Analytic sources of inequivalence of the velocity gauge and length gauge

Rainer Dick*

Department of Physics and Engineering Physics, University of Saskatchewan, 116 Science Place, Saskatoon, SK S7N 5E2, Canada

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It has been known for many years now that the descriptions of electromagnetic couplings in velocity gauge and length gauge can yield different results for atoms and molecules in strong fields. We point out that it is mathematically consistent to mix velocity gauge for some components of a material with length gauge for other components, although this should not be possible for a *bona fide* gauge transformation. For many-particle systems in a Hartree approximation, it is even possible to mix velocity gauge and length gauge for different particles of the same kind. Four main sources of analytic differences between velocity gauge and length gauge are then identified, and it is pointed out that these sources imply differences between velocity gauge and length gauge in particular for subfemtosecond spectroscopy, for experiments involving strong fields, and for chiral materials. Finally, it is emphasized that the transformation from velocity gauge to length gauge is just a particular example of a picture-changing unitary transformation. However, all these transformations lead to nonunitary shifts of the Hamiltonian, irrespective of whether the transformation can be described as a gauge transformation. Therefore, all these descriptions of quantum optics in dipole approximation are formally equivalent if agreement is achieved which particular formulation of the time-dependent interaction terms perturbs the "true" energy eigenstates of a system. However, this is where the actual discrepancies between velocity gauge, length gauge, and also other formulations such as acceleration gauge originate. This implies a generalization of the results of Galstyan *et al.* [Phys. Rev. A **93**, 023422 (2016)] from two different classes of theoretical formulations to many different classes of theoretical formulations.

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I. INTRODUCTION

It has been known for many decades [1,2] that describing matter-photon couplings in the dipole approximation yields different results for observables if a "velocity-gauge" interaction Hamiltonian

$$H_{Iv} = \int d^3\boldsymbol{x} \sum_i \left\{ \frac{i q_i \hbar}{2m_i} \boldsymbol{A}(t) \cdot [\psi_i^+(\boldsymbol{x},t) \overleftrightarrow{\nabla} \psi_i(\boldsymbol{x},t)] + \frac{q_i^2}{2m_i} \psi_i^+(\boldsymbol{x},t) \boldsymbol{A}^2(t) \psi_i(\boldsymbol{x},t) \right\} \tag{1}$$

is used instead of a "length-gauge" interaction Hamiltonian

$$H_{Il} = -\int d^3\boldsymbol{x} \sum_i q_i \psi_i^+(\boldsymbol{x},t) \boldsymbol{x} \cdot \boldsymbol{E}(t) \psi_i(\boldsymbol{x},t). \tag{2}$$

Indeed, a well-know textbook example is provided by the differential electron-photon-scattering cross section for scattering of photons with initial momentum $\hbar\boldsymbol{k}$ and polarization $\boldsymbol{\epsilon}(\boldsymbol{k})$ off electronic states $|n\rangle$. The velocity-gauge Hamiltonian (1) yields (see, e.g., Refs. [3,4])

$$\frac{d\sigma}{d\Omega} = \frac{\alpha_S^2}{c^2} \frac{k'}{k} \left| \frac{\hbar}{m_e} \delta_{n'n} \boldsymbol{\epsilon}'(\boldsymbol{k}') \cdot \boldsymbol{\epsilon}(\boldsymbol{k}) + \sum_{n''} \omega_{n',n''} \omega_{n'',n} \times \left(\frac{\langle n'|\boldsymbol{\epsilon}'(\boldsymbol{k}') \cdot \mathbf{x}|n''\rangle \langle n''|\boldsymbol{\epsilon}(\boldsymbol{k}) \cdot \mathbf{x}|n\rangle}{\omega_{n'',n} - ck - i\epsilon} + \frac{\langle n'|\boldsymbol{\epsilon}(\boldsymbol{k}) \cdot \mathbf{x}|n''\rangle \langle n''|\boldsymbol{\epsilon}'(\boldsymbol{k}') \cdot \mathbf{x}|n\rangle}{\omega_{n'',n} + ck' - i\epsilon} \right) \right|^2, \tag{3}$$

where $k' = k - (\omega_{n',n}/c)$ and the sum over virtual states $|n''\rangle$ includes integration over continuous quantum numbers.

On the other hand, the length-gauge Hamiltonian (2) yields the original Kramers–Heisenberg formula [5]

$$\frac{d\sigma}{d\Omega} = \alpha_S^2 c^2 k k'^3 \left| \sum_{n''} \left(\frac{\langle n'|\boldsymbol{\epsilon}'(\boldsymbol{k}') \cdot \mathbf{x}|n''\rangle \langle n''|\boldsymbol{\epsilon}(\boldsymbol{k}) \cdot \mathbf{x}|n\rangle}{\omega_{n'',n} - ck - i\epsilon} + \frac{\langle n'|\boldsymbol{\epsilon}(\boldsymbol{k}) \cdot \mathbf{x}|n''\rangle \langle n''|\boldsymbol{\epsilon}'(\boldsymbol{k}') \cdot \mathbf{x}|n\rangle}{\omega_{n'',n} + ck' - i\epsilon} \right) \right|^2. \tag{4}$$

The two expressions are practically equivalent if near-resonance conditions can be satisfied in the sense that there are nonvanishing transition matrix elements $\langle n'|\boldsymbol{\epsilon}'(\boldsymbol{k}') \cdot \mathbf{x}|n''\rangle$ and $\langle n''|\boldsymbol{\epsilon}(\boldsymbol{k}) \cdot \mathbf{x}|n\rangle$ with the properties $\omega_{n'',n} \simeq ck$ and $\omega_{n',n''} \simeq -ck'$, or if there are nonvanishing matrix elements $\langle n'|\boldsymbol{\epsilon}(\boldsymbol{k}) \cdot \mathbf{x}|n''\rangle$ and $\langle n''|\boldsymbol{\epsilon}'(\boldsymbol{k}') \cdot \mathbf{x}|n\rangle$ with the properties $\omega_{n'',n} \simeq -ck'$ and $\omega_{n',n''} \simeq ck$. Indeed, there will be observational bias towards observation of scattered photons where these near resonance conditions are met, while at the same time electronic energy-level systems are generically dense enough to meet these requirements. This implies equivalence of Eqs. (3) and (4) for all practical purposes. However, discrepancies between the predictions from velocity- and length-gauge Hamiltonians have become observationally relevant in strong-field systems. Indeed, it is known that the two Hamiltonians (1) and (2) can yield very different and occasionally contradictory results in strong electromagnetic fields [6–14]. In particular, for electron detachment in strong fields, authors have argued for velocity gauge on the basis of analytic advantages [15,16] and momentum conservation [17], whereas Schlicher *et al.* [2] and also Cohen–Tannoudji *et al.* [18] have emphasized the advantage of the formulation of length gauge in terms of kinetic momentum $m\boldsymbol{v}$ and electric field $\boldsymbol{E}(t)$, and many

*rainer.dick@usask.ca

studies in recent years report that length gauge yields results in better agreement with observations [6,7,14,19–24]. Yet again, for high-order harmonic generation in diatomic molecules at large internuclear separation, Chirilă and Lein found good agreement with the semiclassical three-step mechanism only in velocity gauge [25].

For another example, Zhang and Nakajima report discrepancies between velocity- and length-gauge predictions for the photoelectron angular distribution in hydrogen, in particular for elliptical polarization [8], while Majety *et al.* [26] report advantages in using length gauge at short distances and velocity gauge at longer distances for the calculation of photoelectron spectra from single-electron systems, helium, and hydrogen molecules.

Furthermore, Dong *et al.* find unphysical oscillations in the photoinduced carrier densities in graphene if they use length gauge, whereas velocity gauge does not exhibit that problem [11]. On the other hand, theoretical investigations both in velocity gauge [27–31] and in length gauge [32] show the possibility of strong third-harmonic generation in graphene, but Ishikawa [31] reports a reduction of the nonlinear response due to the interplay of interband and intraband dynamics in velocity gauge, whereas Al-Naib *et al.* find an increase in length gauge [32]. These observations shed light on another important difference between length gauge and velocity gauge in systems with energy bands: velocity gauge is diagonal in quasimomentum and therefore *prima facie* less sensitive to intraband effects, whereas length gauge is less sensitive to interband interactions. The choice of length gauge [33–35] versus velocity gauge [36,37] for the study of optical transitions and higher-harmonic generation in solids therefore also depends on a judicious estimate of relative importance of intraband and interband interactions.

The choice of interaction term naturally affects numerical implementations and calculational efficiencies for integrations of the time-dependent Schrödinger equation [9,10,38], but it is also important to recognize physical differences between the interaction Hamiltonians (1) and (2). Close inspection of the transformation which maps (1) into (2) demonstrates that the transformation can also be understood as a redefinition of the Schrödinger field without any transformation of the gauge fields. As a consequence, we will also find that the transformation can be selectively applied to different particle species, i.e., we could perform the transformation, e.g., only for electrons, but not for protons in a many-particle system, without violating any mathematical consistency conditions. Indeed, the transition from velocity gauge to length gauge is only a subset of picture-changing operations which also include, e.g., the transformation into "acceleration gauge" [18,39,40], and understanding the transition from velocity gauge to length gauge in this wider framework also helps to understand sources of differences between velocity gauge and length gauge. The advantage of an improved understanding of analytic sources of differences between those different pictures of time evolution illuminates the question why one picture can be better or worse than another picture in describing specific physical systems or effects.

The transformation between velocity and length gauges for a nonrelativistic many-particle system is reexamined in Sec. II from a first-principles perspective, which emphasizes the separation of internal short-wavelength components and external long-wavelength components in the dipole approximation. Selective application to different particle species will be discussed in Sec. III. The appearance of analytic differences between velocity gauge and length gauge in physical systems is first discussed in the single electron approximation in Sec. IV. Section V revisits the question of analytic differences between velocity gauge and length gauge in the wider scope of picture-changing operations, which also includes the transition to acceleration gauge. Section VI summarizes our conclusions.

II. DIPOLE APPROXIMATION AND THE TRANSITION TO LENGTH GAUGE

Quantum optics with photons in the sub-keV energy range is conveniently analyzed in dipole approximation for the radiation fields $\boldsymbol{E}_\gamma(\boldsymbol{x},t) \simeq \boldsymbol{E}_\gamma(t)$, $\boldsymbol{B}_\gamma(\boldsymbol{x},t) \simeq \boldsymbol{B}_\gamma(t)$, since wavelengths in excess of 10 nm do not resolve atomic or molecular length scales. Furthermore, the preferred gauge for the electromagnetic potentials in quantum optics of atomic or molecular systems is Coulomb gauge,

$$\nabla \cdot \boldsymbol{A}(\boldsymbol{x},t) = 0, \quad \Phi(\boldsymbol{x},t) = \frac{1}{4\pi\epsilon_0} \int d^3\boldsymbol{x}' \frac{\varrho(\boldsymbol{x}',t)}{|\boldsymbol{x}-\boldsymbol{x}'|}, \tag{5}$$

$$\left(\frac{1}{c^2}\frac{\partial^2}{\partial t^2} - \Delta\right)\boldsymbol{A}(\boldsymbol{x},t) = \mu_0 \boldsymbol{J}(\boldsymbol{x},t), \tag{6}$$

where the current $\boldsymbol{J}$ includes the contributions from the longitudinal components of the electric field, which cancel the longitudinal components from $\boldsymbol{j}$,

$$\boldsymbol{J}(\boldsymbol{x},t) = \boldsymbol{j}(\boldsymbol{x},t) - \epsilon_0 \frac{\partial}{\partial t} \nabla \Phi(\boldsymbol{x},t), \quad \nabla \cdot \boldsymbol{J}(\boldsymbol{x},t) = 0; \tag{7}$$

see, e.g., Ref. [41]. The vector potential is then

$$\boldsymbol{A}(\boldsymbol{x},t) = \boldsymbol{A}_J(\boldsymbol{x},t) + \boldsymbol{A}_\gamma(t), \tag{8}$$

where the contribution from the local charges is

$$\boldsymbol{A}_J(\boldsymbol{x},t) = \int d^3\boldsymbol{x}' \frac{\mu_0}{4\pi|\boldsymbol{x}-\boldsymbol{x}'|} \boldsymbol{J}\left(\boldsymbol{x}', t - \frac{|\boldsymbol{x}-\boldsymbol{x}'|}{c}\right), \tag{9}$$

and the freely evolving part $\boldsymbol{A}_\gamma(t)$ accounts for the external radiation fields

$$\boldsymbol{E}_\gamma(t) = -d\boldsymbol{A}_\gamma(t)/dt \tag{10}$$

and

$$\boldsymbol{B}_\gamma(t) = \nabla \times \boldsymbol{A}_\gamma(\boldsymbol{x},t)|_{\boldsymbol{k}\cdot\boldsymbol{x}\to 0}. \tag{11}$$

The advantage of the Coulomb gauge is therefore twofold: First, the Coulomb gauge explicitly accounts for atomic or molecular potentials and currents without the potential pitfall of double counting of electromagnetic interactions through longitudinal photon operators. Furthermore, the Coulomb gauge separates the long-wavelength radiation contribution $\boldsymbol{A}_\gamma(t)$ on the one hand from the local contributions $\Phi(\boldsymbol{x},t)$ and $\boldsymbol{A}_J(\boldsymbol{x},t)$ due to the charges in the atoms or molecules on the other hand.

We also note that the contributions from $\boldsymbol{A}_J(\boldsymbol{x},t)$ to Hamilton operators can usually be neglected: Atomic orbitals scale with electron charge at least like e^3, which implies that terms $e\boldsymbol{A}_J(\boldsymbol{x},t)$ from local vector potentials scale at least

like e^8. Therefore we omit the index γ for the radiation contributions from now on, $\boldsymbol{A}_\gamma(t) \equiv \boldsymbol{A}(t)$.

The semiclassical Hamiltonian in the Coulomb gauge and in the dipole approximation then takes the form

$$\begin{aligned} H_v = \int d^3\boldsymbol{x} \sum_i \Big(& \frac{1}{2m_i} \big\{ \hbar^2 \nabla \psi_i^+(\boldsymbol{x},t) \cdot \nabla \psi_i(\boldsymbol{x},t) \\ & + i q_i \hbar \boldsymbol{A}(t) \cdot [\psi_i^+(\boldsymbol{x},t) \overset{\leftrightarrow}{\nabla} \psi_i(\boldsymbol{x},t)] \\ & + q_i^2 \psi_i^+(\boldsymbol{x},t) \boldsymbol{A}^2(t) \psi_i(\boldsymbol{x},t) \big\} + \psi_i^+(\boldsymbol{x},t) V_i(\boldsymbol{x},t) \psi_i(\boldsymbol{x},t) \Big) \\ & + \frac{1}{2} \sum_{ij} \int d^3\boldsymbol{x} \int d^3\boldsymbol{x}' \psi_i^+(\boldsymbol{x},t) \psi_j^+(\boldsymbol{x}',t) \\ & \times V_{ij}(\boldsymbol{x}-\boldsymbol{x}',t) \psi_j(\boldsymbol{x}',t) \psi_i(\boldsymbol{x},t). \end{aligned} \tag{12}$$

This is the second-quantized Hamiltonian with respect to the matter fields $\psi_i(\boldsymbol{x},t)$ in the Heisenberg picture,

$$[\psi_i(\boldsymbol{x},t), \psi_j^+(\boldsymbol{x}',t)]_\pm = \delta_{ij} \delta(\boldsymbol{x}-\boldsymbol{x}'), \tag{13}$$

while the radiation fields are treated classically, e.g., as expectation values of photon operators with long-wavelength coherent photon states. Spin labels are suppressed, since the subleading Pauli terms are (as usual) not included in Eq. (12).

The single-particle potentials $V_i(\boldsymbol{x},t)$ include the contributions to the scalar potential $\Phi(\boldsymbol{x},t)$ (5) due to fixed charges (e.g., massive ion cores),

$$V_i(\boldsymbol{x},t) \supseteq \sum_I \frac{q_i Q_I}{4\pi\epsilon_0 |\boldsymbol{x}-\boldsymbol{X}_I|}, \tag{14}$$

while the two-particle interaction potentials contain the contributions to $\Phi(\boldsymbol{x},t)$ from the dynamical charged particles,

$$V_{ij}(\boldsymbol{x}-\boldsymbol{x}',t) \supseteq \frac{q_i q_j}{4\pi\epsilon_0 |\boldsymbol{x}-\boldsymbol{x}'|}, \tag{15}$$

see, e.g., Ref. [42] for extensive discussions of the roles of Coulomb potentials and asymptotic conditions in atomic collision theory and electron exchange processes.

The quantum optics Hamiltonian (12) with vector potentials $\boldsymbol{A}(\boldsymbol{x},t)$ would yield equations of motion

$$i\hbar \frac{\partial}{\partial t} \psi_i(\boldsymbol{x},t) = [\psi_i(\boldsymbol{x},t), H],$$

which are form invariant under gauge transformations

$$\begin{aligned} \psi_i'(\boldsymbol{x},t) &= \exp[i q_i \varphi(\boldsymbol{x},t)/\hbar] \psi_i(\boldsymbol{x},t), \\ \boldsymbol{A}'(\boldsymbol{x},t) &= \boldsymbol{A}(\boldsymbol{x},t) + \nabla \varphi(\boldsymbol{x},t), \\ V_i'(\boldsymbol{x},t) &= V_i(\boldsymbol{x},t) - q_i \frac{\partial}{\partial t} \varphi(\boldsymbol{x},t), \\ V_{ij}'(\boldsymbol{x}-\boldsymbol{x}',t) &= V_{ij}(\boldsymbol{x}-\boldsymbol{x}',t). \end{aligned}$$

However, as a consequence of the dipole approximation, the Schrödinger equations

$$\begin{aligned} i\hbar \frac{\partial}{\partial t} \psi_i(\boldsymbol{x},t) &= [\psi_i(\boldsymbol{x},t), H_v] \\ &= -\frac{[\hbar\nabla - i q_i \boldsymbol{A}(t)]^2}{2m_i} \psi_i(\boldsymbol{x},t) + V_i(\boldsymbol{x},t) \psi_i(\boldsymbol{x},t) \\ &\quad + \sum_j \int d^3\boldsymbol{x}' \psi_j^+(\boldsymbol{x}',t) V_{ij}(\boldsymbol{x}-\boldsymbol{x}',t) \\ &\quad \times \psi_j(\boldsymbol{x}',t) \psi_i(\boldsymbol{x},t), \end{aligned} \tag{16}$$

following from the Hamiltonian (12), are manifestly form invariant only under restricted gauge transformations where the gauge function $\varphi(\boldsymbol{x},t)$ is constrained to be at most linear in spatial coordinates,

$$\varphi(\boldsymbol{x},t) = \boldsymbol{a}(t) \cdot \boldsymbol{x} + b(t) \Rightarrow \boldsymbol{A}'(t) = \boldsymbol{A}(t) + \boldsymbol{a}(t). \tag{17}$$

These gauge transformations also trivially preserve Coulomb gauge.

The minimal coupling terms in the effective dipole Hamiltonian can therefore be absorbed into the Schrödinger fields through the particular transformation with gauge function $\varphi(\boldsymbol{x},t) = -\boldsymbol{x} \cdot \boldsymbol{A}(t)$,

$$\psi_i(\boldsymbol{x},t) \Rightarrow \psi_i^{(l)}(\boldsymbol{x},t) = \exp\left[-\frac{i}{\hbar} q_i \boldsymbol{x} \cdot \boldsymbol{A}(t) \right] \psi_i(\boldsymbol{x},t), \tag{18}$$

$$\boldsymbol{A}(t) \Rightarrow \boldsymbol{A}'(t) = \boldsymbol{A}(t) - \nabla[\boldsymbol{x} \cdot \boldsymbol{A}(t)] = 0, \tag{19}$$

$$\begin{aligned} V_i(\boldsymbol{x},t) \Rightarrow V_i'(\boldsymbol{x},t) &= V_i(\boldsymbol{x},t) + q_i \frac{\partial}{\partial t} \boldsymbol{x} \cdot \boldsymbol{A}(t) \\ &= V_i(\boldsymbol{x},t) - q_i \boldsymbol{x} \cdot \boldsymbol{E}(t). \end{aligned} \tag{20}$$

The transformation (18) preserves the canonical (anti-) commutation relations (13).

The resulting Hamiltonian in terms of the *new* Schrödinger fields and the *old* gauge fields and potentials is [after dropping the label (l) for the Schrödinger fields in length gauge]

$$\begin{aligned} H_l = \int d^3\boldsymbol{x} \sum_i \Big(& \frac{\hbar^2}{2m_i} \nabla \psi_i^+(\boldsymbol{x},t) \cdot \nabla \psi_i(\boldsymbol{x},t) \\ & + \psi_i^+(\boldsymbol{x},t) V_i(\boldsymbol{x},t) \psi_i(\boldsymbol{x},t) \\ & - q_i \psi_i^+(\boldsymbol{x},t) \boldsymbol{x} \cdot \boldsymbol{E}(t) \psi_i(\boldsymbol{x},t) \Big) \\ & + \frac{1}{2} \sum_{ij} \int d^3\boldsymbol{x} \int d^3\boldsymbol{x}' \psi_i^+(\boldsymbol{x},t) \psi_j^+(\boldsymbol{x}',t) \\ & \times V_{ij}(\boldsymbol{x}-\boldsymbol{x}',t) \psi_j(\boldsymbol{x}',t) \psi_i(\boldsymbol{x},t). \end{aligned} \tag{21}$$

Equation (21) is the second-quantized Hamiltonian in length gauge, whereas Eq. (12) is the Hamiltonian in velocity gauge.

In spite of our previous observation that the velocity-gauge Hamiltonian (12) preserves form invariance of the equations of motion (16) under the restricted gauge transformations (17), the equations of motion resulting from Eq. (21),

$$\begin{aligned} i\hbar \frac{\partial}{\partial t} \psi_i(\boldsymbol{x},t) &= [\psi_i(\boldsymbol{x},t), H_l] \\ &= -\frac{\hbar^2}{2m_i} \Delta \psi_i(\boldsymbol{x},t) + V_i(\boldsymbol{x},t) \psi_i(\boldsymbol{x},t) \\ &\quad - q_i \boldsymbol{x} \cdot \boldsymbol{E}(t) \psi_i(\boldsymbol{x},t) \\ &\quad + \sum_j \int d^3\boldsymbol{x}' \psi_j^+(\boldsymbol{x}',t) V_{ij}(\boldsymbol{x}-\boldsymbol{x}',t) \\ &\quad \times \psi_j(\boldsymbol{x}',t) \psi_i(\boldsymbol{x},t), \end{aligned} \tag{22}$$

do *not* have the same form as the equations resulting from Eq. (12) because we explicitly substituted $\boldsymbol{A}'(t) = 0$ and $V_i'(\boldsymbol{x},t) = V_i(\boldsymbol{x},t) - q_i \boldsymbol{x} \cdot \boldsymbol{E}(t)$.

There are different ways to understand the discrepancy between Eqs. (16) and (22). In a geometric analogy, we can think of the general formulation of equations of motion of a gauge theory as the form-invariant formulation of the theory, akin to the tensor formulation of the Einstein equation in General Relativity. However, formulating the equations of motion in a particular gauge is akin to choosing a particular coordinate system, and manifest form invariance is lost once the equations are formulated in that system.

Alternatively, we can think of the transformation that led to Eq. (22) as an incomplete gauge transformation which mixes the *transformed* Schrödinger fields with the *original* potentials through $V_i(\boldsymbol{x},t)$ and

$$-q_i \boldsymbol{x} \cdot \boldsymbol{E}(t) = q_i \boldsymbol{x} \cdot d\boldsymbol{A}(t)/dt.$$

In a geometric analog, this is like using mixed coordinates $x' = x \cos\phi + y \sin\phi$ and y, but not the transformed coordinate $y' = y \cos\phi - x \sin\phi$ after a rotation in two dimensions. In that picture, the transformation that led from Eq. (16) to Eq. (22) is rather the analog of an affine transformation in field space instead of a full gauge transformation: the transition from Eq. (12) to Eq. (21) only involved the field redefinition (18), which we can think of as a basic dressing operation, but not a complete gauge transformation. We also note that, contrary to the original Hamiltonian (12), the length-gauge Hamiltonian (21) yields equations of motion (22) which are manifestly form invariant only under even more restricted gauge transformations $\varphi(\boldsymbol{x},t) = b(t)$. This point of view sheds new light on the gauge problem from a different angle. In particular, we will see explicitly in Sec. III that, contrary to an ordinary full gauge transformation, the transition to length gauge can be selectively applied only to particular Schrödinger field operators, thus implying the possibility of selective transformations only for particular particle species.

The loss of manifest form invariance cannot affect the underlying physics, and ultimately the Hamiltonians (12) and (21) should yield the same physical results [18,40,43] if evaluated analytically or with sufficiently high precision, and yet careful evaluations have led to very different results in many instances. Before elucidating these points any further, we also note that implementing the dipole approximation on the level of potentials $\boldsymbol{A}(\boldsymbol{x},t) \to \boldsymbol{A}(t)$ eliminates magnetic effects from the outset, thus changing the Lorentz invariants of the theory and effectively reducing the equations (10) and (11) only to the one equation (10) [44].

III. SWITCHING TO LENGTH GAUGE ONLY FOR PARTICULAR SPECIES

The fact that the transition between velocity gauge and length gauge can be viewed as an incomplete gauge transformation, *viz.* acting only on the matter fields, has the interesting implication that we can *separately choose for each matter field* $\psi_i(\boldsymbol{x},t)$ whether we wish to represent its couplings in velocity gauge or in length gauge. Recall that, in second quantization, different matter fields $\psi_i(\boldsymbol{x},t)$, $1 \leqslant i \leqslant n$, represent different particle species, i.e., the assertion above implies that, in a system of electrons, protons, and α particles, we may, e.g., choose to represent the electrons and α particles in velocity gauge and the protons in length gauge. Suppose we have n different species of charged particles, and we retain velocity gauge for n_v of them, labeled by indices $1 \leqslant i \leqslant n_v$, while we switch to length gauge for $n_l = n - n_v$. We will label the latter field operators with indices $1 \leqslant \bar{\imath} \equiv i - n_v \leqslant n_l$. The Hamiltonian in the mixed representation of electromagnetic interactions then takes the form

$$\begin{aligned} H_{vl} = & \int d^3\boldsymbol{x} \sum_{i=1}^{n_v} \frac{1}{2m_i} \big\{ \hbar^2 \nabla \psi_i^+(\boldsymbol{x},t) \cdot \nabla \psi_i(\boldsymbol{x},t) \\ & + i q_i \hbar \boldsymbol{A}(t) \cdot [\psi_i^+(\boldsymbol{x},t) \overleftrightarrow{\nabla} \psi_i(\boldsymbol{x},t)] \\ & + q_i^2 \psi_i^+(\boldsymbol{x},t) \boldsymbol{A}^2(t) \psi_i(\boldsymbol{x},t) \big\} \\ & + \int d^3\boldsymbol{x} \sum_{\bar{\imath}=1}^{n_l} \bigg(\frac{\hbar^2}{2m_{\bar{\imath}}} \nabla \psi_{\bar{\imath}}^+(\boldsymbol{x},t) \cdot \nabla \psi_{\bar{\imath}}(\boldsymbol{x},t) \\ & - q_{\bar{\imath}} \psi_{\bar{\imath}}^+(\boldsymbol{x},t) \boldsymbol{x} \cdot \boldsymbol{E}(t) \psi_{\bar{\imath}}(\boldsymbol{x},t) \bigg) \\ & + \sum_{i=1}^{n} \psi_i^+(\boldsymbol{x},t) V_i(\boldsymbol{x},t) \psi_i(\boldsymbol{x},t) \\ & + \frac{1}{2} \sum_{i,j=1}^{n} \int d^3\boldsymbol{x} \int d^3\boldsymbol{x}' \psi_i^+(\boldsymbol{x},t) \psi_j^+(\boldsymbol{x}',t) \\ & \times V_{ij}(\boldsymbol{x} - \boldsymbol{x}',t) \psi_j(\boldsymbol{x}',t) \psi_i(\boldsymbol{x},t). \end{aligned} \tag{23}$$

If we are using a semiclassical approximation by replacing the photon operators $\boldsymbol{A}(t)$, $\boldsymbol{E}(t) = -d\boldsymbol{A}(t)/dt$, with classical fields, the operator H_{vl} preserves particle numbers. In this case we can map the second-quantized Schrödinger equation for states in Fock space,

$$i\hbar \frac{d}{dt} |\Psi(t)\rangle = H_{vl} |\Psi(t)\rangle, \tag{24}$$

into decoupled wave equations for many-particle wave functions within each sector of Fock space by using the *ansatz*

$$\begin{aligned} |\Psi(t)\rangle = & \int d^3\boldsymbol{x}_1 \psi_{i_1}^+(\boldsymbol{x}_1) \cdots \int d^3\boldsymbol{x}_N \psi_{i_N}^+(\boldsymbol{x}_N) |0\rangle \\ & \times \Psi_{i_1,\ldots,i_N}(\boldsymbol{x}_1,\ldots,\boldsymbol{x}_N,t), \end{aligned} \tag{25}$$

where $\psi_i^+(\boldsymbol{x})$ are the Schrödinger picture creation operators. We assume ordering of the particle species labels i_k in such a way that all labels $i_k \leqslant n_v$ correspond to field operators in velocity gauge, while all labels $i_k > n_v$ correspond to field operators in length gauge. Use of the canonical (anti-)commutation relations of the field operators and linear independence of the N-particle states then reduces the second-quantized Schrödinger equation (24) to the wave equation

$$i\hbar \frac{\partial}{\partial t} \Psi_{i_1,\ldots,i_N}(\boldsymbol{x}_1,\ldots,\boldsymbol{x}_N,t) = H_{vl}^{(1)} \Psi_{i_1,\ldots,i_N}(\boldsymbol{x}_1,\ldots,\boldsymbol{x}_N,t), \tag{26}$$

with the first-quantized Hamiltonian

$$H_{vl}^{(1)} = -\sum_{k|i_k\leqslant n_v}\frac{\hbar^2}{2m_{i_k}}\left(\frac{\partial}{\partial \boldsymbol{x}_k}-\frac{i}{\hbar}q_{i_k}\boldsymbol{A}(t)\right)^2 - \sum_{k|i_k>n_v}\left(\frac{\hbar^2}{2m_{i_k}}\frac{\partial^2}{\partial \boldsymbol{x}_k^2}+q_{i_k}\boldsymbol{x}_k\cdot\boldsymbol{E}(t)\right) + \frac{1}{2}\sum_{j,k=1}^{N}V_{i_j,i_k}(\boldsymbol{x}_j-\boldsymbol{x}_k,t)+\sum_{k=1}^{N}V_{i_k}(\boldsymbol{x}_k,t). \quad (27)$$

Furthermore, note that, within the framework of a Hartree approximation, an *ansatz*

$$\Psi_{i_1,\ldots,i_N}(\boldsymbol{x}_1,\ldots,\boldsymbol{x}_N,t)=\prod_{k=1}^{N}\Psi_{i_k}(\boldsymbol{x}_k,t)$$

is used for the many-particle wave functions without imposing symmetry or antisymmetry for bosons or fermions of the same species. In this case the transformation between velocity-gauge and length-gauge wave functions,

$$\Psi_{i_k}^{(v)}(\boldsymbol{x}_k,t)\Leftrightarrow\Psi_{i_k}^{(l)}(\boldsymbol{x}_k,t)=\exp\left[-\frac{i}{\hbar}q_{i_k}\boldsymbol{x}_k\cdot\boldsymbol{A}(t)\right]\Psi_{i_k}^{(v)}(\boldsymbol{x}_k,t), \quad (28)$$

can even be *separately imposed for each particle in the system*, i.e., within the limitations of Hartree approximations to many-particle systems, we could even treat, e.g., electrons in outer orbitals of an atom in velocity gauge and electrons in inner orbitals in length gauge!

IV. ANALYTIC DIFFERENCES BETWEEN VELOCITY GAUGE AND LENGTH GAUGE

The different form of interaction terms in the equations of motion (16) and (22) and the corresponding Hamiltonians (12) and (21) affects numerical procedures which can contribute to different results for the two descriptions of quantum optical interactions [9,10,38]. However, in spite of the formal analytic equivalence of the theories through the transformation (18), there are also important sources of analytical *inequivalence* between the two formalisms.

We further pursue this question for an analytic origin of differences between velocity gauge and length gauge in a single-electron approximation. Examinations of atoms and molecules in strong electromagnetic fields also use this approximation, and it will simplify the equations considerably without limiting the conclusions on the origin of differences between velocity gauge or length gauge. The corresponding Hamiltonians are

$$H_v=\int d^3\boldsymbol{x}\left(\frac{1}{2m_e}\{\hbar^2\nabla\psi^+(\boldsymbol{x},t)\cdot\nabla\psi(\boldsymbol{x},t) - ie\hbar\boldsymbol{A}(t)\cdot[\psi^+(\boldsymbol{x},t)\overset{\leftrightarrow}{\nabla}\psi(\boldsymbol{x},t)] + e^2\psi^+(\boldsymbol{x},t)\boldsymbol{A}^2(t)\psi(\boldsymbol{x},t)\}+\psi^+(\boldsymbol{x},t)V(\boldsymbol{x})\psi(\boldsymbol{x},t)\right), \quad (29)$$

and

$$H_l=\int d^3\boldsymbol{x}\left(\frac{\hbar^2}{2m_e}\nabla\Psi^+(\boldsymbol{x},t)\cdot\nabla\Psi(\boldsymbol{x},t) + \Psi^+(\boldsymbol{x},t)V(\boldsymbol{x})\Psi(\boldsymbol{x},t)+e\Psi^+(\boldsymbol{x},t)\boldsymbol{x}\cdot\boldsymbol{E}(t)\Psi(\boldsymbol{x},t)\right), \quad (30)$$

where $V(\boldsymbol{x})$ is the effective potential experienced by the electron. The transformation of the electron field operators to length gauge is

$$\Psi(\boldsymbol{x},t)=\exp\left[ie\boldsymbol{x}\cdot\boldsymbol{A}(t)/\hbar\right]\psi(\boldsymbol{x},t), \quad (31)$$

or equivalently in terms of the $\boldsymbol{k}$-space operators,

$$\Psi(\boldsymbol{k},t)=\psi(\boldsymbol{k}-e\boldsymbol{A}(t)/\hbar,t). \quad (32)$$

Strong-electric-field situations are usually analyzed in terms of numerical solutions of the corresponding Schrödinger wave equations. The equations (16) and (22) for the operators become the corresponding Schrödinger wave equations for wave functions $\tilde{\psi}(\boldsymbol{x},t)$ and $\tilde{\Psi}(\boldsymbol{x},t)$, respectively, after switching into the Schrödinger picture and expanding the states in the one-particle sector of Fock space. The velocity-gauge Schrödinger equation in Fock space,

$$i\hbar\frac{d}{dt}|\Phi(t)\rangle=H_v|\Phi(t)\rangle,$$

applied to single-particle states,

$$|\Phi(t)\rangle=\int d^3\boldsymbol{x}\psi^+(\boldsymbol{x})|0\rangle\tilde{\psi}(\boldsymbol{x},t), \quad (33)$$

yields (after dropping the tilde from the wave function)

$$i\hbar\frac{\partial}{\partial t}\psi(\boldsymbol{x},t)=-\frac{[\hbar\nabla+ie\boldsymbol{A}(t)]^2}{2m_e}\psi(\boldsymbol{x},t)+V(\boldsymbol{x})\psi(\boldsymbol{x},t), \quad (34)$$

and similarly in length gauge,

$$i\hbar\frac{\partial}{\partial t}\Psi(\boldsymbol{x},t)=-\frac{\hbar^2}{2m_e}\Delta\Psi(\boldsymbol{x},t)+V(\boldsymbol{x})\Psi(\boldsymbol{x},t) + e\boldsymbol{x}\cdot\boldsymbol{E}(t)\Psi(\boldsymbol{x},t). \quad (35)$$

Nevertheless, equations (31) and (32) now apply to the wave functions and map the solutions of Eq. (34) bijectively and unitarily into the solutions of Eq. (35). So how can we get different physical results from equations (34) and (35)?

There are several effects here that need to be taken into account. On the one hand, since the exponent in Eq. (31) is only first order in e, any perturbative results from Eqs. (34) and (35) will differ beyond first order or, stated differently, what is formally second-order perturbation theory in Eq. (35) is not actually second-order perturbation theory in Eq. (34). For first-order matrix elements, the familiar identity

$$\hbar\mathbf{p}=im_e[H,\mathbf{x}] \quad (36)$$

ensures

$$\langle f|\mathbf{p}|i\rangle=im_e\omega_{fi}\langle f|\mathbf{x}|i\rangle \quad (37)$$

between energy eigenstates. This implies equivalence of velocity and length forms of transition matrix elements in

the limit of large transition times, $S_{fi} = \langle f|U_D(\infty,-\infty)|i\rangle$, where $U_D(t,t')$ is the time evolution operator in the interaction (Dirac) picture. Equation (37) then takes the form which actually ensures strict equivalence between first-order matrix elements in velocity and length gauge,

$$\langle f|\mathbf{p}|i\rangle \to \pm i m_e c k \langle f|\mathbf{x}|i\rangle. \tag{38}$$

See Refs. [45,46] for the inclusion of relativistic corrections in Eq. (36) in atomic systems and confirmation that high-precision numerical evaluations of first-order matrix elements for transitions in helium respect equivalence at the 10 ppb level. At the level of second-order matrix elements, Jentschura and Pachucki showed that Eq. (36) can also be used to demonstrate equivalence between *velocity-gauge* and *length-gauge* results for polarizabilities of isotropic states [47]. This is remarkable, because we have to distinguish between *length form* and *velocity form* of matrix elements on the one hand, and *length-gauge* and *velocity-gauge* results on the other hand. The identity (37) can always be used to transform matrix elements in either gauge, without ever invoking the gauge transformation (18), and at the level of first-order perturbation theory, Eq. (38) can then be used to demonstrate equivalence of length-gauge and velocity-gauge results if the amplitudes come with an energy-conserving factor $\delta(\omega_{fi} \mp ck)$. However, this does not generically work at second or higher order. For example, the differential scattering cross section (3) is the *velocity-gauge* electron-photon-scattering cross section in *length form*, and the comparison with Eq. (4) shows the differences with the *length-gauge* electron-photon-scattering cross section in *length form*.

Indeed, due to the difficulties of interpretation of the transition from velocity gauge to length gauge through the transformation (18), Fulton had argued that the dipole approximation should only be implemented at the end of calculations, when any changes between length and velocity representations should only be based on the relations (37) and (38). This reduces the problem to the discussion of gauge compatibility of the different forms of matrix elements. Fulton demonstrated applicability of the technique (37) and (38) for interaction potentials which depend on the locations of many particles [48]. Furthermore, Johnson and collaborators used the relativistic versions of (37) and (38) to demonstrate equivalence of length gauge and velocity gauge of single-photon transition matrix elements in first order in photon operators for relativistic many-electron systems [49,50]. They were not concerned with loss of gauge equivalence for higher-order photon transitions but demonstrated that, in first order in photon operators, gauge equivalence is maintained for perturbations of the initial and final electron states through relativistic Coulomb and Breit potentials.

The transition from Eq. (37) to Eq. (38) is justified by the fact that these matrix elements appear with factors $\delta(\omega_{fi} \mp ck)$ for large observation times $\Delta t = t - t'$. However, for monochromatic perturbation operators with frequency $\omega = \pm ck$ and small pulse duration Δt, these factors are replaced already in first order by a Dirichlet kernel $\sin[(\omega_{fi} \mp ck)\Delta t/2]/\pi(\omega_{fi} \mp ck)$. This is a matter of concern for the description of subfemtosecond spectroscopy in velocity or length gauge, since the resulting energy uncertainty for time resolution $\Delta t \lesssim 10^{-15}$ s is already of the order of $\hbar/\Delta t \gtrsim$ 0.7 eV. This implies discrepancies between first-order matrix elements in length gauge and velocity gauge at least at the several-percent level or higher, if the subfemtosecond experiments are performed with UV laser pulses, and even higher uncertainties for subfemtosecond experiments at lower wavelengths [51,52]. Many aspects of pulse duration on quantum-mechanical signals and spectral analysis are discussed in Ref. [53].

Furthermore, we should expect even larger differences between velocity gauge and length gauge for electron detachment in strong fields. Before the detachment, energy eigenstates would be defined in terms of atomic or molecular wave functions. However, after the detachment, wave functions are usually described in terms of dominant electric field and subdominant potential terms, i.e., as Volkov solutions. This implies that equation (36) and therefore also equations (37) and (38) do not apply and cannot be used to infer equivalence of velocity-gauge and length-gauge matrix elements.

A third analytic caveat regarding the equivalence of velocity gauge and length gauge concerns the fact that the mapping (31), $|\Psi(t)\rangle = \exp[ie\mathbf{x}\cdot\boldsymbol{A}(t)/\hbar]|\psi(t)\rangle$, is *not a unitary mapping of the Hamiltonian* [54]. To elucidate this point, note that the mapping of the Hamiltonians in

$$i\hbar\frac{d}{dt}|\psi(t)\rangle = H_v(t)|\psi(t)\rangle \Rightarrow i\hbar\frac{d}{dt}|\Psi(t)\rangle = H_l(t)|\Psi(t)\rangle$$

transforms

$$H_v(t) = \frac{[\mathbf{p} + e\boldsymbol{A}(t)]^2}{2m_e} + V(\mathbf{x})$$

into

$$\begin{aligned} H_l(t) &= \exp[ie\mathbf{x}\cdot\boldsymbol{A}(t)/\hbar]\left(H_v(t) - e\mathbf{x}\cdot\frac{d\boldsymbol{A}(t)}{dt}\right) \\ &\quad\times \exp[-ie\mathbf{x}\cdot\boldsymbol{A}(t)/\hbar] \\ &= \frac{\mathbf{p}^2}{2m_e} + V(\mathbf{x}) - e\mathbf{x}\cdot\frac{d\boldsymbol{A}(t)}{dt}, \end{aligned} \tag{39}$$

and therefore the matrix elements of the Hamiltonians in velocity gauge and length gauge differ,

$$\begin{aligned} &\langle\Phi(t)|H_l(t)|\Psi(t)\rangle \\ &\quad= \langle\phi(t)|H_v(t)|\psi(t)\rangle - e\langle\phi(t)|\mathbf{x}|\psi(t)\rangle\cdot\frac{d\boldsymbol{A}(t)}{dt}. \end{aligned} \tag{40}$$

This implies that $H_l(t)$ and $H_v(t)$ evolve the system differently through the eigenstates of the unperturbed system during an electromagnetic pulse. We also note that Eq. (40) can yield different energy expectation values for systems which are not invariant under parity, or which have degeneracy between even and odd states. Note that the question for equivalence of energy expectation values for systems with, e.g., a single mirror symmetry but no other parity invariance hinges on the polarization of the incident radiation.

These observations are of no concern if we only care about time evolution of a system from a state before a classical electromagnetic pulse was applied to a state after the pulse has ceased. However, if we wish to derive information about optical responses of the system from shifts of energy levels and eigenstates during application of a strong external field, then we have to anticipate principally different results

from velocity- and length-gauge descriptions. These results can ultimately be mapped through application of Eq. (31). However, this will yield unique, generally-agreed-upon results only if we agree whether the velocity gauge states $|\psi(t)\rangle$ or the length-gauge states $|\Psi(t)\rangle$ correspond to unperturbed system eigenstates in the evaluation of any matrix elements of $H_v(t)$ or $H_l(t)$ or their corresponding interaction terms. Stated differently, the question is which state we consider as a basic atomic or molecular state, and which state we consider as a kind of dressed state through application of a basic dressing operation $\exp[\pm ie\boldsymbol{x}\cdot\boldsymbol{A}(t)/\hbar]$. After all, the transformation (31) impresses the Fourier components $\boldsymbol{A}(\omega)$ along with all higher harmonics onto the dressed wave function. Integrations of equations (34) and (35) starting with the same atomic or molecular state in a laser field of frequency ω_0 would therefore describe different physical situations. This confirms the observation of Galstyan *et al.* of two principally different families of solutions in quantum optical systems [38].

We can also generate the transformation (31) of the Schrödinger field operators from velocity gauge to length gauge as a unitary transformation in the form

$$\Psi(\boldsymbol{x},t) = \Sigma(t)\psi(\boldsymbol{x},t)\Sigma^{+}(t), \tag{41}$$

with

$$\Sigma(t) = \exp\left[\frac{e}{i\hbar}\int d^3\boldsymbol{x}\,\psi^{+}(\boldsymbol{x},t)\boldsymbol{x}\cdot\boldsymbol{A}(t)\psi(\boldsymbol{x},t)\right]. \tag{42}$$

To fully appreciate the implications of this observation, we have to go beyond the semiclassical approximation and restore the photon operators in the dipole approximation,

$$\boldsymbol{A}(t) = \sqrt{\frac{\hbar\mu_0 c}{(2\pi)^3}}\int\frac{d^3\boldsymbol{k}}{\sqrt{2k}}\sum_{\alpha=1}^{2}\boldsymbol{\epsilon}_\alpha(\boldsymbol{k})[a_\alpha(\boldsymbol{k})\exp(-ickt) + a_\alpha^{+}(\boldsymbol{k})\exp(ickt)], \tag{43}$$

with $\boldsymbol{k}\cdot\boldsymbol{\epsilon}_\alpha(\boldsymbol{k}) = 0$, $\boldsymbol{\epsilon}_\alpha(\boldsymbol{k})\cdot\boldsymbol{\epsilon}_\beta(\boldsymbol{k}) = \delta_{\alpha\beta}$, $[a_\alpha(\boldsymbol{k}),a_\beta(\boldsymbol{k}')] = 0$, and $[a_\alpha(\boldsymbol{k}),a_\beta^{+}(\boldsymbol{k}')] = \delta_{\alpha\beta}\delta(\boldsymbol{k}-\boldsymbol{k}')$.

Even after restoring $\boldsymbol{A}(t)$ as a photon operator, the unitary transformation (42) leaves the vacuum invariant, but charged particle states pick up a photon cloud either in length gauge or in velocity gauge. Suppose we start with the single charged particle state in velocity gauge,

$$|\phi(t)\rangle = \int d^3\boldsymbol{x}\,\psi^{+}(\boldsymbol{x})|0\rangle\phi(\boldsymbol{x},t). \tag{44}$$

The corresponding length-gauge state is

$$|\Phi(t)\rangle = \int d^3\boldsymbol{x}\,\Psi^{+}(\boldsymbol{x})|0\rangle\phi(\boldsymbol{x},t) = \Sigma(0)|\phi(t)\rangle = \int d^3\boldsymbol{x}\exp\left(\frac{e}{i\hbar}\boldsymbol{x}\cdot\boldsymbol{A}\right)\psi^{+}(\boldsymbol{x})|0\rangle\phi(\boldsymbol{x},t), \tag{45}$$

where $\boldsymbol{A}\equiv\boldsymbol{A}(t=0)$ is the photon operator in the Schrödinger picture. The length-gauge state would be dressed by a coherent photon state.

On the other hand, if we suppose that the length-gauge state is a charged single-particle state, we would infer a dressed velocity-gauge state

$$|\phi(t)\rangle = \int d^3\boldsymbol{x}\exp(ie\boldsymbol{x}\cdot\boldsymbol{A}/\hbar)\Psi^{+}(\boldsymbol{x})|0\rangle\phi(\boldsymbol{x},t). \tag{46}$$

Note that the photon components in the dressed length-gauge states (45) or dressed velocity-gauge states (46) correspond to coherent states (see the appendix for definitions and conventions for coherent states)

$$|\boldsymbol{\zeta}\rangle = \exp(\pm ie\boldsymbol{x}\cdot\boldsymbol{A}/\hbar)|0\rangle,$$

with amplitudes

$$\zeta_\alpha(\boldsymbol{k},\boldsymbol{x}) = \pm i\frac{e}{4\pi}\sqrt{\frac{\mu_0 c}{\pi\hbar k}}\boldsymbol{x}\cdot\boldsymbol{\epsilon}_\alpha(\boldsymbol{k}).$$

V. ACCELERATION GAUGE AND SHIFTS IN PHASE SPACE

The apparent differences in velocity-gauge and length-gauge results have always been puzzling, since the transformation (18) can be implemented as a gauge transformation (19) and (20). On the other hand, every unitary time-dependent transformation of wave functions or quantum states constitutes a change of the picture of quantum dynamics. U(1) gauge transformations can be expressed in terms of a Hermitian phase factor $\varphi(\mathbf{x},t)$, like the transformation (18) expressed on the states,

$$|\psi^{(l)}(t)\rangle = \Upsilon_{lv}(t)|\psi^{(v)}(t)\rangle, \qquad \Upsilon_{lv}(t) = \exp[-iq\mathbf{x}\cdot\boldsymbol{A}(t)/\hbar]. \tag{47}$$

However, more general U(1) transformations with Hermitian phase factors $\varphi(\mathbf{p},\mathbf{x},t)$ can just as well be used to change the picture of quantum dynamics, and we must just as well be able to recover the correct physical predictions from any unitarily related picture of quantum dynamics. The only special property of the particular subgroup of picture-changing operations which are gauge transformations is the fact that *the equations of the corresponding gauge theory remain form invariant as long as we do not commit to any particular gauge* (just like the equations of Geometry or General Relativity remain form invariant under diffeomorphisms as long as we do not commit to any particular coordinate system). Therefore it helps to illuminate the problem of apparent inequivalence of velocity gauge and length gauge from the more general perspective of picture-changing transformations, which we limit to the case of time-dependent U(1) transformations $\Upsilon(t) = \exp[iq\varphi(\mathbf{p},\mathbf{x},t)/\hbar]$.

One example is provided by the Galilei transformation with boost parameter $\boldsymbol{v}_G$,

$$\Upsilon_G(t) = \exp[-i\mathbf{p}\cdot\boldsymbol{v}_G t/\hbar]\exp[im\mathbf{x}\cdot\boldsymbol{v}_G/\hbar], \tag{48}$$

which transforms the velocity-gauge Hamiltonian

$$H_v(t) = \frac{[\mathbf{p}-q\boldsymbol{A}(t)]^2}{2m} + V(\mathbf{x}) \tag{49}$$

into

$$H_G(t) = \frac{[\mathbf{p} - m\boldsymbol{v}_G - q\boldsymbol{A}(t)]^2}{2m} + V(\mathbf{x} - \boldsymbol{v}_G t) + \boldsymbol{v}_G \cdot \mathbf{p}.$$

Another well-known example is provided by the acceleration-frame transformation [18,39,40] (see also Ref. [55] for references to earlier occurrences of this transformation in the physics literature)

$$\Upsilon_{av}(t) = \exp\left(-i\frac{q}{\hbar}\frac{\mathbf{p}}{m}\cdot\int_{t_0}^{t} d\tau \boldsymbol{A}(\tau)\right) \times \exp\left(i\frac{q^2}{2m\hbar}\int_{t_0}^{t} d\tau \boldsymbol{A}^2(\tau)\right), \tag{50}$$

which shifts the operator for particle location according to

$$\mathbf{x} \to \mathbf{X}(t) = \Upsilon_{av}(t)\cdot\mathbf{x}\cdot\Upsilon_{av}^{+}(t) = \mathbf{x} - \frac{q}{m}\int_{t_0}^{t} d\tau \boldsymbol{A}(\tau),$$

and transforms the velocity-gauge Hamiltonian (49) into the "acceleration-gauge" (or "Henneberger frame" [40]) Hamiltonian

$$H_a(t) = \frac{\mathbf{p}^2}{2m} + V(\mathbf{X}(t)).$$

The transformation which maps the length-gauge Hamiltonian

$$H_l(t) = \frac{\mathbf{p}^2}{2m} + V(\mathbf{x}) + q\mathbf{x}\cdot\frac{d\boldsymbol{A}(t)}{dt}$$

into $H_a(t)$ is then given by

$$\begin{aligned}\Upsilon_{al}(t) &= \Upsilon_{av}(t)\cdot\Upsilon_{lv}^{+}(t)\\ &= \exp\left\{i\frac{q^2}{2m\hbar}\int_{t_0}^{t} d\tau[\boldsymbol{A}^2(\tau) - \boldsymbol{A}(t)\cdot\boldsymbol{A}(\tau)]\right\}\\ &\quad\times\exp\left\{i\frac{q}{\hbar}\left[\mathbf{x}\cdot\boldsymbol{A}(t) - \frac{\mathbf{p}}{m}\cdot\int_{t_0}^{t} d\tau\boldsymbol{A}(\tau)\right]\right\}.\end{aligned}$$

All these cases are captured in the group of shift operators in phase space,

$$\begin{aligned}\Upsilon(t) &= \exp\left[iq\mathbf{p}\cdot\boldsymbol{c}(t)/\hbar\right]\exp\left[iq\mathbf{x}\cdot\boldsymbol{a}(t)/\hbar\right]\\ &= \exp\left[iq\varphi(\mathbf{p},\mathbf{x},t)/\hbar\right]\end{aligned} \tag{51}$$

with

$$\varphi(\mathbf{p},\mathbf{x},t) = \mathbf{x}\cdot\boldsymbol{a}(t) + \mathbf{p}\cdot\boldsymbol{c}(t) + \frac{q}{2}\boldsymbol{a}(t)\cdot\boldsymbol{c}(t), \tag{52}$$

and these reduce to U(1) gauge transformations if $\boldsymbol{c}(t) = 0$.

Besides the advantage of Eq. (51) of also including Galilei transformations (48) and the Henneberger transformation (50), it is also instructive to think about the question of equivalence of time-dependent gauge transformations in the larger framework of time-dependent unitary transformations. A time-dependent unitary transformation

$$|\psi'(t)\rangle = \Upsilon(t)|\psi(t)\rangle \tag{53}$$

shifts the Hamiltonian according to

$$\begin{aligned}H'(t) &= \Upsilon(t)\cdot\left[H(t) - i\hbar\frac{d}{dt}\right]\cdot\Upsilon^{+}(t)\\ &= \Upsilon(t)\cdot H(t)\cdot\Upsilon^{+}(t) - i\hbar\sum_{n=1}^{\infty}\frac{1}{n!}\left(\frac{iq}{\hbar}\right)^{n}{}^{n}[\,\varphi(t),d/dt]\\ &= \Upsilon(t)\cdot H(t)\cdot\Upsilon^{+}(t)\\ &\quad - q\sum_{n=1}^{\infty}\frac{1}{n!}\left(\frac{iq}{\hbar}\right)^{n-1}{}^{n-1}[\,\varphi(t),\dot\varphi(t)].\end{aligned} \tag{54}$$

This is the necessary and sufficient condition for the correct transformation law

$$U'(t,t') = \Upsilon(t)U(t,t')\Upsilon^{+}(t') \tag{55}$$

of the time evolution operators in the two pictures.

Equation (54) implies for the phase-space shift operators (51) and (52) the following transformation of Hamiltonians:

$$\begin{aligned}H'(t) &= \Upsilon(t)\cdot H(t)\cdot\Upsilon^{+}(t) - q\dot\varphi(t) - \frac{iq^2}{2\hbar}[\varphi(t),\dot\varphi(t)]\\ &= \Upsilon(t)\cdot H(t)\cdot\Upsilon^{+}(t) - q\mathbf{x}\cdot\dot{\boldsymbol{a}}(t) - q\mathbf{p}\cdot\dot{\boldsymbol{c}}(t)\\ &\quad - q^2\dot{\boldsymbol{a}}(t)\cdot\boldsymbol{c}(t).\end{aligned} \tag{56}$$

Therefore, while Eq. (55) of course implies

$$\langle\phi'(t)|U'(t,t')|\psi'(t)\rangle = \langle\phi(t)|U(t,t')|\psi(t)\rangle, \tag{57}$$

the matrix elements of the Hamiltonians in the two representations are related by

$$\begin{aligned}\langle\phi'(t)|H'(t)|\psi'(t)\rangle &= \langle\phi(t)|H(t)|\psi(t)\rangle - q\dot{\boldsymbol{a}}(t)\cdot\langle\phi(t)|\mathbf{x}|\psi(t)\rangle\\ &\quad - q\dot{\boldsymbol{c}}(t)\cdot\langle\phi(t)|\mathbf{p}|\psi(t)\rangle\\ &\quad - q^2\boldsymbol{a}(t)\cdot\dot{\boldsymbol{c}}(t)\langle\phi(t)|\psi(t)\rangle,\end{aligned} \tag{58}$$

which generalizes Eq. (40). Equivalence, e.g., of velocity and length gauge has been argued on the basis of the identity (57); see, e.g., Ref. [18]. However, actual calculations of transition probabilities, decay rates, or scattering cross sections are based on the matrix elements of the interaction picture (or Dirac picture) time evolution operator between energy eigenstates,

$$S_{nm}(t,t') = \langle\psi_n|U_D(t,t')|\psi_m\rangle, \tag{59}$$

with the interaction picture time evolution operator

$$\begin{aligned}U_D(t,t') &= \exp\left(iH_0t/\hbar\right)U(t,t')\exp(-iH_0t'/\hbar)\\ &= \mathrm{T}\exp\left(-\frac{i}{\hbar}\int_{t'}^{t} d\tau H_D(\tau)\right),\end{aligned} \tag{60}$$

the Hamiltonian

$$H_D(t) = \exp\left(iH_0t/\hbar\right)W(t)\exp\left(-iH_0t/\hbar\right),$$

and the identification of $W(t)$, e.g., from

$$H_0(\mathbf{p},\mathbf{x}) = H(\mathbf{p},\mathbf{x},t) - W(\mathbf{p},\mathbf{x},t) = \frac{\mathbf{p}^2}{2m} + V(\mathbf{x}), \tag{61}$$

in a single (quasi-)particle approximation.

The initial and final times in Eq. (59) are usually taken as $t \to \infty$, $t' \to -\infty$, but the scattering-matrix elements both for finite and infinite times are principally just a matrix representation of the integration of the Schrödinger equation in the interaction picture [56]. However, applications of the different pictures (53) and (54) to the same system are based on the same identification (61) of the unperturbed Hamiltonian and therefore use the same eigenstates in Eq. (59), although $U_D'(t,t') \neq U_D(t,t')$. This is in short the origin of the principal difference of the scattering cross sections (3) and (4). More

specifically, scattering-matrix elements in the two pictures (53) and (54) are related by

$$\begin{aligned} S'_{nm}(t,t') &= \langle\psi_n|U'_D(t,t')|\psi_m\rangle \\ &= \sum_{kl} \exp[i(\omega_{nk}t + \omega_{lm}t')]\langle\psi_n|\Upsilon(t)|\psi_k\rangle \\ &\quad \times S_{kl}(t,t')\langle\psi_l|\Upsilon^+(t')|\psi_m\rangle. \end{aligned} \tag{62}$$

This is a more precise formulation of the observation from Eq. (40) that the different formulations evolve a system differently through the eigenstates of the unperturbed system. Equation (62) implies that transition probabilities, decay rates, and scattering cross sections will generically be different in the different pictures (53) and (54), as explicitly demonstrated for the case of velocity gauge versus length gauge in equations (3) and (4). On the other hand, if the picture-changing operation $\Upsilon(t)$ involves semiclassical electromagnetic potentials which correspond to pulses of finite duration $\Delta t < t - t'$, then the scattering-matrix elements $S_{nm}(t,t')$, with initial time t' before the time t_0 of onset of the pulse and final time t after cessation of the pulse, are *invariant* with respect to Eq. (53), and therefore *any observable computed from those scattering-matrix elements would also be invariant under the picture-changing operation* (53),

$$S'_{nm}(t,t') = S_{nm}(t,t'), \quad t' < t_0 < t_0 + \Delta t < t. \tag{63}$$

This does not apply to the scattering matrices leading to the scattering cross sections (3) and (4), since their calculation involves photon operators which describe spontaneous photon emission or absorption at any time, as opposed to time-constrained semiclassical pulses.

However, if Eq. (63) holds for semiclassical pulses, why then do many researchers find strikingly different results in studies of strong laser pulses, which are used, e.g., for electron detachment and higher-harmonic generation? From the previous observations, we can easily identify four sources for different results in different pictures:

(1) The scattering-matrix formalism will generically yield different results in the different pictures (53) and (54) due to the transformation law (62), unless it is applied to semiclassical pulses of finite duration [Eq. (63)].

(2) Due to the nonperturbative nature of the strong fields involved, theoretical investigations of strong-field experiments do not use the scattering-matrix formalism but employ numerical integrations of the time-dependent Schrödinger equation. Given the inhomogeneous transformation law (54) of the Hamiltonian during picture changing, it is not surprising that numerical integration of the Schrödinger equations for $H(t)$ and $H'(t)$ in the different pictures can lead to very different conclusions.

(3) The modeling of electron detachment in strong fields involves a change of H_0 during the experiment, since the bound-electron state is dominated by the attractive atomic or molecular potentials, whereas the ionized state is presumed to be dominated by the strong external radiation field. This renders the relations (37) and (38) useless for trying to establish equivalence even only at first order.

(4) As pointed out already before, the replacement of the energy-preserving δ function through the Dirichlet kernel

$$\delta(\omega_{nm} \mp ck) \to \frac{\sin[(\omega_{nm} \mp ck)\Delta t/2]}{\pi(\omega_{nm} \mp ck)}$$

for subfemtosecond pulses of frequency $\pm ck$ invalidates the relation (38) for equivalence between velocity- and length-gauge matrix elements,

$$\langle\psi_f|\mathbf{p}|\psi_i\rangle = im\omega_{fi}\langle\psi_f|\mathbf{x}|\psi_i\rangle \neq \pm imck\langle\psi_f|\mathbf{x}|\psi_i\rangle.$$

Note that this effect alone cannot change the *numerical equivalence* (63) of scattering-matrix elements for times t' and t before and after semiclassical pulses, but we cannot expect to be able to transform the scattering-matrix elements any more into the same analytic form.

VI. CONCLUSIONS

Four sources of analytic differences between velocity gauge and length gauge have been identified. The difference between $|\omega_{fi}|\langle f|\mathbf{x}|i\rangle$ and $ck\langle f|\mathbf{x}|i\rangle$ for short observation times or short pulses can be interpreted as a manifestation of energy-time uncertainty, but it implies a discrepancy between theoretical calculations in length gauge versus velocity gauge at the several percent level or higher for subfemtosecond spectroscopy.

Furthermore, equation (36) does not hold if the initial and final states are eigenstates of different Hamiltonians, as often appears in strong-field electron detachment. Therefore, no corresponding equivalence of velocity-gauge matrix elements and length-gauge matrix elements can be inferred from Eq. (36) in these cases.

In addition the scattering-matrix formalism in the different pictures. Eqs. (53) and (54) will generically yield different results due to the transformation law (62), unless it is applied to semiclassical pulses of finite duration [Eq. (63)].

Finally, the inequivalence (40) between matrix elements of the Hamilton operators implies potentially large analytic uncertainties for systems without parity invariance or with accidental degeneracies between even and odd states. This is in agreement with the observations of a high level of agreement between velocity- and length-gauge calculations for helium atoms [46] and the observation of equivalence of second-order matrix elements for polarizabilities [47] and two-photon transitions [54] in rotationally symmetric systems, while at the same time numerically evaluated matrix elements in velocity gauge and length gauge can be very different for chiral compounds [12,13], and calculated ionization properties can be very different for hydrogen [8,14,16,26].

From an analytic perspective, we would expect to find strong differences between velocity gauge and length gauge both for systems without parity invariance, and for strong-field electron detachment. This seems to be corroborated by numerical calculations in both cases and also by analytic work on strong-field electron detachment.

So far everything has been formulated in a neutral way without stating a preference for velocity gauge or length gauge or any of the other pictures implied in Eqs. (51)–(54). The transformation between velocity gauge and length gauge has always been formulated as a transition from velocity

gauge to length gauge; see, e.g., Eqs. (31) and (32), but everything can just as well be formulated in the opposite direction. However, since the different pictures can produce different results even at the analytic level, for the reasons outlined above, is there a picture that should be preferred for principal reasons? This question has been asked time and again especially for velocity gauge and length gauge. The results (41)–(46) indicate that, if the wave function in one of the pictures is considered as fundamental, the wave function in the other picture corresponds to a state which is dressed with a coherent photon component. However, this does not break the tie with respect to the question of which picture should be considered as "fundamental." In light of the fact that both velocity gauge and length gauge have proven to be particularly successful in their own right in different physical situations, the practical aspect of this question is rather: which of these different situations are better described by dressed states, and why? An argument which has often been made in favor of length gauge (besides the simpler form of the interaction term) is the apparent formulation in terms of gauge-invariant quantities $\boldsymbol{p} = m\boldsymbol{v}$ and $\boldsymbol{E}(t)$; see, e.g., Refs. [2,18]. Indeed, the momentum in length-gauge wave functions $\Psi(\boldsymbol{x},t)$ and velocity-gauge wave functions $\psi(\boldsymbol{x},t)$ is

$$\boldsymbol{P} = \int d^3\boldsymbol{x}\frac{\hbar}{i}\sum_i \Psi_i^+(\boldsymbol{x},t)\nabla\Psi_i(\boldsymbol{x},t) \tag{64}$$

$$= \int d^3\boldsymbol{x}\frac{\hbar}{i}\sum_i \psi_i^+(\boldsymbol{x},t)\nabla\psi_i(\boldsymbol{x},t) - \int d^3\boldsymbol{x}\sum_i q_i\psi_i^+(\boldsymbol{x},t)\boldsymbol{A}(t)\psi_i(\boldsymbol{x},t); \tag{65}$$

see also Appendix B, where these relations are derived on the level of Schrödinger field operators and taking into account the contributions from the electromagnetic fields to energy and momentum conservation. These relations and the equations (12) and (21) for Hamiltonians in velocity gauge and length gauge show that, in length gauge, we can identify momentum with particle velocity $\boldsymbol{p} = m\boldsymbol{v}$, whereas velocity gauge is of course based on the canonical relation $\boldsymbol{p} = m\boldsymbol{v} + q\boldsymbol{A}$ for particle momentum in external fields. As noted above, this apparent formulation in terms of gauge-invariant quantities has been alluded to as one advantage of the length-gauge formulation. Furthermore, the direct identification of momentum and velocity also makes length gauge advantageous for investigations of electron detachment [6,7,19–24] and applications of the Keldysh formalism [10,14], since intuitive methods using electron motion are better captured in that formalism. This advantage disappears for large internuclear separation in molecules, when the dipole operator during electron transfer can become an excessively large inherent perturbation of the system [25,26].

On the other hand, a different view can be expressed on the basis of reductionism. Recall that the Dirac Hamiltonian for quantum electrodynamics in Coulomb gauge has the form (see, e.g., Sec. 21.4 in Ref. [4])

$$H = \int d^3\boldsymbol{x}\left(\frac{\epsilon_0}{2}\boldsymbol{E}_\perp^2(\boldsymbol{x},t) + \frac{1}{2\mu_0}\boldsymbol{B}^2(\boldsymbol{x},t)\right) + \sum_i H_i + \sum_{ij} V_{ij}, \tag{66}$$

with $\boldsymbol{E}_\perp(\boldsymbol{x},t) = -\partial\boldsymbol{A}(\boldsymbol{x},t)/\partial t$, $\boldsymbol{B}(\boldsymbol{x},t) = \nabla\times\boldsymbol{A}(\boldsymbol{x},t)$, and the kinetic and Coulomb terms for fermion species with labels i,j (summations over the Dirac indices s are implicitly included in H_i)

$$H_i = -c\int d^3\boldsymbol{x}\overline{\psi}_i(\boldsymbol{x},t)\boldsymbol{\gamma}\cdot[i\hbar\nabla + q_i\boldsymbol{A}(\boldsymbol{x},t)]\psi_i(\boldsymbol{x},t) + m_ic^2\int d^3\boldsymbol{x}\overline{\psi}_i(\boldsymbol{x},t)\psi_i(\boldsymbol{x},t), \tag{67}$$

$$V_{ij} = \sum_{s,s'}\int d^3\boldsymbol{x}\int d^3\boldsymbol{x}'\frac{q_iq_j}{8\pi\epsilon_0|\boldsymbol{x}-\boldsymbol{x}'|} \times\psi_{is}^+(\boldsymbol{x},t)\psi_{js'}^+(\boldsymbol{x}',t)\psi_{js'}(\boldsymbol{x}',t)\psi_{is}(\boldsymbol{x},t). \tag{68}$$

This Hamiltonian reduces to the velocity-gauge Hamiltonian (12) in the nonrelativistic semiclassical dipole approximation, and therefore from a *reductionist* point of view the velocity-gauge Hamiltonian should be considered as more fundamental than the length-gauge Hamiltonian or any other picture of quantum dynamics in the semiclassical dipole approximation. This is also directly manifested in the $\psi^+\boldsymbol{A}^2\psi$ term in Eq. (1) and the corresponding first term in the scattering cross section (3), which arise in the nonrelativistic limit from intermediate virtual positron states. Another case in point concerns the question of which gauge naturally relates to standard atomic energy eigenstates. The $\boldsymbol{k}$-space representation of hydrogen eigenstates $|n,\ell,m_\ell\rangle$ is concentrated around $\boldsymbol{k}=0$ within a characteristic radius $k \lesssim (na)^{-1}$, where a is the Bohr radius, e.g., for the $1s$ state,

$$\langle\boldsymbol{k}|\Phi_{1,0,0}(t)\rangle = \frac{\sqrt{2a}^3}{\pi}\frac{\exp(-iE_1t/\hbar)}{[1+(ka)^2]^2}. \tag{69}$$

The transformation (31) and (32) therefore implies that the $\boldsymbol{k}$-space representation of the $1s$ state in velocity gauge, $\psi_{1,0,0}(\boldsymbol{k},t)$, would be concentrated around the wave number $-e\boldsymbol{A}(t)/\hbar$ if the length-gauge wave function $\Psi_{1,0,0}(\boldsymbol{k},t)$ would agree with Eq. (69). On the other hand, the length-gauge wave function $\Psi_{1,0,0}(\boldsymbol{k},t)$ would be concentrated in a radius $k \lesssim a^{-1}$ around the wave number $e\boldsymbol{A}(t)/\hbar$ if the velocity-gauge wave function $\psi_{1,0,0}(\boldsymbol{k},t)$ would agree with Eq. (69). The latter result would seem to make more sense on the basis of the length-gauge Schrödinger equation (35). Assume an extremely strong field such that (35) approximately reduces to

$$\frac{\partial}{\partial t}\Psi(\boldsymbol{x},t) \simeq \frac{ie}{\hbar}\boldsymbol{x}\cdot\frac{d\boldsymbol{A}(t)}{dt}\Psi(\boldsymbol{x},t). \tag{70}$$

This has the solution

$$\Psi(\boldsymbol{x},t) \simeq \exp\left(i\frac{e}{\hbar}\boldsymbol{x}\cdot[\boldsymbol{A}(t)-\boldsymbol{A}(t_0)]\right)\Psi(\boldsymbol{x},t_0), \tag{71}$$

which corresponds to propagation of the initial state with a plane-wave factor with wave number $e\boldsymbol{A}(t)/\hbar$. This would lend credibility to the conclusion that standard atomic eigenstates correspond to the velocity-gauge picture. It does not constitute mathematical proof, however, that the velocity-gauge picture is indeed more fundamental or superior in any way. Furthermore, in times of discussions about emergent phenomena in condensed-matter physics and quantum gravity, the reductionist argument given above may not carry much weight anymore and, indeed, declaring one gauge in any way as

more fundamental or superior than any other gauge was never the objective of this investigation, which was only driven by the desire to understand why we can have strikingly different results in different gauges in the dipole approximation. Recall from the introduction that the length gauge produced superior results in many instances. From the reductionist point of view, the question is not whether length gauge is valid. The question is why those systems are better described by states which are dressed in the form (45). On the other hand, if the length-gauge picture is considered more fundamental, the question arises why systems which are better analyzed in velocity gauge are better described by dressed states of the form (46).

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APPENDIX A: COHERENT STATES

The Coulomb-gauge photon field operator in the interaction picture is

$$\begin{aligned}\boldsymbol{A}(\boldsymbol{x},t) = \sqrt{\frac{\hbar\mu_0 c}{(2\pi)^3}}\int\frac{d^3\boldsymbol{k}}{\sqrt{2k}}\sum_{\alpha=1}^{2}\boldsymbol{\epsilon}_\alpha(\boldsymbol{k})\{a_\alpha(\boldsymbol{k})\\ \times\ \exp\left[i(\boldsymbol{k}\cdot\boldsymbol{x}-ckt)\right]\\ + a_\alpha^+(\boldsymbol{k})\exp\left[-i(\boldsymbol{k}\cdot\boldsymbol{x}-ckt)\right]\},\end{aligned}\tag{A1}$$

with $\boldsymbol{k}\cdot\boldsymbol{\epsilon}_\alpha(\boldsymbol{k})=0$, $\boldsymbol{\epsilon}_\alpha(\boldsymbol{k})\cdot\boldsymbol{\epsilon}_\beta(\boldsymbol{k})=\delta_{\alpha\beta}$, $[a_\alpha(\boldsymbol{k}),a_\beta(\boldsymbol{k}')]=0$, and $[a_\alpha(\boldsymbol{k}),a_\beta^+(\boldsymbol{k}')]=\delta_{\alpha\beta}\delta(\boldsymbol{k}-\boldsymbol{k}')$.

The corresponding electric- and magnetic-field operators

$$\boldsymbol{E}(\boldsymbol{x},t)=-\partial\boldsymbol{A}(\boldsymbol{x},t)/\partial t,\quad \boldsymbol{B}(\boldsymbol{x},t)=\nabla\times\boldsymbol{A}(\boldsymbol{x},t),$$

yield expectation values corresponding to a classical electromagnetic wave,

$$\begin{aligned}\langle\zeta|\boldsymbol{A}(\boldsymbol{x},t)|\boldsymbol{\zeta}\rangle &= \boldsymbol{\mathcal{A}}(\boldsymbol{x},t) = \sqrt{\frac{\hbar\mu_0 c}{(2\pi)^3}}\int\frac{d^3\boldsymbol{k}}{\sqrt{2k}}\\ &\quad\times\sum_{\alpha=1}^{2}\boldsymbol{\epsilon}_\alpha(\boldsymbol{k})\{\zeta_\alpha(\boldsymbol{k})\exp\left[i(\boldsymbol{k}\cdot\boldsymbol{x}-ckt)\right]\\ &\quad+\zeta_\alpha^+(\boldsymbol{k})\exp\left[-i(\boldsymbol{k}\cdot\boldsymbol{x}-ckt)\right]\},\end{aligned}\tag{A2}$$

$$\langle\zeta|\boldsymbol{E}(\boldsymbol{x},t)|\boldsymbol{\zeta}\rangle = \boldsymbol{\mathcal{E}}(\boldsymbol{x},t) = -\partial\boldsymbol{\mathcal{A}}(\boldsymbol{x},t)/\partial t,$$

$$\langle\zeta|\boldsymbol{B}(\boldsymbol{x},t)|\boldsymbol{\zeta}\rangle = \boldsymbol{\mathcal{B}}(\boldsymbol{x},t) = \nabla\times\boldsymbol{\mathcal{A}}(\boldsymbol{x},t),$$

if we use the coherent photon state [57]

$$|\boldsymbol{\zeta}\rangle=\exp\left\{\int d^3\boldsymbol{k}\left[\zeta(\boldsymbol{k})\cdot a^+(\boldsymbol{k})-\zeta^+(\boldsymbol{k})\cdot a(\boldsymbol{k})\right]\right\}|0\rangle,$$

where the definitions $\zeta(\boldsymbol{k})\cdot a^+(\boldsymbol{k})=\sum_{\alpha=1}^{2}\zeta_\alpha(\boldsymbol{k})a_\alpha^+(\boldsymbol{k})$ and $|\zeta(\boldsymbol{k})|^2=\sum_{\alpha=1}^{2}\zeta_\alpha^+(\boldsymbol{k})\zeta_\alpha(\boldsymbol{k})$ were used.

The expectation values for photon number, energy, and momentum in the coherent state are

$$\langle n\rangle=\int d^3\boldsymbol{k}|\zeta(\boldsymbol{k})|^2,\quad \langle H_0\rangle=\int d^3\boldsymbol{k}\hbar ck|\zeta(\boldsymbol{k})|^2,$$

and

$$\begin{aligned}\langle\boldsymbol{P}\rangle &= \left\langle\int d^3\boldsymbol{x}\epsilon_0\boldsymbol{E}(\boldsymbol{x},t)\times\boldsymbol{B}(\boldsymbol{x},t)\right\rangle\\ &= \int d^3\boldsymbol{k}\hbar\boldsymbol{k}|\zeta(\boldsymbol{k})|^2.\end{aligned}$$

APPENDIX B: ENERGY AND MOMENTUM IN COULOMB GAUGE

The Lagrange density for coupled electromagnetic and nonrelativistic matter fields without any particular choice of gauge is given by

$$\begin{aligned}\mathcal{L} = \sum_i\Bigg[&\frac{i\hbar}{2}\left(\psi_i^+\cdot\frac{\partial\psi_i}{\partial t}-\frac{\partial\psi_i^+}{\partial t}\cdot\psi_i\right)-q_i\psi_i^+\Phi\psi_i\\ &-\frac{\hbar^2}{2m_i}\nabla\psi_i^+\cdot\nabla\psi_i - i\frac{q_i\hbar}{2m_i}\boldsymbol{A}\cdot(\psi_i^+\overleftrightarrow{\nabla}\psi_i)\\ &-\frac{q_i^2}{2m_i}\psi_i^+\boldsymbol{A}^2\psi_i\Bigg]-\frac{1}{4\mu_0}F_{\mu\nu}F^{\mu\nu}.\end{aligned}\tag{B1}$$

Here $\Phi=-cA_0$ is the electric potential, and we use the definition of an alternating derivative operator $\psi^+\overleftrightarrow{\nabla}\psi\equiv\psi^+\cdot\nabla\psi-\nabla\psi^+\cdot\psi$. The summation over i in (B1) refers to different kinds of nonrelativistic particles (e.g., electrons, protons, etc.), and a summation over spin labels is implicitly understood.

The canonical energy-momentum tensor following from the Lagrange density (B1),

$$\begin{aligned}\Theta_\mu{}^\nu &= \eta_\mu{}^\nu\mathcal{L}+\frac{1}{\mu_0}\partial_\mu A_\lambda\cdot F^{\nu\lambda}\\ &\quad-\sum_i\left(\partial_\mu\psi_i\frac{\partial\mathcal{L}}{\partial(\partial_\nu\psi_i)}+\partial_\mu\psi_i^+\frac{\partial\mathcal{L}}{\partial\left(\partial_\nu\psi_i^+\right)}\right),\end{aligned}$$

is rendered gauge invariant in the usual way by adding the trivially conserved tensor

$$\delta\Theta_\mu{}^\nu=-\frac{1}{\mu_0}\partial_\lambda\left(A_\mu F^{\nu\lambda}\right)=-A_\mu j^\nu-\frac{1}{\mu_0}\partial_\lambda A_\mu\cdot F^{\nu\lambda},$$

$$\partial_\nu\delta\Theta_\mu{}^\nu\equiv 0.$$

The improved energy-momentum tensor $t_\mu{}^\nu=\Theta_\mu{}^\nu+\delta\Theta_\mu{}^\nu$ yields in particular the energy density $\mathcal{H}=-t_0{}^0$ for quantum optics,

$$\begin{aligned}\mathcal{H} = \frac{\epsilon_0}{2}\boldsymbol{E}^2+\frac{1}{2\mu_0}\boldsymbol{B}^2+\sum_i\frac{1}{2m_i}\Big[&\hbar^2\nabla\psi_i^+\cdot\nabla\psi_i\\ &+iq_i\hbar\boldsymbol{A}\cdot(\psi_i^+\overleftrightarrow{\nabla}\psi_i)+q_i^2\psi_i^+\boldsymbol{A}^2\psi_i\Big],\end{aligned}\tag{B2}$$

and the gauge-invariant momentum density $\mathcal{P}_a=t_a{}^0/c$,

$$\boldsymbol{\mathcal{P}}=\epsilon_0\boldsymbol{E}\times\boldsymbol{B}+\frac{1}{2i}\sum_i(\hbar\psi_i^+\overleftrightarrow{\nabla}\psi_i-2iq_i\psi_i^+\boldsymbol{A}\psi_i).\tag{B3}$$

We split the electric-field components in Coulomb gauge $\nabla\cdot\boldsymbol{A}=0$ according to $\boldsymbol{E}_\parallel=-\nabla\Phi$ and $\boldsymbol{E}_\perp=-\partial\boldsymbol{A}/\partial t$. The equation for the electrostatic potential decouples from the

vector potential,

$$\Delta\Phi = -\frac{1}{\epsilon_0}\sum_i q_i \psi_i^+ \psi_i,$$

and is solved by

$$\Phi(\boldsymbol{x},t) = \frac{1}{4\pi\epsilon_0}\int d^3\boldsymbol{x}' \sum_i \frac{q_i}{|\boldsymbol{x}-\boldsymbol{x}'|}\psi_i^+(\boldsymbol{x}',t)\psi_i(\boldsymbol{x}',t).$$

Furthermore, the two components of the electric field are orthogonal in the Coulomb gauge,

$$\begin{aligned}\int d^3\boldsymbol{x}\boldsymbol{E}_\parallel(\boldsymbol{x},t)\cdot\boldsymbol{E}_\perp(\boldsymbol{x},t) &= \int d^3\boldsymbol{k}\boldsymbol{E}_\parallel(\boldsymbol{k},t)\cdot\boldsymbol{E}_\perp(-\boldsymbol{k},t)\\ &= -\int d^3\boldsymbol{x}\Phi(\boldsymbol{x},t)\frac{\partial}{\partial t}\nabla\cdot\boldsymbol{A}(\boldsymbol{x},t) = 0, \qquad \text{(B4)}\end{aligned}$$

and the contribution from $\boldsymbol{E}_\parallel$ to the Hamiltonian generates the Coulomb potentials in the Hamiltonian,

$$\begin{aligned}H_C &= \frac{\epsilon_0}{2}\int d^3\boldsymbol{x}\boldsymbol{E}_\parallel^2(\boldsymbol{x},t) = -\frac{\epsilon_0}{2}\int d^3\boldsymbol{x}\Phi(\boldsymbol{x},t)\Delta\Phi(\boldsymbol{x},t)\\ &= \frac{1}{2}\int d^3\boldsymbol{x}\Phi(\boldsymbol{x},t)\varrho(\boldsymbol{x},t)\\ &= \sum_{ij}\int d^3\boldsymbol{x}\int d^3\boldsymbol{x}'\frac{q_iq_j}{8\pi\epsilon_0|\boldsymbol{x}-\boldsymbol{x}'|}\\ &\quad\times\psi_i^+(\boldsymbol{x},t)\psi_j^+(\boldsymbol{x}',t)\psi_j(\boldsymbol{x}',t)\psi_i(\boldsymbol{x},t). \qquad \text{(B5)}\end{aligned}$$

The resulting Hamiltonian in Coulomb gauge therefore has the form

$$\begin{aligned}H = \int d^3\boldsymbol{x}\Bigg(&\sum_i\frac{1}{2m_i}\big\{\hbar^2\nabla\psi_i^+(\boldsymbol{x},t)\cdot\nabla\psi_i(\boldsymbol{x},t)\\ &+iq_i\hbar\boldsymbol{A}(\boldsymbol{x},t)\cdot[\psi_i^+(\boldsymbol{x},t)\overset{\leftrightarrow}{\nabla}\psi_i(\boldsymbol{x},t)]\\ &+q_i^2\psi_i^+(\boldsymbol{x},t)\boldsymbol{A}^2(\boldsymbol{x},t)\psi_i(\boldsymbol{x},t)\big\}\\ &+\frac{\epsilon_0}{2}\boldsymbol{E}_\perp^2(\boldsymbol{x},t)+\frac{1}{2\mu_0}\boldsymbol{B}^2(\boldsymbol{x},t)\Bigg)\\ &+\sum_{ij}\int d^3\boldsymbol{x}\int d^3\boldsymbol{x}'\frac{q_iq_j}{8\pi\epsilon_0|\boldsymbol{x}-\boldsymbol{x}'|}\\ &\times\psi_i^+(\boldsymbol{x},t)\psi_j^+(\boldsymbol{x}',t)\psi_j(\boldsymbol{x}',t)\psi_i(\boldsymbol{x},t). \qquad \text{(B6)}\end{aligned}$$

The momentum operator in Coulomb gauge follows from Eq. (B3) and

$$\begin{aligned}\int d^3\boldsymbol{x}\epsilon_0\boldsymbol{E}_\parallel\times\boldsymbol{B} &= -\int d^3\boldsymbol{x}\epsilon_0\Phi\Delta\boldsymbol{A} = \int d^3\boldsymbol{x}\varrho\boldsymbol{A}\\ &= \int d^3\boldsymbol{x}\sum_i q_i\psi_i^+\boldsymbol{A}\psi_i\end{aligned}$$

as

$$\begin{aligned}\boldsymbol{P} &= \int d^3\boldsymbol{x}\left(\frac{\hbar}{i}\sum_i\psi_i^+\nabla\psi_i - \sum_i q_i\psi_i^+\boldsymbol{A}\psi_i + \epsilon_0\boldsymbol{E}\times\boldsymbol{B}\right)\\ &= \int d^3\boldsymbol{x}\left(\frac{\hbar}{i}\sum_i\psi_i^+\nabla\psi_i + \epsilon_0\boldsymbol{E}_\perp\times\boldsymbol{B}\right). \qquad \text{(B7)}\end{aligned}$$

All these results hold at the operator level within the full second-quantized theory [4], with the only approximation of nonrelativistic charged fields. If we now specify to the semiclassical theory in dipole approximation, the transformation to Schrödinger field operators in length gauge

$$\Psi_i(\boldsymbol{x},t) = \exp\left[-\frac{i}{\hbar}q_i\boldsymbol{x}\cdot\boldsymbol{A}(t)\right]\psi_i(\boldsymbol{x},t)$$

yields expressions [after leaving out the formally divergent kinetic contributions from the semiclassical fields $\boldsymbol{E}(t) = -d\boldsymbol{A}(t)/dt$ and $\boldsymbol{B}(t) = [\nabla\times\boldsymbol{A}(\boldsymbol{x},t)]_{k\cdot x\to 0}$ and neglecting the contributions from $\boldsymbol{A}_{\boldsymbol{J}}(\boldsymbol{x},t)$ (9) for the reasons pointed out after Eq. (11)]

$$\begin{aligned}H = \int d^3\boldsymbol{x}\Bigg[&\sum_i\frac{\hbar^2}{2m_i}\nabla\Psi_i^+(\boldsymbol{x},t)\cdot\nabla\Psi_i(\boldsymbol{x},t)\\ &-q_i\Psi_i^+(\boldsymbol{x},t)\boldsymbol{x}\cdot\boldsymbol{E}(t)\Psi_i(\boldsymbol{x},t)\Bigg]\\ &+\sum_{ij}\int d^3\boldsymbol{x}\int d^3\boldsymbol{x}'\frac{q_iq_j}{8\pi\epsilon_0|\boldsymbol{x}-\boldsymbol{x}'|}\\ &\times\Psi_i^+(\boldsymbol{x},t)\Psi_j^+(\boldsymbol{x}',t)\Psi_j(\boldsymbol{x}',t)\Psi_i(\boldsymbol{x},t), \qquad \text{(B8)}\end{aligned}$$

and

$$\begin{aligned}\boldsymbol{P} &= \int d^3\boldsymbol{x}\left[-i\hbar\sum_i\Psi_i^+(\boldsymbol{x},t)\nabla\Psi_i(\boldsymbol{x},t)+\epsilon_0\boldsymbol{E}_\parallel(\boldsymbol{x},t)\boldsymbol{B}(t)\right]\\ &= \int d^3\boldsymbol{x}\frac{\hbar}{i}\sum_i\Psi_i^+(\boldsymbol{x},t)\nabla\Psi_i(\boldsymbol{x},t) \qquad \text{(B9)}\\ &= \int d^3\boldsymbol{x}\frac{\hbar}{i}\sum_i\psi_i^+(\boldsymbol{x},t)\nabla\psi_i(\boldsymbol{x},t)\\ &\quad-\int d^3\boldsymbol{x}\sum_i q_i\psi_i^+(\boldsymbol{x},t)\boldsymbol{A}(t)\psi_i(\boldsymbol{x},t), \qquad \text{(B10)}\end{aligned}$$

since $\int d^3\boldsymbol{x}\boldsymbol{E}_\parallel(\boldsymbol{x},t)\times\boldsymbol{B}(t)$ is a surface term.

Both the momentum (B9) in terms of the length-gauge Schrödinger operators and the kinetic terms in Eq. (B8) support the identification $\boldsymbol{p} = m\boldsymbol{v}$, whereas the kinetic terms in Eq. (B6) and the expression (B10) agree with the general classical relation $\boldsymbol{p} = m\boldsymbol{v} + q\boldsymbol{A}$ for the canonical particle momentum in external fields.

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