\mathbf{A}_{\rm T}]. \tag{30}$$

Finally we define a unitary transformation from a fixed gauge g to a fixed gauge g' by

$$U_{gg'} \equiv \exp\left(i\int d^3x\{\chi_g(\mathbf{x},[\hat{\mathbf{A}}_{\rm T}]) - \chi_{g'}(\mathbf{x},[\hat{\mathbf{A}}_{\rm T}])\}\rho(\mathbf{x})\right), \tag{31}$$

an example of which is the well known Power-Zienau-Woolley transformation [2–4] used to obtain the Hamiltonian in the Poincaré gauge from the Hamiltonian in the Coulomb gauge. Such a gauge transformation is not to be confused with the residual gauge (symmetry) transformation given in Eq. (19).

D. The Hamiltonian in the gauge g

To obtain the Hamiltonian in the gauge g we need to determine the effect of the transformation in Eq. (28) on the various operators of the theory, namely, ψ, $\psi^\dagger$, $\mathbf{A}$, and $\boldsymbol{\Pi}$. In doing so we will resume denoting operators without carets. Clearly U_g leaves the vector potential $\mathbf{A}$ unchanged, while the action of $\hat{\mathbf{A}}$ on $\mathcal{H}_g$ is given in Eq. (30). The effect on the Dirac field operator ψ is that of a gauge transformation

$$U_g\psi U_g^{-1} = e^{ie(\alpha-\chi_g)}\psi. \tag{32}$$

The canonical momentum $\boldsymbol{\Pi}$ transforms as

$$U_g\boldsymbol{\Pi}U_g^{-1} = \boldsymbol{\Pi} + \mathbf{P}_g, \tag{33}$$

so that in the new representation $\boldsymbol{\Pi}$ represents the (negative of) the gauge-dependent displacement operator $\mathbf{D}_g \equiv \mathbf{E} + \mathbf{P}_g$. Using Eq. (16) we find the constraint G and the residual gauge transformation Ω transform as

$$\begin{aligned} U_g G U_g^{-1} &= \boldsymbol{\nabla}\cdot\boldsymbol{\Pi}, \\ U_g\Omega[\beta]U_g^{-1} &= \exp\left(i\int d^3x(\boldsymbol{\Pi}\cdot\boldsymbol{\nabla})\beta(\mathbf{x})\right), \end{aligned} \tag{34}$$

which are independent of the gauge g. The constraint G implies that the longitudinal canonical momentum $\boldsymbol{\Pi}_{\rm L}$ vanishes on $\mathcal{H}_g$. On the one hand this means $\mathbf{P}_{\rm L}$ alone represents (the negative of) the longitudinal electric field and on the other that the Hamiltonian density on $\mathcal{H}_g$ can be written in terms of the transverse operators $\mathbf{A}_{\rm T}$ and $\boldsymbol{\Pi}_{\rm T}$ only,

$$\begin{aligned} \mathscr{H} = {} & -i\psi^\dagger\boldsymbol{\alpha}\cdot[\boldsymbol{\nabla} - ie(\mathbf{A}_{\rm T} + \boldsymbol{\nabla}\chi_g)]\psi + (\beta m + e\phi_e)\psi^\dagger\psi \\ & + \tfrac{1}{2}(\mathbf{P}_{\rm L})^2 + \tfrac{1}{2}\left[\left(\boldsymbol{\Pi}_{\rm T} + \mathbf{P}^g_{\rm T}\right)^2 + (\boldsymbol{\nabla}\times\mathbf{A}_{\rm T})^2\right], \end{aligned} \tag{35}$$

where $\mathbf{A}_{\rm T} + \boldsymbol{\nabla}\chi_g$ is simply the vector potential $\mathbf{A}$ in the gauge g. Equation (35) gives a Hamiltonian in an arbitrary gauge, which is fully relativistic in the material degrees of freedom. It is one of the main results of this paper.

The commutator of the transverse operators follows from Eq. (13) and is given by

$$[A_{{\rm T},i}(\mathbf{x}), \Pi_{{\rm T},j}(\mathbf{x}')] = i\delta^{\rm T}_{ij}(\mathbf{x} - \mathbf{x}'), \tag{36}$$

with $\delta^{\rm T}$ denoting the transverse delta function. We note also that by denoting the Fourier transforms of $\mathbf{A}_{\rm T}$ and $\boldsymbol{\Pi}_{\rm T}$ with tildes we can define photon creation and annihilation operators in the usual way

$$a_\lambda(\mathbf{k}) = \sqrt{\frac{1}{2\omega}}[\omega\tilde{A}_{{\rm T},\lambda}(\mathbf{k}) + i\tilde{\Pi}_{{\rm T},\lambda}(\mathbf{k})], \tag{37}$$

where $\lambda = 1,2$ denotes one of two polarization directions orthogonal to $\mathbf{k}$. The bosonic commutator

$$[a_\lambda(\mathbf{k}), a^\dagger_{\lambda'}(\mathbf{k}')] = \delta_{\lambda\lambda'}\delta(\mathbf{k} - \mathbf{k}') \tag{38}$$

follows from Eq. (36).

E. The Dirac equation in the gauge g

It is an instructive exercise to calculate in the arbitrary gauge g the equation of motion for the Dirac field operator ψ, which should be the Dirac equation in the presence of a Maxwell field. The calculation demonstrates how the scalar potential, like the longitudinal vector potential, is reexpressed through the functional χ_g.

Writing first the products of Dirac field operators appearing in Eq. (35) in normal order, we obtain

$$\begin{aligned} i\dot{\psi} = {} & \Bigg[\boldsymbol{\alpha}\cdot[-i\boldsymbol{\nabla} - e(\mathbf{A}_{\rm T} + \boldsymbol{\nabla}\chi_g)] + \beta m + e\phi_e \\ & + \frac{e}{4\pi}\int d^3x'\frac{\rho(\mathbf{x}')}{|\mathbf{x} - \mathbf{x}'|} \\ & - e\int d^3x'\boldsymbol{g}_{\rm T}(\mathbf{x},\mathbf{x}')\cdot\left(\boldsymbol{\Pi}_{\rm T} + \mathbf{P}^g_{\rm T}\right)\Bigg]\psi. \end{aligned} \tag{39}$$

The first term on the second line of Eq. (39) is equal to eV, with V denoting the static Coulomb potential of charges. The transverse electric field in the gauge g is $\mathbf{E}_{\rm T} = -(\boldsymbol{\Pi}_{\rm T} + \mathbf{P}^g_{\rm T})$ and it is straightforward to verify that $\mathbf{E}_{\rm T} = -\dot{\mathbf{A}}_{\rm T}$, as in Eq. (4). These equalities imply that Eq. (39) is indeed the Dirac equation

$$i\dot{\psi} = [\boldsymbol{\alpha}\cdot(-i\boldsymbol{\nabla} - e\mathbf{A}) + \beta m + e(\phi_e + \phi)]\psi, \tag{40}$$

where we have defined the scalar potential anew by

$$\phi = V - \frac{\partial\chi_g}{\partial t}, \tag{41}$$

which is analogous to Eq. (6).

III. IMPLICATIONS OF THE FORMALISM

Having obtained the Hamiltonian in an arbitrary gauge, we discuss in this section some implications resulting from the freedom to choose $\boldsymbol{g}_{\rm T}$. In Sec. III A we point out that as in the nonrelativistic case a canonical partitioning of the Hamiltonian is gauge dependent and therefore leads to gauge-dependent definitions of quantum subsystems [10].

By means of analogy with classical electrodynamics, in Sec. III B we demonstrate how gauge-invariant subsystem components might be defined. Finally in Sec. III C, we consider

an application of such a definition in the context of energy transfer and causality in Fermi's two-atom problem.

A. Ambiguity in defining quantum systems

To understand what the arbitrariness of $\boldsymbol{g}_{\mathrm{T}}$ might mean with regard to the physical predictions of our formulation it is important to identify the physical observables in a given gauge g. In the Weyl gauge the canonical momentum $\mathbf{\Pi}$ is the negative of the electric field $\mathbf{E}$. In the gauge g we have $\mathbf{E} = -U_g \mathbf{\Pi} U_g^{-1} = -(\mathbf{\Pi} + \mathbf{P}_g)$. The operator $\mathbf{\Pi}_{\mathrm{T}} + \mathbf{P}_{\mathrm{T}}^g$ appearing in Eq. (35) therefore represents the negative of the transverse electric field $\mathbf{E}_{\mathrm{T}}$. This identity was used in obtaining the Dirac equation (40).

Due to the gauge dependence of $\mathbf{P}_{\mathrm{T}}^g$, the operator $\mathbf{\Pi}_{\mathrm{T}}$ is implicitly gauge dependent in that it represents a different physical observable in each different gauge. Explicitly $\mathbf{\Pi}_{\mathrm{T}}$ represents the transverse component of (the negative of) the gauge-dependent displacement operator $\mathbf{D}_g$.

Now, the most common way to use a Hamiltonian (density) such as Eq. (35) is to split it into free and interacting components as follows:

$$\mathscr{H} = \mathscr{H}_0 + \mathscr{H}_I, \tag{42}$$

where

$$\begin{aligned}
&\mathscr{H}_0 \equiv \mathscr{H}_D + \mathscr{H}_{\mathrm{EM}},\\
&\mathscr{H}_D \equiv -i\psi^\dagger \boldsymbol{\alpha}\cdot\boldsymbol{\nabla}\psi + (\beta m + e\phi_e)\psi^\dagger\psi + \tfrac{1}{2}(\mathbf{P}_{\mathrm{L}})^2 + \tfrac{1}{2}(\mathbf{P}_{\mathrm{T}}^g)^2,\\
&\mathscr{H}_M \equiv \tfrac{1}{2}\big[\mathbf{\Pi}_{\mathrm{T}}^2 + (\boldsymbol{\nabla}\times\mathbf{A}_{\mathrm{T}})^2\big],
\end{aligned} \tag{43}$$

and

$$\mathscr{H}_I \equiv -e\psi^\dagger \boldsymbol{\alpha}\cdot(\mathbf{A}_{\mathrm{T}} + \boldsymbol{\nabla}\chi_g)\psi + \mathbf{\Pi}_{\mathrm{T}}\cdot\mathbf{P}_{\mathrm{T}}^g. \tag{44}$$

The reason for this splitting is of course that the sets of operators $\{\psi,\psi^\dagger\}$ and $\{\mathbf{A}_{\mathrm{T}},\mathbf{\Pi}_{\mathrm{T}}\}$ are mutually commuting. The component $\mathscr{H}_D$ represents the Dirac field subsystem, $\mathscr{H}_M$ represents the Maxwell field subsystem, and $\mathscr{H}_I$ represents their interaction. The problem with such a splitting is that the subsystem components so defined are like the operator $\mathbf{\Pi}_{\mathrm{T}}$, implicitly gauge dependent and as a result physically ambiguous.

Given this ambiguity it is natural to try and determine what kind of calculations can be carried out that yield results independent of g, i.e., that are gauge invariant. For example, S-matrix elements on energy shell are gauge invariant to all orders in perturbation theory [14,15]. Consequently, the conceptual difficulty regarding the definition of subsystems does not effect scattering theory in any way. The invariance of S-matrix elements rests on the fact that the bare states (eigenstates of the free-energy operator H_0) coincide asymptotically with eigenstates of the total Hamiltonian, that is, that bare states are asymptotically stable.

In order that conventional (perturbative) calculations using bare states produce gauge-invariant results a condition of free-energy conservation must be satisfied [14]. Otherwise calculations will in general yield gauge-dependent results. Free-energy conservation is a condition that must be imposed from outside the theory, so various approximations, which essentially ensure free-energy conservation by giving rise to a δ function in the initial energy minus the final energy of the process under study, are used throughout nonrelativistic QED and quantum optics; they include the resonant state on an energy shell approximation [14,16], the pole approximation [17], the Markovian approximation [17], and the Fermi approximations [3,6].

A general survey of the types of calculation used in practice in nonrelativistic QED and their dependence on g can be found in Ref. [14]. An example of the difference in predicted results from calculations in two different gauges is well known in nonrelativistic QED where the Coulomb gauge (minimal coupling) Hamiltonian and Poincaré gauge (multipolar) Hamiltonian yield different results for, among other things, the theoretical lineshape of spontaneous emission [4]. The source of this difference lies in the use of physically different canonical operators in determining the lineshape observable.

The ambiguity regarding subsystems defined using canonical variables is also present at the classical level. It occurs when moving to the Hamiltonian formalism from the Lagrangian formalism. Since canonical momenta are defined in terms of the Lagrangian, equivalent Lagrangians yielding equivalent Hamiltonians will in general not yield physically equivalent canonical momenta. Again, a well known example is given by the Coulomb gauge and Poincaré gauge formulations of classical electrodynamics.

At the quantum level the ambiguity in the definition of subsystems can be viewed as a generic trait of interacting theories, whether they are relativistic, field theoretic, or otherwise. Given a Hamiltonian dependent on two sets of mutually commuting operators $\{x_i\}$ and $\{y_i\}$ and a splitting of the Hamiltonian of the form $H = H_x(\{x_i\}) + H_y(\{y_i\}) + H_{xy}(\{x_i,y_i\})$, an equivalent Hamiltonian is obtained by a unitary transformation $H' = UHU^{-1}$. In general, the subsystem components of H' will not be equivalent to those of H, i.e., $H'_x \neq UH_xU^{-1}$, with the same being true for H_y and H_{xy}. The importance of this fact for concepts such as quantum entanglement and decoherence has been recognized in the philosophy literature [18].

B. Classical-type gauge-invariant subsystems and operators

In the preceding section we discussed the gauge dependence of splitting the Hamiltonian into free and interaction components and we reviewed the situation regarding the kinds of calculation that yield gauge-invariant results. In this section we address the complimentary question as to whether *manifestly* gauge-invariant subsystem components can be defined from the outset. This question has so far received no direct attention in the literature, but it is important if one wishes to identify how gauge-invariant results might be obtained outside the range of validity of the simplifying assumptions of scattering theory.

To see how we might define gauge-invariant subsystems consider first the case of a free classical electron coupled to a classical Maxwell field. For this system the Hamiltonian in *any* gauge can be written

$$H = \frac{1}{2}m\dot{\mathbf{r}}^2 + \frac{1}{2}\int d^3\mathbf{x}[\mathbf{E}(\mathbf{x})^2 + \mathbf{B}(\mathbf{x})^2], \tag{45}$$

which represents the total energy of the system as the sum of the kinetic energy of the electron and the energy of the

electromagnetic (EM) field. Regarding the electron variables, what varies between gauges is the identification of the gauge-invariant velocity $\dot{\mathbf{r}}$, with the electron canonical momentum $\mathbf{p} = m\dot{\mathbf{r}} - e\mathbf{A}(\mathbf{r})$ [3,10,19].

The classical electron velocity and canonical momentum have clear analogs in QED; the velocity density

$$\mathscr{V} = -i\psi^\dagger \boldsymbol{\alpha}\cdot[\boldsymbol{\nabla} - ie(\mathbf{A}_\mathrm{T} + \boldsymbol{\nabla}\chi_g)]\psi \tag{46}$$

is manifestly gauge invariant, while the canonical momentum density $\mathscr{P} = -i\psi^\dagger\boldsymbol{\alpha}\cdot\boldsymbol{\nabla}\psi$ depends on g. Analogously to Eq. (45), we can split the Hamiltonian into two gauge-invariant components

$$\begin{aligned}\mathscr{H} &= \mathscr{H}_M + \mathscr{H}_\mathrm{EM},\\ \mathscr{H}_M &= -i\psi^\dagger\boldsymbol{\alpha}\cdot[\boldsymbol{\nabla} - ie(\mathbf{A}_\mathrm{T} + \boldsymbol{\nabla}\chi_g)]\psi\\ &\quad + (\beta m + e\phi_e)\psi^\dagger\psi,\\ \mathscr{H}_\mathrm{EM} &= \tfrac{1}{2}(\mathbf{P}_\mathrm{L})^2 + \tfrac{1}{2}\left[\left(\boldsymbol{\Pi}_\mathrm{T} + \mathbf{P}^g_\mathrm{T}\right)^2 + (\boldsymbol{\nabla}\times\mathbf{A}_\mathrm{T})^2\right],\end{aligned} \tag{47}$$

with the first component representing the energy density of the matter field and the second component the energy density of the EM field. With these definitions the Hamiltonian naturally represents the energy of the system as the sum of energies of the subsystems rather than as the sum of superficially defined free and interaction energies.

We note that the first term in $\mathscr{H}_\mathrm{EM}$ represents the energy density of the longitudinal EM field, while the second represents the energy density of the transverse EM field. The subsystems defined in this way are coupled because the velocity density $\mathscr{V}$ and the electric field $\mathbf{E}$ do not commute; using for simplicity the Coulomb gauge ($\boldsymbol{g}_\mathrm{T} \equiv 0$), we obtain

$$\begin{aligned}&[\mathscr{V}(\mathbf{x}), E_j(\mathbf{x}')]\\ &\quad= [i\psi^\dagger(\mathbf{x})\boldsymbol{\alpha}\cdot[\boldsymbol{\nabla} - ie\mathbf{A}_\mathrm{T}(\mathbf{x})]\psi(\mathbf{x}), \Pi_{\mathrm{T},j}(\mathbf{x}') + P_{\mathrm{L},j}(\mathbf{x}')]\\ &\quad= ie\psi^\dagger(\mathbf{x})\alpha_i\left(\delta^\mathrm{T}_{ij}(\mathbf{x}-\mathbf{x}') - \nabla_i\nabla_j\frac{1}{4\pi|\mathbf{x}-\mathbf{x}'|}\right)\psi(\mathbf{x})\\ &\quad\equiv ie\psi^\dagger(\mathbf{x})\alpha_i\delta_{ij}\delta(\mathbf{x}-\mathbf{x}')\psi(\mathbf{x}) \equiv ij_j(\mathbf{x})\delta(\mathbf{x}-\mathbf{x}').\end{aligned} \tag{48}$$

The δ function ensures that $\mathscr{V}(\mathbf{x})$ and $\mathbf{E}(\mathbf{x}')$ are compatible observables for $\mathbf{x} \neq \mathbf{x}'$. Moreover it ensures that the matter field and the EM field energies are compatible in disjoint regions $\mathcal{R}$ and $\mathcal{R}'$ in $\mathbb{R}^3$:

$$\left[H^\mathcal{R}_M, H^{\mathcal{R}'}_\mathrm{EM}\right] \equiv \int_\mathcal{R} d^3x \int_{\mathcal{R}'} d^3x'[\mathscr{H}_M(\mathbf{x}), \mathscr{H}_\mathrm{EM}(\mathbf{x}')] = 0. \tag{49}$$

Using the commutator Eq. (48) and the commutator of the electric field and magnetic field energy,

$$\frac{1}{2}\int d^3x'[E_i(\mathbf{x}), B_j(\mathbf{x}')^2] = i\epsilon_{ijk}\nabla_j B_k(\mathbf{x}), \tag{50}$$

the equation of motion for the electric field is found to be

$$\dot{\mathbf{E}} = \boldsymbol{\nabla}\times\mathbf{B} - \mathbf{j}, \tag{51}$$

which is just one of Maxwell's equations. The remaining Maxwell equation

$$\dot{\mathbf{B}} = -\boldsymbol{\nabla}\times\mathbf{E} \tag{52}$$

is found in a similar fashion as the equation of motion for the magnetic field.

C. Energy and causality

In recent years a great deal of attention has been paid to the nature of energy transfer between separated material systems [20–25], specifically in the context of the two-atom problem of Fermi [13]. In order to investigate causality at the microscopic level, Fermi considered two identical spatially separated atoms A and B. Initially atom A is energetically excited while atom B is in its ground state and there are no photons present in the EM field. The question posed by Fermi was; when does atom B begin to move out of its ground state due to atom A? Einstein causality would appear to require that any changes in the energy of atom B be independent of atom A for all times less than the time it would take for a signal produced by atom A traveling at the maximal speed of propagation c to reach atom B. The most recent work concerning the Fermi problem was the proposal of a circuit QED experiment designed to test for any possible violations of causality [24].

The majority of theoretical proofs of causality in the Fermi problem involve using the bare states of a nonrelativistic Hamiltonian in the Poincaré gauge and the electric dipole approximation (EDA) [21–24]. The EDA dictates that the atoms in the Fermi problem couple to the Maxwell field at the c-number atomic center-of-mass positions. The problem itself can then be formulated in terms of a well defined center-of-mass separation. Moreover, the EDA ensures that the Poincaré gauge dipole canonical momenta $\mathbf{p}_i = m\dot{\mathbf{r}}_i, i = A, B$ which define the bare atomic energies, are purely kinetic [2,3].

At the same time in the Poincaré gauge the field canonical momentum is identified as the (negative of the) local multipolar transverse displacement field [2,3]. Outside the atoms, which in the EDA means away from the center-of-mass positions, this field coincides with the retarded electric field. The bare energies of the atoms are coupled through this field, which ensures that there are no violations of causality.

A different proof presented in Ref. [25] uses the abstract language of algebraic quantum-field theory, relying quite generally on the primitively causal nature of relativistic quantum field theory resulting from the hyperbolicity of the relevant equations of motion. Here we will show using the gauge-invariant definition of $\mathscr{H}_M$ in Eq. (47) that changes in the energy density of the matter field at a point $(t,\mathbf{x})$ are independent of the matter field at all points, which cannot be connected to $(t,\mathbf{x})$ by a causal signal. The energy of the matter field in some closed region $\mathcal{R} \subset \mathbb{R}^3$ is merely

$$H^\mathcal{R}_M(t) = \int_\mathcal{R} d^3x\, \mathscr{H}_M(\mathbf{x},t). \tag{53}$$

Since $\mathscr{H}_M$ is gauge invariant the result does not rely on the use of a particular gauge and avoids any approximations.

We begin by calculating the equation of motion for $\mathscr{H}_M$, which for simplicity is carried out in the Coulomb gauge

$$\begin{aligned}i\dot{\mathscr{H}}_M(\mathbf{x}) &= -\frac{1}{2}\int d^3x'\{[e\psi^\dagger(\mathbf{x})\alpha_i A_{\mathrm{T},i}(\mathbf{x})\psi(\mathbf{x}), \Pi_{\mathrm{T},j}(\mathbf{x}')^2]\\ &\quad + [i\psi^\dagger(\mathbf{x})\alpha_i\nabla_i\psi(\mathbf{x}), P_{\mathrm{L},j}(\mathbf{x}')^2]\}\\ &= ij_i(\mathbf{x})[E_{\mathrm{T},i}(\mathbf{x}) + E_{\mathrm{L},i}(\mathbf{x})] = ij_i(\mathbf{x})E_i(\mathbf{x}).\end{aligned} \tag{54}$$

Thus, together with the Maxwell equations (51) and (52) we have a system of equations, which can be written

$$\begin{aligned} \dot{\mathscr{H}}_M(\mathbf{x},t) &= \mathbf{j}(\mathbf{x},t)\cdot\mathbf{E}(\mathbf{x},t), \\ \mathbf{j}(\mathbf{x},t) &= \nabla\times\mathbf{B}(\mathbf{x},t) - \dot{\mathbf{E}}(\mathbf{x},t), \\ \Box\mathbf{E}(\mathbf{x},t) &= -\nabla\rho(\mathbf{x},t) - \dot{\mathbf{j}}(\mathbf{x},t), \\ \Box\mathbf{B}(\mathbf{x},t) &= \nabla\times\mathbf{j}(\mathbf{x},t), \end{aligned} \tag{55}$$

where $\Box$ is the d'Alembertian $\partial^2/\partial t^2 - \nabla^2$.

Now, first we note that using the second equation the first can be written in terms of the electric and magnetic fields alone. Second we note that the remaining two equations are inhomogeneous wave equations for the (Cartesian components of the) electric and magnetic fields with source terms $\boldsymbol{\nu}(\mathbf{x},t) \equiv \nabla\rho(\mathbf{x},t) + \dot{\mathbf{j}}(\mathbf{x},t)$ and $\boldsymbol{\mu}(\mathbf{x},t) \equiv -\nabla\times\mathbf{j}(\mathbf{x},t)$, respectively. These equations are hyperbolic and have well known retarded solutions of the form

$$\begin{aligned} \mathbf{E}(\mathbf{x},t) &= \mathbf{E}_0(\mathbf{x},t) + \mathbf{E}_r(\mathbf{x},t), \\ \mathbf{B}(\mathbf{x},t) &= \mathbf{B}_0(\mathbf{x},t) + \mathbf{B}_r(\mathbf{x},t), \end{aligned} \tag{56}$$

where $\mathbf{E}_0$ and $\mathbf{B}_0$ satisfy the homogeneous equations $\Box\mathbf{E}_0 = \Box\mathbf{B}_0 = 0$ and $\mathbf{E}_r$ and $\mathbf{B}_r$ depend respectively on the sources $\boldsymbol{\nu}(\mathbf{x}',t_r)$ and $\boldsymbol{\mu}(\mathbf{x}',t_r)$ at the retarded time $t_r = t - |\mathbf{x}-\mathbf{x}'|$ [26,27]. Using these solutions, we can conclude that as desired $\dot{\mathscr{H}}_M(\mathbf{x},t)$ at the point $(t,\mathbf{x})$ depends on the matter field at points $(t_r,\mathbf{x}')$ only.

IV. CONCLUSION

In this paper we have derived a Hamiltonian in an arbitrary noncovariant gauge, which could be taken as the starting point for the relativistic description of atoms and molecules. It also serves well as a means by which the gauge freedom of noncovariant canonical QED can be explored. We have discussed some implications of this gauge freedom and highlighted that a canonical partitioning of the Hamiltonian is manifestly gauge dependent. We have shown that a classical-type partitioning of the Hamiltonian in terms of velocities instead of canonical operators is gauge invariant and suggested a possible application of such a partitioning in relation to the problem of causality in spatially separated material systems.

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