

Arbitrary gauge quantization of light-matter theories with time-dependent constraints

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We provide a general framework for the quantization of light-matter theories with time-dependent holonomic constraints. Unless time dependence is present from the outset at the Lagrangian level, different gauges generally produce nonequivalent canonical theories. The irrotational gauge is defined as the one that also yields a correct theory when time dependence is introduced at the Hamiltonian level. Our framework unifies examples of such gauges found in existing literature. In particular, we show that for describing time-dependent light-matter interactions the Coulomb gauge is not generally irrotational, so it does not enjoy any special status.

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I. INTRODUCTION

Controlling microscopic light-matter interactions has an almost unbounded range of applications. For example, the active control of spacetime metamaterials for microwave engineering, photonics, and even acoustics is rapidly progressing [1–4], while quantum descriptions of time-varying media have also revealed novel phenomena [5–7]. The external control of superconducting devices allows state control [8] and stabilization [9,10], quantum switches [11], entanglement harvesting [12], fast quantum gates [13–16], and improved qubit connectivity [17,18]. Subcycle interaction switching with quantum wells [19] and ultrastrong switching with superconducting circuits [20] were achieved some time ago, and more recently using an electron gas [21]. Controlled ultrastrong interactions within electromagnetic cavities containing atoms and molecules [22–25], and solid-state emitters [26] also possess numerous applications, including chemical reaction control [27,28].

Modeling direct control requires the introduction of prescribed time dependence, which is also important for understanding loss and decoherence [29–32], as well as fundamental vacuum phenomena, such as the Unruh effect [33–35] and the dynamical Casimir effect [36,37]. The simplest method to achieve this is to swap out certain model parameters for time-dependent functions, but whether such a procedure is truly justified will often be unclear. In particular, it will generally result in nonequivalent theories when implemented in different gauges [38]. A number of recent articles have addressed the question of whether and how the resulting descriptions may or may not be valid [38–42].

A more refined approach, pursued here, is to prescribe physically motivated time-dependent constraints. We begin in Sec. II with a general presentation of the ways in which time dependence can be included in both Lagrangian and Hamiltonian (canonical) theories with holonomic constraints. We demonstrate that, in general, only if it is present within the constraints from the outset of the Lagrangian description, does a *unique* canonical description follow. This description, in turn, enables one to determine if the more naive approach of introducing time dependence phenomenologically at a later stage is valid, and if it is, which gauge provides the valid naive theory. The name given to this gauge is *irrotational* [43]. In Sec. III, we use the framework of Sec. II to consolidate disparate examples of the irrotational gauge found within existing literature [39,43]. The previously debated status of the Coulomb gauge for describing time-dependent light-matter interactions is determined conclusively by our demonstration that it does not generally coincide with the irrotational gauge. In Sec. IV, we explain how our results relate to those within existing literature.

II. TIME-DEPENDENT CONSTRAINTS

A. Elimination of coordinates using constraints

For simplicity, we restrict our attention to systems described by N discrete coordinates and velocities $\{x_n, \dot{x}_n\}$ with $n = 0, 1, \dots, N-1$, noting that the extension to include continuously labeled coordinates (fields) does not result in different conclusions. Without loss of generality, we may begin with a Lagrangian, $L\{x_n, \dot{x}_n\}$, that does not depend on time t . It is, however, assumed to be accompanied by a finite number of generally time-dependent holonomic constraints, $f_\mu^t\{x_n\} = 0$, where $\mu = 0, 1, \dots, M-1$ with $M < N$.

The constraints allow M of the coordinates x_n to be eliminated in favor of $N - M$ unconstrained coordinates $\{q_i^g : i = 0, 1, \dots, N'\}$, where $N' = N - (M + 1)$. We express this as

$$x_n = g_n^t\{q_i^g\}, \quad (1)$$

where the time dependence of the functions g_n^t is due to that of f_μ^t . There is generally no unique way, however, to eliminate

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$N - M$ of the x_n . A different elimination results in a different collection of functions $\{g'_n\}$ and different coordinates $q_i^{g'}$ such that $x_n = g'_n\{q_i^{g'}\}$. The sets of coordinates and velocities labeled by g and g' are related by a generally time-dependent bijective (invertible) point transformation:

$$\begin{aligned} q_i^{g'} &= q_i^g(\{q_j^g\}, t), \\ \dot{q}_i^{g'} &= \sum_{j=0}^{N'} \frac{\partial q_i^{g'}}{\partial q_j^g} \dot{q}_j^g + \partial_t q_i^{g'}(\{q_j^g\}, t). \end{aligned} \quad (2)$$

Writing the Lagrangian L in terms of the coordinates $\{q_i^g\}$ defines an explicitly time-dependent Lagrangian L_g^t as

$$L\{x_n, \dot{x}_n\} = L\{g'_n\{q_i^g\}, \dot{g}'_n\{q_i^g, \dot{q}_i^g\}\} =: L_g^t\{q_i^g, \dot{q}_i^g\}, \quad (3)$$

where

$$\dot{g}'_n = \sum_{j=0}^{N'} \frac{\partial g'_n}{\partial q_j^g} \dot{q}_j^g + \partial_t g'_n \quad (4)$$

is seen to be linearly dependent on the velocities $\dot{q}_j$. The Lagrangian L_g^t generally differs from any Lagrangian $L_g(t)$ that is obtained by phenomenologically introducing time dependence within the parameters of a time-independent Lagrangian L_g .

Any two Lagrangian functions L_g^t and $L_{g'}^t$ are distinct and are related using Eqs. (2) and (3) as $L_g^t\{q_i^g, \dot{q}_i^g\} = L_{g'}^t\{q_i^{g'}\{q_i^g, t\}, \dot{q}_i^{g'}\{q_i^g, \dot{q}_i^g, t\}\}$. Extending this formalism to deal with nonholonomic constraints is a difficult problem. We can, however, include some such cases by allowing $L_g^t\{q_i^g, \dot{q}_i^g\}$ and $L_{g'}^t\{q_i^{g'}, \dot{q}_i^{g'}\}$ to differ by a total time derivative of a function of the coordinates as

$$L_{g'}^t\{q_j^{g'}, \dot{q}_j^{g'}\} = L_g^t\{q_j^g, \dot{q}_j^g\} + \frac{d}{dt} F_{gg'}^t\{q_j^g\}. \quad (5)$$

In this case, the two Lagrangians, although not equal, remain equivalent.

Our treatment covers a broad class of systems, with two representative subclasses discussed in Sec. III. The systems excluded are those subject to nonholonomic constraints that cannot be reduced to holonomic systems via gauge transformations. A classic example is a nonslipping rolling wheel. A description of such systems requires the nonvariational Lagrange-d'Alembert principle, and although their external control could conceivably be of interest, this lies beyond the present scope.

B. The canonical formalism and the irrotational gauge

Our aim now is to compare the results of introducing an explicit time dependence prior to the construction of the Lagrangian versus afterward, for instance, at the Hamiltonian level.

1. Hamiltonian

Construction of the Hamiltonian proceeds in the usual way. We assume that the imposition of constraints has necessarily resulted in a nondegenerate Lagrangian, such that there are no

identically zero canonical momenta. The latter are defined by

$$p_j^g = \frac{\partial L_g^t}{\partial \dot{q}_j^g}, \quad (6)$$

where L_g^t is given in Eq. (3). For different g , the canonical momenta are related by

$$p_i^{g'} = \sum_{j=0}^{N'} p_j^g \frac{\partial q_j^g}{\partial q_i^{g'}} + \frac{\partial F_{gg'}^t}{\partial q_i^{g'}}, \quad (7)$$

from which it follows that $\partial p_i^{g'}/\partial p_j^g = \partial q_j^g/\partial q_i^{g'}$, and therefore that the transformation $g \leftrightarrow g'$ is canonical. The velocities $\dot{q}_i^g$ and canonical momenta are in one-to-one correspondence, such that the former can be written as time-dependent functions of the canonical variables: $\dot{q}_j^g \equiv \dot{q}_j^g(q_i^g, p_i^g, t)$. The Hamiltonian is then defined as

$$H_g^t\{q_i^g, p_i^g\} := \sum_{j=0}^{N'} p_j^g \dot{q}_j^g\{q_i^g, p_i^g, t\} - L_g^t\{q_j^g, \dot{q}_j^g\{q_i^g, p_i^g, t\}\} \quad (8)$$

and it is explicitly time dependent. For different g , the Hamiltonians are related by

$$\begin{aligned} H_{g'}^t\{q_i^{g'}, p_i^{g'}\} &= H_g^t\{q_i^g, p_i^g\} + \sum_{i=0}^{N'} p_i^{g'} \partial_t q_i^{g'}(\{q_j^g\}, t) \\ &\quad - \partial_t F_{gg'}^t\{q_i^g\}. \end{aligned} \quad (9)$$

We have therefore constructed a physically unique canonical theory, including a description of any external control that can be described using holonomic constraints and gauge transformations.

Upon quantization, the canonical variables become canonical operators and we consider dynamics in the Schrödinger picture, meaning operators change only due to explicit time dependence. The transformation $g \leftrightarrow g'$ is implemented by an explicitly time-dependent unitary operator $U_{gg'}(t)$ as

$$q_i^{g'}(\{q_j^g\}, t) = U_{gg'}(t)^\dagger q_i^g U_{gg'}(t), \quad (10)$$

$$p_i^{g'}(\{q_j^g, p_j^g\}, t) = U_{gg'}(t)^\dagger p_i^g U_{gg'}(t). \quad (11)$$

Each of the canonical operator sets (labeled by g and g') is explicitly time dependent when expressed in terms of the other set. It is sufficient for us to consider separately the limiting cases of (1) a pure point transformation, $F_{gg'} \equiv 0$, and (2) a pure “gauge” transformation $q_i^{g'} = q_i^g$. A general treatment of point transformations [case (1)] can be found in Ref. [44]. We will restrict our attention to pure translations such that $\partial q_i^g/\partial q_j^{g'} = \delta_{ij}$. This suffices to provide a clear example in Sec. III of the significance of different treatments of explicit time dependence. The unitary transformation required to translate each coordinate q_i by an arbitrary time-dependent function $c_i(t)$ is simply

$$\begin{aligned} U_{gg'}(t) &= \exp(iS_{gg'}^t), \\ S_{gg'}^t &= \sum_{i=0}^{N'} c_i(t) p_i^g. \end{aligned} \quad (12)$$

Equations (10) and (11) read $q_i^{g'}(\{q_j^g\}, t) = q_i^g - c_i(t)$ and $p_i^{g'} = p_i^g$, respectively, while Eq. (9) reads

$$H_g^t\{q_i^{g'}, p_i^{g'}\} = H_g^t\{q_i^g, p_i^g\} - \partial_t S_{gg'}^t. \quad (13)$$

Letting $q_i^g \equiv q_i$ and $p_i \equiv p_i^g$, and noting that $\partial_t S_{gg'}^t = -i\dot{U}_{gg'}(t)U_{gg'}(t)^\dagger = i\dot{U}_{gg'}(t)^\dagger U_{gg'}(t)$ and $\partial_t[U_{gg'}(t)U_{gg'}(t)^\dagger] \equiv 0$, we see using Eqs. (10) and (11) that the two Hamiltonians H_g^t and $H_{g'}^t$ are equivalent:

$$H_g^t\{q_i, p_i\} = U_{gg'}(t)H_{g'}^t\{q_i, p_i\}U_{gg'}(t)^\dagger + i\dot{U}_{gg'}(t)U_{gg'}(t)^\dagger. \quad (14)$$

In case (2) of a pure “gauge” transformation, $q_i^{g'} = q_i^g$, the required unitary transformation is

$$U_{gg'}(t) = \exp(iF_{gg'}^t) \quad (15)$$

such that Eqs. (10) and (11) read $q_i^{g'} = q_i^g$ and $p_i^{g'} = p_i^g + \partial F_{gg'}^t / \partial q_i^g$, respectively, while Eq. (9) reads

$$H_{g'}^t\{q_i^{g'}, p_i^{g'}\} = H_g^t\{q_i^g, p_i^g\} - \partial_t F_{gg'}^t. \quad (16)$$

Letting $q_i^g \equiv q_i$ and $p_i \equiv p_i^g$, and applying the same manipulations that lead from Eq. (13) to Eq. (14), one again obtains Eq. (14).

2. Naive Hamiltonians and the irrotational gauge

Let us now suppose that the theory does not possess any explicit time dependence through either the constraints $f_\mu = 0$ or the gauge function $F_{gg'}$. The point transformation $q_i^{g'} = q_i^g\{q_j^g\}$ is then also time independent as are Eqs. (10) and (11) for the quantum canonical transformation $g \leftrightarrow g'$. Equation (9) becomes simply $H_g^t\{q_i^{g'}, p_i^{g'}\} = H_g\{q_i^g, p_i^g\}$. Letting $q_i^g \equiv q_i$ and $p_i \equiv p_i^g$, one therefore obtains

$$H_{g'}\{q_i, p_i\} = U_{gg'}H_g\{q_i, p_i\}U_{gg'}^\dagger. \quad (17)$$

Suppose now that we introduce time dependence into the Hamiltonian H_g in the Schrödinger picture as $H_g \rightarrow H_g(t)$. This could, for example, be the result of introducing t dependence into the functions g_n upon which H_g depends, resulting in concurrent t dependence of the g'_n and $U_{gg'}$. However, if the functional forms of H_g , $H_{g'}$, and $U_{gg'}$ in terms of the canonical operators remain unchanged, then Eq. (17) will continue to hold in the form $H_{g'}^t(t) = U_{gg'}(t)H_g(t)U_{gg'}(t)^\dagger$. The Hamiltonians $H_g(t)$ and $H_{g'}(t)$ obtained in this way are therefore not equivalent. An equivalence class of Hamiltonians can of course be constructed for each fixed g by allowing g' to vary as

$$\mathcal{S}_g = \{H_{g'}^t(t) = H_g(t) + i\dot{U}_{gg'}(t)U_{gg'}(t)^\dagger : g' \text{ variable}\}, \quad (18)$$

but distinct such classes $\mathcal{S}_g$ and $\mathcal{S}_{g'}$ are not equivalent. Thus, introducing time dependence at the Hamiltonian level does not in general result in a unique (unambiguous) description.

The freedom to choose among the different descriptions labeled by g will henceforth be called gauge freedom. In general, one cannot guarantee the existence of a gauge for

which

$$X_g(t) := H_g^t - H_g(t) = 0, \quad (19)$$

where H_g^t is obtained by assuming explicit time dependence from the outset as in the previous section. However, if such a gauge does exist, its naive model $H_g(t)$ is physically valid, because it coincides with H_g^t . We adopt the terminology proposed in Ref. [43] and call such gauges *irrotational*. Given any naive model $H_g(t)$, one can derive a correct model from it as $H_{g_{\text{irr}}}^t(t) = U_{gg_{\text{irr}}}(t)H_g(t)U_{gg_{\text{irr}}}(t)^\dagger$, without any additional contributions arising from the time dependence of the rotation. Note, however, that the naive g -gauge model $H_g(t)$ is not itself correct unless $g = g_{\text{irr}}$. The correct g -gauge model, H_g^t , can be obtained from the irrotational gauge model by inserting $H_{g_{\text{irr}}}^t \equiv H_{g_{\text{irr}}}(t)$ into the right-hand side of Eq. (14). Letting $U_{gg'}(t) = \exp[i\mathcal{G}_{gg'}^t]$, this gives an expression for $X_g(t)$ in terms of g_{irr} as

$$X_g(t) := i\dot{U}_{g_{\text{irr}}g}(t)U_{g_{\text{irr}}g}(t)^\dagger = -\partial_t \mathcal{G}_{g_{\text{irr}}g}^t. \quad (20)$$

For the examples of a translation and a pure gauge transformation given in the previous section, we have $\mathcal{G}_{gg'}^t = S_{gg'}^t$ and $\mathcal{G}_{gg'}^t = F_{gg'}^t$, respectively.

III. APPLICATION TO LIGHT-MATTER SYSTEMS

We provide two example applications of the framework presented in Sec. II.

A. Superconducting circuit with variable external flux

Reference [43] presents a systematic approach to quantizing electrical circuits that contain loops threaded by *external* fluxes. The authors consider an arbitrary circuit consisting of N elements spread across M primitive loops L_j (each not containing any further loops). Denoting the external flux threading loop L_j by $\phi_j(t)$, and denoting the n th branch flux by x_n , there are M linear constraints expressible in the form $\Phi(t) = R\mathbf{x}$, where $\Phi(t) = (\phi_0(t), \dots, \phi_{M-1}(t))$, $\mathbf{x} = (x_0, \dots, x_{N-1})$, and R is an $M \times N$ matrix. It is defined by $R_{jn} = 1$ (respectively, $R_{jn} = -1$), when $\phi_j(t)$ and $x_n \in L_j$ possess the same (respectively, opposite) orientations, while $R_{jn} = 0$ if $x_n \notin L_j$. The x_n are also related to the $N - M$ unconstrained but gauge-dependent fluxes $\mathbf{q}^g = (q_0^g, \dots, q_{N-M-1}^g)$ using an $(N - M) \times N$ gauge matrix $g = G$, as $\mathbf{q}^g = G\mathbf{x}$. Defining $\mathbf{Q}^g := (\mathbf{q}^g, \Phi(t))$, one obtains $\mathbf{Q}^g = S\mathbf{x}$, where the $N \times N$ matrix $S = (G, R)$ is, by construction, invertible; $\det S \neq 0$, such that $\mathbf{x} = S^{-1}\mathbf{Q}^g$ [43]. This last expression is a particular instance of Eq. (1) in which the x_n happen to be *linear combinations* of the variables q_i^g and the time-dependent functions $\phi_j(t)$, with time-independent coefficients S_{nm} . Clearly then, the construction of a canonical description of a circuit threaded by external fluxes constitutes an example application of the general framework we have provided in Sec. II.

By way of explicit demonstration, we choose the simplest example presented in Ref. [43]: a superconducting circuit consisting of two junctions labeled right ($n = 0$) and left ($n = 1$) with corresponding capacitances C_n and Josephson energies E_{Jn} . The junctions are connected via two nodes, one of which is the ground node. A generally time-dependent external flux

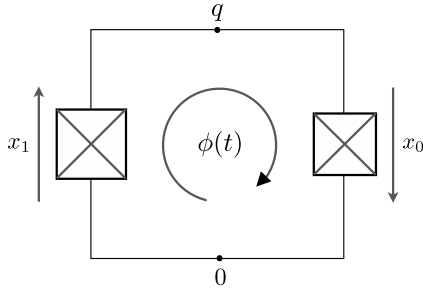


FIG. 1. Two Josephson junctions form a closed loop, L_0 , which is threaded by an external flux $\phi(t)$. The right and left branch fluxes are x_0 and x_1 , respectively. Since x_0 and x_1 both possess the same orientation as $\phi(t)$, we have $R_{00} = 1 = R_{01}$. The lower node (0) is designated as ground, and the nonground node flux is $q = G\mathbf{x}$. The latter linear combination of the x_n is determined by the choice of spanning tree (gauge).

$\phi(t)$ threads the circuit loop, as depicted in Fig. 1. Since there is only a single loop, both branch fluxes x_n possess the same orientation as $\phi(t)$, such that

$$\phi(t) = R\mathbf{x} = x_0 + x_1, \quad R = \begin{pmatrix} 1 & 1 \end{pmatrix}. \quad (21)$$

In Ref. [43], the choice of spanning tree (gauge g) is encoded into two parameters, denoted therein by m_1 and m_r . These determine the circuit's nonground flux coordinate q in terms of the branch fluxes via the 1×2 matrix $G = (m_r \ m_1)$, giving $q = m_r x_0 + m_1 x_1$ [45]. The standard gauge choices $(m_r \ m_1) = (0 \ 1)$ and $(m_r \ m_1) = (-1 \ 0)$ are particular cases that assign the flux q entirely to the right and left branches, respectively. One can alternatively express q in terms of the sum $m_+ = m_1 + m_r$ and the difference $m_\Delta = m_1 - m_r$. Here, we will restrict our attention to the cases defined by $m_\Delta = 1$ and $m_1 = \alpha \in [0, 1]$. This suffices to reveal the important physical conclusions, while allowing us to encode the gauge choice g more simply into the single parameter $g = \alpha$, the corresponding matrix being $G = (\alpha \ -1 \ \alpha)$. We continue to be able to smoothly interpolate between the standard gauges of choosing the right arm as spanning tree ($\alpha = 0$) and choosing the left arm as spanning tree ($\alpha = 1$).

In terms of α , the nonground node flux $q = m_1 x_1 + m_r x_0$ is

$$q = G\mathbf{x} = \alpha x_1 - (1 - \alpha)x_0, \quad (22)$$

so by letting $\mathbf{Q} = (q, \phi(t))$, one obtains $\mathbf{Q} = S\mathbf{x}$, where

$$S = \begin{pmatrix} \alpha - 1 & \alpha \\ 1 & 1 \end{pmatrix}. \quad (23)$$

One can eliminate x_1 and x_0 in favor of q and $\phi(t)$ using Eqs. (21) and (22) to obtain [cf. Eq. (1)]

$$x_0 = g'_{\alpha,0}(q) = \alpha\phi(t) - q, \quad (24)$$

$$x_1 = g'_{\alpha,1}(q) = (1 - \alpha)\phi(t) + q, \quad (25)$$

or more compactly, $\mathbf{x} = S^{-1}\mathbf{Q}$. Each different gauge α defines a different set of time-dependent functions $g'_{\alpha,n}$.

The Lagrangian is the difference between the physical kinetic and potential energies:

$$L\{x_n, \dot{x}_n\} = \sum_{n=0,1} \left[\frac{1}{2} C_n \dot{x}_n^2 - V_n(x_n) \right], \quad (26)$$

where

$$V_n(x_n) = -E_{Jn} \cos\left(\frac{2\pi}{\varphi} x_n\right) \quad (27)$$

is the potential energy of the n th junction, in which φ is the elementary flux quantum. Let us first suppose that the external flux $\phi(t) \equiv \phi$ is steady, in which case the $g'_{\alpha,n} \equiv g_{\alpha,n}$ are time independent. Defining $L_\alpha(q, \dot{q}) := L\{g_{\alpha,n}(q), \dot{g}_{\alpha,n}(q, \dot{q})\}$ and $p := \partial L_\alpha / \partial \dot{q}$, one finds the corresponding Hamiltonian H_α to be

$$H_\alpha(q, p) = \frac{p^2}{2C} + V_0(\alpha\phi - q) + V_1([1 - \alpha]\phi + q), \quad (28)$$

where $C := C_0 + C_1$. The Hamiltonians of different gauges are unitarily related as in Eq. (17) by

$$H_{\alpha'} = R_{\alpha\alpha'} H_\alpha R_{\alpha\alpha'}^\dagger, \quad (29)$$

where

$$R_{\alpha\alpha'} := e^{iS_{\alpha\alpha'}}, \quad S_{\alpha\alpha'} := (\alpha - \alpha')p\phi. \quad (30)$$

The canonical theory so obtained is physically unique and unambiguous only for a steady flux. Indeed, if we denote by $H_\alpha(t)$ and $R_{\alpha\alpha'}(t)$ the result of making the replacement $\phi \rightarrow \phi(t)$ within Eqs. (28) and (30), respectively, then Eq. (29) becomes $H_{\alpha'}(t) = R_{\alpha\alpha'}(t) H_\alpha(t) R_{\alpha\alpha'}(t)^\dagger$, such that $H_\alpha(t)$ and $H_{\alpha'}(t)$ are not equivalent.

If one instead assumes that the external flux is time dependent from the outset and defines $L_\alpha^t(q, \dot{q}) = L\{g'_{\alpha,n}(q), \dot{g}'_{\alpha,n}(q, \dot{q})\}$ and $p := \partial L_\alpha^t / \partial \dot{q}$ as in Eqs. (3) and (6), one finds the Hamiltonian given by Eq. (8) to be

$$H_\alpha^t = H_\alpha(t) + X_\alpha(t), \quad (31)$$

where

$$X_\alpha(t) := \left(\alpha - \frac{C_1}{C} \right) p \dot{\phi}(t). \quad (32)$$

In writing Eq. (31), we have dropped the purely external kinetic term proportional to $\phi(t)^2$, which only affects the energy zero point. In the Schrödinger picture, we immediately have that

$$X_\alpha(t) - X_{\alpha'}(t) = \partial_t S_{\alpha\alpha'}^t, \quad (33)$$

where

$$\partial_t S_{\alpha\alpha'}^t := (\alpha - \alpha') p \dot{\phi}(t) = -i \dot{R}_{\alpha\alpha'}(t) R_{\alpha\alpha'}(t)^\dagger, \quad (34)$$

and since $R_{\alpha\alpha'}(t) H_\alpha(t) R_{\alpha\alpha'}(t)^\dagger = H_{\alpha'}(t)$, it follows from Eq. (31) that

$$H_{\alpha'}^t = R_{\alpha\alpha'}(t) H_\alpha^t R_{\alpha\alpha'}(t)^\dagger + i \dot{R}_{\alpha\alpha'}(t) R_{\alpha\alpha'}(t)^\dagger \quad (35)$$

as in Eq. (14). We have therefore now obtained a physically unique time-dependent description.

According to Eqs. (19) and (31), the irrotational gauge for which the naive Hamiltonian $H_\alpha(t)$ is correct is the solution $\alpha = \alpha_{\text{irr}}$ of $X_\alpha(t) \equiv 0$, such that for any α we have

$X_\alpha(t) = -\partial_t S_{\alpha_{\text{irr}}}$, as in Eq. (20). According to Eq. (32), the required value is

$$\alpha_{\text{irr}} := \frac{C_1}{C_0 + C_1}. \quad (36)$$

Notice that this is not a constant value independent of other model parameters, meaning that its value depends on the underlying physical context. The extremal right (left) gauge $\alpha = 0$ ($\alpha = 1$) becomes effectively irrotational only when the right (left) capacitance dominates the left (right) capacitance: $C_0 \gg C_1$ ($C_1 \gg C_0$). Concurrently, the node flux q of the irrotational gauge,

$$q = \frac{1}{C}(C_1 x_1 - C_0 x_0), \quad (37)$$

is a quasirelative flux; it is the difference between the physical fluxes after each one has been weighted by the ratio of the corresponding junction capacitance to the total capacitance.

B. A moving atom

As a second example, we consider the description of time-dependent light-matter interactions [39–41]. Introducing a phenomenological modulation $\mu(t)$ within the light-matter interaction Hamiltonians of different gauges results in nonequivalent models [38,39]. This is another example of the general result given above in Sec. II B 2, and it necessitates a more fundamental description of the underlying control mechanism. In practice, shuttling material systems around is one of only a few methods available to modulate a light-matter interaction, and so we here use holonomic constraints to describe such motion explicitly.

1. System and constraints

For simplicity, we consider a bound material system consisting of only two charges $e_n = \mp e$, $n = 0, 1$, with masses m_n and positions $\mathbf{r}_n$. The densities of charge and current are

$$\rho(\mathbf{x}) \equiv \rho(\mathbf{x}, \{\mathbf{r}_n\}) = \sum_{n=0,1} e_n \delta(\mathbf{x} - \mathbf{r}_n), \quad (38)$$

$$\mathbf{J}(\mathbf{x}) \equiv \mathbf{J}(\mathbf{x}, \{\mathbf{r}_n\}) = \sum_{n=0,1} e_n \dot{\mathbf{r}}_n \delta(\mathbf{x} - \mathbf{r}_n), \quad (39)$$

where $\mathbf{x}$ denotes a point in space. These densities satisfy identically the local conservation of charge:

$$\partial_\mu j^\mu = \dot{\rho} + \nabla \cdot \mathbf{J} = 0, \quad (40)$$

where $j^0 = \rho$, $j^i = (\mathbf{J})_i$, and repeated spacetime indices are summed. Equation (40) does not have to be imposed as a constraint. To describe the gross motion of the two-charge system, however, we introduce the holonomic constraint

$$m_0 \mathbf{r}_0 + m_1 \mathbf{r}_1 = M \mathbf{R}(t), \quad (41)$$

where $M := m_1 + m_2$ and the center-of-mass position $\mathbf{R}(t)$ is assumed to be explicitly time dependent. The explicitly time-independent reduced coordinates for the material system are the components of the relative position $\mathbf{r} = \mathbf{r}_0 - \mathbf{r}_1$. The electromagnetic field is described by potential A and the field tensor $F = dA$ possesses components $F_{\mu\nu} = \partial_\mu A_\nu - \partial_\nu A_\mu$.

The longitudinal and transverse parts of the vector potential $\mathbf{A}$ are gauge dependent and gauge invariant, respectively.

We encode gauge freedom into the real parameter α and choose the holonomic constraint [38,46–49]

$$\int d^3x \mathbf{g}_\alpha(\mathbf{x}, \mathbf{x}', \mathbf{R}(t)) \cdot \mathbf{A}(\mathbf{x}) = 0 \quad (42)$$

in which $\mathbf{g}_\alpha = \mathbf{g}_L + \mathbf{g}_{\alpha T}$, where

$$\mathbf{g}_L(\mathbf{x}, \mathbf{x}', \mathbf{o}) := -\nabla \frac{1}{4\pi |\mathbf{x} - \mathbf{x}'|} \quad (43)$$

is independent of $\mathbf{o}$ and α , and

$$\begin{aligned} \mathbf{g}_{\alpha T}(\mathbf{x}, \mathbf{x}', \mathbf{o}) &:= -\alpha \int_0^{\mathbf{x}'} d\mathbf{s}_\lambda \cdot \delta^T(\mathbf{x} - \mathbf{s}), \\ \mathbf{s}_\lambda &:= \mathbf{o} + \lambda[\mathbf{x}' - \mathbf{o}], \quad \lambda \in [0, 1]. \end{aligned} \quad (44)$$

The transverse δ function possesses the Fourier representation

$$\delta_{ij}^T(\mathbf{x}) = \int \frac{d^3k}{(2\pi)^3} (\delta_{ij} - \hat{k}_i \hat{k}_j) e^{i\mathbf{k} \cdot \mathbf{x}}, \quad (45)$$

which is seen to be the difference between $\delta_{ij}\delta(\mathbf{x})$ and the corresponding longitudinal δ function $\delta_{ij}^L(\mathbf{x})$. Constraint (42) is sufficiently general to yield the Coulomb ($\alpha = 0$) and multipolar ($\alpha = 1$) gauges as particular cases. The α -gauge polarization field

$$\mathbf{P}_\alpha(\mathbf{x}) := - \int d^3x' \mathbf{g}_\alpha(\mathbf{x}, \mathbf{x}', \mathbf{R}(t)) \rho(\mathbf{x}') \quad (46)$$

satisfies $-\nabla \cdot \mathbf{P}_\alpha = -\nabla \cdot \mathbf{P}_L = \rho$ identically. The α -independent longitudinal part is the Coulomb gauge polarization $\mathbf{P}_L = \nabla \phi_{\text{Coul}}$, where

$$\phi_{\text{Coul}}(\mathbf{x}) := -(\nabla^{-2} \rho)(\mathbf{x}) = \int d^3x' \frac{\rho(\mathbf{x}')}{4\pi |\mathbf{x} - \mathbf{x}'|} \quad (47)$$

is the Coulomb potential.

Line integral expressions of the type in Eq. (44) were introduced by Dirac relatively early on Ref. [50]. Most recently, Woolley has provided a discussion in Ref. [51]. Equation (44) can be motivated by first noting that the Coulomb field $-\mathbf{E}_L = \mathbf{P}_L$ of our neutral atom can be written as

$$\mathbf{P}_L(\mathbf{x}) = q \int_0^{\mathbf{r}} d\mathbf{s} \cdot \delta^L(\mathbf{x} - \mathbf{s}) \quad (48)$$

independent of both the choice of origin $\mathbf{o}$ and specific path $\mathbf{s}$ that ends at $\mathbf{r}$. For the choices $\mathbf{o} = \mathbf{R}(t)$ and $\mathbf{s}$ a straight line, Eq. (44) gives for $\alpha = 1$ the total multipolar polarization

$$\mathbf{P}_1(\mathbf{x}) = \mathbf{d} \int_0^1 d\lambda \delta(\mathbf{x} - \mathbf{R}(t) - \lambda[\mathbf{r} - \mathbf{R}(t)]). \quad (49)$$

Unlike the Coulomb field $\mathbf{P}_L$, this field is strictly *localized inside the atom*, and since the canonical formalism yields the transverse photonic momentum $\mathbf{\Pi} = -\mathbf{E} - \mathbf{P}$ [46,52], we see that whether $\mathbf{\Pi}$ is locally connected to the local field $\mathbf{E}$ depends on the localization properties of $\mathbf{P}$. In the Coulomb gauge $\mathbf{P} = \mathbf{P}_L = -\mathbf{E}_L$ is nonlocal, and so therefore, is $\mathbf{\Pi} = -\mathbf{E}_T = -\mathbf{E} + \mathbf{E}_L$. In contrast, for any origin $\mathbf{o}$ and path $\mathbf{s}$ confined within the atom, the line-integral form of $\mathbf{g}_T$ [Eq. (44)] yields $\mathbf{\Pi} = -\mathbf{E}$ at all points outside the atom, providing the best possible transverse representation of the total electric

field [52]. The PZW straight line choice in Eq. (44) is particularly noteworthy because it enables a simple multipolar development of the polarization and corresponding magnetization fields. In particular, the interaction Hamiltonian can be expanded in a series of multipole moments, facilitating a quantum description that parallels classical multipolar radiation theory.

Healy showed that the line integral form of $\mathbf{g}_T$ does not in fact provide the most general partition of the charge and current densities into polarization and magnetization fields [53]. One could also consider imposing more general gauge constraints that, for example, are nonlinear in $\mathbf{A}$, unlike Eq. (42) [54]. Here, however, such generalizations offer little advantage: Our simple α -gauge formalism is the minimal extension needed to track how gauge choice affects an explicitly time-dependent canonical theory.

2. Hamiltonian

We now proceed with the construction of the canonical description. The two time-dependent holonomic constraints, Eqs. (41) and (42), determine the constrained variables $\mathbf{r}_0$, $\mathbf{r}_1$, and $\mathbf{A} = \mathbf{A}_T + \mathbf{A}_L$, in terms of two unconstrained variables, $\mathbf{r} = \mathbf{r}_0 - \mathbf{r}_1$, and $\mathbf{A}_T$, and the time-dependent external vector $\mathbf{R}(t)$, as [cf. Eq. (1)]

$$\mathbf{r}_0 = \mathbf{g}_0^t(\mathbf{r}) := \mathbf{R}(t) + \frac{m_1}{M}\mathbf{r}, \quad (50)$$

$$\mathbf{r}_1 = \mathbf{g}_1^t(\mathbf{r}) := \mathbf{R}(t) - \frac{m_0}{M}\mathbf{r}, \quad (51)$$

$$\mathbf{A}(\mathbf{x}) = \mathbf{g}_{2,\alpha}^t([\mathbf{A}_T], \mathbf{x}) := \mathbf{A}_T(\mathbf{x}) + \nabla \chi_\alpha([\mathbf{A}_T], \mathbf{x}, \mathbf{R}(t)), \quad (52)$$

$$\chi_\alpha([\mathbf{A}_T], \mathbf{x}, \mathbf{R}(t)) := \int d^3x' \mathbf{g}_\alpha(\mathbf{x}', \mathbf{x}, \mathbf{R}(t)) \cdot \mathbf{A}_T(\mathbf{x}'). \quad (53)$$

Any function F of the $\mathbf{r}_n$ and $\mathbf{A}_L$ becomes explicitly time dependent when written in terms of $\mathbf{r}$, $\mathbf{A}_T$, and $\mathbf{R}(t)$, and we will henceforth abbreviate it as $F^t := F(\mathbf{r}_1(\mathbf{r}, \mathbf{R}(t)), \mathbf{r}_2(\mathbf{r}, \mathbf{R}(t)), [\mathbf{A}_L([\mathbf{A}_T], \mathbf{R}(t))])$. Simple examples depending only on the $\mathbf{r}_n$ and their velocities include ρ^t , $\mathbf{J}^t$, and $\mathbf{P}_{\alpha}^t$.

We must now construct a Lagrangian function of $\mathbf{r}$, $\mathbf{A}_T$, and $\mathbf{R}(t)$, and their velocities. The standard QED Lagrangian assuming nonrelativistic charges is

$$L(\mathbf{r}_1, \mathbf{r}_2, [A_\mu, (dA)_{\mu\nu}]) = \frac{1}{2}m_1\dot{\mathbf{r}}_1 + \frac{1}{2}m_2\dot{\mathbf{r}}_2 - \int d^3x \left(j^\mu A_\mu + \frac{1}{4}F^{\mu\nu}F_{\mu\nu} \right), \quad (54)$$

which is subject not only to the holonomic constraints (41) and (42), but also to Gauss' law

$$\nabla \cdot \mathbf{E} = \rho \quad (55)$$

in which the electric field $\mathbf{E} = -\dot{\mathbf{A}} - \nabla A_0$ depends on $\dot{\mathbf{A}}$. One way to arrive at the correct canonical quantum theory in this case is to follow Dirac by systematically constructing Lie brackets consistent with the constraints [38,46–49]. However, the same final result can be obtained more straightforwardly by judiciously imposing the constraints at the outset. This yields an α - and t -dependent Lagrangian function of the unconstrained coordinates and velocities, $L_\alpha^t[\mathbf{r}, \mathbf{A}_T, \dot{\mathbf{r}}, \dot{\mathbf{A}}_T]$,

which is of exactly the type appearing in Eq. (3). Subsequently, the procedure in Sec. II can be applied, including textbook quantization [46]. To this end, note first that since $\mathbf{E} = -\partial_t \mathbf{A}_T - \nabla \phi_{\text{Coul}} = -\partial_t \mathbf{A} - \nabla A_0$ is manifestly gauge invariant, Eq. (52) suffices to fix A_0 up to a constant as

$$A_0 = \phi_{\text{Coul}} - \partial_t \chi_\alpha. \quad (56)$$

Gauss' law (55) implies further that $\mathbf{E}_L = -\mathbf{P}_L = -\nabla \phi_{\text{Coul}}$, such that

$$\int d^3x \mathbf{E}_L(\mathbf{x})^2 = \int d^3x \phi_{\text{Coul}}(\mathbf{x}) \rho(\mathbf{x}) = -\frac{q^2}{2\pi|\mathbf{r}|} =: 2V(\mathbf{r}) \quad (57)$$

depends only on $\mathbf{r}$ and does not therefore depend explicitly on t . We have dropped here the infinite Coulomb self-energies of the individual charges.

Using Eqs. (40), (56), and (57), the Lagrangian in Eq. (54) can be written as [39]

$$L_\alpha^t[\mathbf{r}, \mathbf{A}_T, \dot{\mathbf{r}}, \dot{\mathbf{A}}_T] := \frac{1}{2}m\dot{\mathbf{r}}^2 + \frac{1}{2}M\dot{\mathbf{R}}(t)^2 - V(\mathbf{r}) + \int d^3x \left[\frac{1}{2}(\dot{\mathbf{A}}_T^2 - (\nabla \times \mathbf{A}_T)^2) + \mathbf{J}^t \cdot \mathbf{A}_T - \frac{d}{dt}(\mathbf{P}_{\alpha T}^t \cdot \mathbf{A}_T) \right], \quad (58)$$

where $m := m_1 m_2 / M$. Lagrangians of different gauges are related as in Eq. (5) by

$$L_{\alpha'}^t - L_\alpha^t = \frac{dF_{\alpha\alpha'}^t}{dt}, \quad (59)$$

where

$$F_{\alpha\alpha'}^t = \int d^3x [\mathbf{P}_{\alpha T}^t(\mathbf{x}) - \mathbf{P}_{\alpha' T}^t(\mathbf{x})] \cdot \mathbf{A}_T(\mathbf{x}). \quad (60)$$

Importantly, the interaction component of L_α^t given by the final line of Eq. (58) includes $\dot{\mathbf{R}}(t)$ -dependent terms coming from both $\dot{\mathbf{P}}_{\alpha T}^t$ and $\mathbf{J}^t$, which are proportional to α and α -independent, respectively. It follows that the Coulomb gauge, $\alpha = 0$, is not generally irrotational.

The correct Hamiltonian for the moving atom, H_α^t , can now be found using L_α^t as in Eq. (8). We provide an explicit expression only for the simple example of an electric dipole, which most clearly serves to illustrate the significance of our treatment. A strong electric dipole approximation (EDA), also known as the long-wavelengths approximation, is defined as [46,55]

$$\rho^t(\mathbf{x}) = -\mathbf{d} \cdot \nabla \delta(\mathbf{x} - \mathbf{R}(t)), \quad (61)$$

$$\mathbf{J}^t(\mathbf{x}) = \dot{\mathbf{d}} \delta(\mathbf{x} - \mathbf{R}(t)) - \dot{\mathbf{R}}(t)(\mathbf{d} \cdot \nabla) \delta(\mathbf{x} - \mathbf{R}(t)), \quad (62)$$

$$\mathbf{P}_{\alpha T}^t(\mathbf{x}) = \mathbf{d} \cdot \delta^T(\mathbf{x} - \mathbf{R}(t)), \quad (63)$$

where $\mathbf{d} = -e\mathbf{r}$. The $\dot{\mathbf{R}}(t)$ -dependent but α -independent term in Eq. (62) for $\mathbf{J}^t$ is especially noteworthy. It is the EDA of the so-called *Röntgen* current associated with the gross translational motion of the atom [55–61]. This accounts for the magnetization possessed by a moving electrically polarized system when observed in the laboratory frame, even though the same system appears unmagnetized to a comoving

observer [56]. Without the Röntgen current, the theory is fundamentally inconsistent, because local conservation of charge, Eq. (40), fails to hold.

The construction of an unambiguous canonical theory now proceeds in an identical way to the example in Sec. III A. The canonical momenta conjugate to $\mathbf{r}$ and $\mathbf{A}_T$ are denoted $\mathbf{p}$ and $\mathbf{\Pi}$, respectively. The Hamiltonian found from L'_α via Eq. (8) can be written as

$$H'_\alpha = H_\alpha(t) + X_\alpha(t). \quad (64)$$

Here, $H_\alpha(t)$ is obtained from the replacement $\mathbf{R} \rightarrow \mathbf{R}(t)$ within the corresponding time-independent Hamiltonian H_α as

$$H_\alpha(t) = \frac{1}{2m}[\mathbf{p} + e(1 - \alpha)\mathbf{A}_T(\mathbf{R}(t))]^2 + V(\mathbf{r}) + \frac{1}{2} \int d^3x [(\mathbf{\Pi} + \mathbf{P}'_{\alpha T})^2 + (\nabla \times \mathbf{A}_T)^2], \quad (65)$$

while $X_\alpha(t)$ is an additional term defined by

$$X_\alpha(t) := -\dot{\mathbf{R}}(t) \cdot [(\mathbf{d} \cdot \nabla_{\mathbf{R}})\mathbf{A}_T(\mathbf{R}(t))] + \alpha(\dot{\mathbf{R}}(t) \cdot \nabla_{\mathbf{R}})\mathbf{d} \cdot \mathbf{A}_T(\mathbf{R}(t)). \quad (66)$$

In writing Eq. (64), we have again dropped the purely external kinetic energy. In the Schrödinger picture, we immediately have that

$$X_\alpha(t) - X_{\alpha'}(t) = \partial_t F_{\alpha\alpha'}^t, \quad (67)$$

where $F_{\alpha\alpha'}^t = (\alpha - \alpha')\mathbf{d} \cdot \mathbf{A}_T(\mathbf{R}(t))$ is the EDA of $F_{\alpha\alpha'}^t$ in Eq. (60), such that

$$\begin{aligned} \partial_t F_{\alpha\alpha'}^t &= (\alpha - \alpha')(\dot{\mathbf{R}}(t) \cdot \nabla_{\mathbf{R}})\mathbf{d} \cdot \mathbf{A}_T(\mathbf{R}(t)) \\ &= -i\dot{R}_{\alpha\alpha'}(t)R_{\alpha\alpha'}(t)^\dagger \end{aligned} \quad (68)$$

in which $R_{\alpha\alpha'}(t) := \exp(iF_{\alpha\alpha'}^t)$. Since $H_{\alpha'}(t) = R_{\alpha\alpha'}(t)H_\alpha(t)R_{\alpha\alpha'}(t)^\dagger$, it follows from Eq. (64) that the Hamiltonians H'_α and $H'_{\alpha'}$ are related by

$$H'_{\alpha'} = R_{\alpha\alpha'}(t)H'_\alpha R_{\alpha\alpha'}(t)^\dagger + i\dot{R}_{\alpha\alpha'}(t)R_{\alpha\alpha'}(t)^\dagger, \quad (69)$$

as in Eq. (14).

In the dipole gauge ($\alpha = 1$), the term $X_\alpha(t)$ defines the well-known dipolar Röntgen interaction

$$X_1(t) = -\mathbf{d} \cdot [\dot{\mathbf{R}}(t) \times \mathbf{B}(\mathbf{R}(t))], \quad (70)$$

where $\mathbf{B} = \nabla \times \mathbf{A}_T$. In words, the dipole couples to an additional electric field generated by its gross motion within the laboratory-frame magnetic field.

The irrotational gauge is defined by the condition $X_\alpha(t) \equiv 0$ for which the existence of a solution, α_{irr} , clearly cannot be guaranteed. One also sees clearly why the Coulomb gauge ($\alpha = 0$) is not generally irrotational; the first term in Eq. (66) is nonvanishing for all α . It results from the essential second term in Eq. (62), which is the EDA of the Röntgen current. In certain situations, α_{irr} does exist, and an example is given in Ref. [39] for the case of a dipole moving through a single Gaussian cavity mode. This is discussed further in what follows.

IV. IMPLICATIONS AND DISCUSSION

A. Modulated light-matter interactions

We now explain how our results relate to those within recent literature on the topic of external time dependence in light-matter systems [39–42]. We begin by noting that Eq. (52) is independent of $\mathbf{r}$ and dependent on both α and $\mathbf{R}(t)$, but for $\alpha = 0$, it is independent of $\mathbf{R}(t)$. It has been argued that for describing time-dependent light-matter interactions, modulation $\mu(t)$ should be introduced in the Coulomb gauge via the transverse potential as $\mathbf{A}_T \rightarrow \mathbf{A}'_T = \mu(t)\mathbf{A}_T$, and only then can a gauge-fixing transformation be performed. Under this assumption, the time-independent Hamiltonian H_α should be replaced by

$$H_0^\alpha(t) = H_\alpha(t) + \tilde{X}_\alpha(t), \quad (71)$$

where $H_\alpha(t)$ is obtained by modulating the light-matter coupling within H_α through $\mu(t)$, and

$$\tilde{X}_\alpha(t) := \dot{\mu}(t) \int d^3x \mathbf{P}_{\alpha T}(\mathbf{x}) \cdot \mathbf{A}_T(\mathbf{x}), \quad (72)$$

where $\mathbf{P}_{\alpha T}$ is defined by the time-independent case ($\mathbf{R}(t) \equiv \mathbf{R}$) of Eq. (46). The Hamiltonian in Eq. (71) can be written as $H_0^\alpha(t) = R_{0\alpha}(t)H_0(t)R_{0\alpha}(t)^\dagger + i\dot{R}_{0\alpha}(t)R_{0\alpha}(t)^\dagger$, where

$$R_{\alpha\alpha'}(t) := \exp \left[i\mu(t) \int d^3x [\mathbf{P}_{\alpha T}(\mathbf{x}) - \mathbf{P}_{\alpha' T}(\mathbf{x})] \cdot \mathbf{A}_T(\mathbf{x}) \right]. \quad (73)$$

The set $\{H_0^\alpha(t)\}$ is seen to be the Coulomb gauge equivalence class $\mathcal{S}_0$ defined in the manner described in Sec. II B 2 via Eq. (18). The authors of Refs. [40,41] conclude that introducing $\mu(t)$ does not result in a different physical theory for each different α , because only the class $\mathcal{S}_0$ is physically valid. Upon examining the physical properties of the canonical subsystems in different gauges, it becomes clear that this conclusion cannot be correct in general.

Since $\tilde{X}_0 \equiv 0$ [cf. Eq. (20)], prescription (71) amounts to the *assumption* that the irrotational gauge is always the Coulomb gauge. Physically, Eq. (71) constitutes the assumption that the electric field $\mathbf{E}' = -\dot{\mathbf{A}}'_T - \nabla\phi_{\text{Coul}}$ becomes explicitly time dependent only through the transverse part $\mathbf{E}'_T = -\dot{\mathbf{A}}'_T$, which equals the field momentum in the Coulomb gauge. Therein, however, the intra-atomic Coulomb energy does not exhaust the contribution of the Coulomb field, $\mathbf{E}_L = -\nabla\phi_{\text{Coul}}$, which according to Eq. (47) responds instantaneously at $\mathbf{x}$ to changes in ρ at any other point $\mathbf{x}'$ [62]. Both $\mathbf{A}_T$ and $-\dot{\mathbf{A}}_T = \mathbf{E}_T = \mathbf{E} - \mathbf{E}_L$ possess instantaneous nonlocal static contributions external to the atom. In contrast, modulation in the multipolar gauge occurs entirely inside the atom through a *local* field $\mathbf{E} + \mathbf{P}_1$. Clearly then, the gauge within which modulation is implemented implicitly determines the assumptions that are being made about the spacetime *localization* properties of the external control. A more detailed discussion of these points can be found in Ref. [38].

B. Beyond phenomenological modulation

When considering more fundamental descriptions of the external control, prescription (71) is immediately seen to fail because the gauge-fixing constraint will not generally be the

only explicitly time-dependent one. For example, a time-dependent external flux allows modulation of resonator-qubit coupling [9,10], but external flux generally defines a separate constraint, such as Eq. (21), from which it follows that the irrotational gauge is not fixed.

The shuttling of material systems is also one of the few methods available to modulate a light-matter interaction, but in this case the correct theory will very obviously include at least one other time-dependent constraint, namely, Eq. (41). Indeed, without Eq. (41) no time dependence can occur at all, because Eq. (41) defines $\mathbf{R}(t)$, which then results in time dependence of the gauge-fixing constraint. In particular, Eq. (52) is necessarily accompanied by the $\mathbf{R}(t)$ -dependent Eqs. (50) and (51) yielding an α -independent term that when combined with any α -dependent terms gives $X_\alpha(t)$. This term reduces to $\tilde{X}_\alpha(t)$ only if the contribution arising from the $\dot{\mathbf{R}}(t)$ -dependent Röntgen current in Eq. (62) is removed from it by hand.

In short, the assumption that the Coulomb gauge is necessarily irrotational is equivalent to the incorrect physical assumption that there is necessarily no Röntgen current. This yields $\tilde{X}_\alpha(t)$ in place of the correct result $X_\alpha(t)$. The Röntgen-current contribution, $X_\alpha(t) - \tilde{X}_\alpha(t)$, that is missing in $H_0^\alpha(t) \in \mathcal{S}_0$ is manifestly gauge invariant and measurable. Within a correct theory, it is present in every gauge, including the Coulomb gauge, and this does not depend on any approximations such as the EDA. A theory without the Röntgen current is fundamentally inconsistent [it violates Eq. (40)] and it will fail to predict the correct physical behavior of even the most elementary processes [55,57–61].

The trivial possibility of constructing an equivalence class $\mathcal{S}_\alpha$ from each $H_\alpha(t)$ was noted in Ref. [39]. The *ab initio* selection of one fixed class, such as $\mathcal{S}_0$, as being preferred, would be arbitrary and as such unjustified. This is why the construction of a first-principles description was deemed necessary in Ref. [39]. Equating its result, H_α^i , with the naive model $H_\alpha(t)$ removes the arbitrariness by determining the correct model $H_{\alpha_{\text{irr}}}(t)$ whenever α_{irr} exists, or else revealing that none of the classes $\mathcal{S}_\alpha$ are strictly valid. It was shown in Ref. [39] using the correct $X_\alpha(t)$ in Eq. (66) that in some cases α_{irr} does indeed exist. Specifically, when considering a harmonic dipole aligned with a Gaussian cavity mode's polarization, which makes an angle θ with the constant motion $\mathbf{R}(t)$ through the cavity (orthogonal to the cavity mode wave vector), one obtains $X_\alpha(t) = -e\dot{\mu}(t)\mathbf{r} \cdot \mathbf{A}_T(\mathbf{0})[\alpha - \cos^2\theta]$, where $\mu(t) = \varphi(\mathbf{R}(t))$ and φ is the Gaussian mode function. Prescription (71) meanwhile yields the incorrect result $\tilde{X}_\alpha(t) = -e\alpha\dot{\mu}(t)\mathbf{r} \cdot \mathbf{A}_T(\mathbf{0})$. As expected, the irrotational gauge $\alpha_{\text{irr}} = \cos^2\theta$ is not a constant and vanishes only when $\theta = \pi/2$. In other words, which of the classes $\mathcal{S}_\alpha$ is the correct one, $\mathcal{S}_{\alpha_{\text{irr}}}$, depends on the microscopic context.

C. Natural line shape

Another well-known problem that is treatable using a phenomenological modulation $\mu(t)$ but for which prescription (71) fails is that of the spectrum of spontaneous emission (natural line shape). An arbitrary gauge but otherwise standard description is given in the Appendix. A bare excited dipole in the vacuum, $|0, e\rangle$, transitions into lower states with the emission of a photon. The line shape is defined as the total spectral

density of photons in the long-time limit. Since the vector $|0, e\rangle$ and the photon number operator represent a different physical state $\mathcal{I}_\alpha$ and observable $\mathcal{O}_\alpha$ in each different gauge α , each spectrum $S_\alpha(\omega) \sim \langle \mathcal{O}_\alpha \rangle_{\mathcal{I}_\alpha(t=\infty)}$ refers to a different physical (gauge-invariant) property, and each would have to be measured using a different experiment [38].

The irrotational gauge concept provides additional insight into the problem, which as was noted by Milonni *et al.* [63] can be cast in terms of a modulated interaction. Specifically, the initial vector $|0, e\rangle$ uniquely represents an energy eigenstate provided the light-matter interaction is only switched on at the initial time. The simplest assumption of a sudden switch-on $\mu(t) = \theta(t)$ (Heaviside step) within the gauge α yields a model $H_\alpha(t)$ that coincides with the time-independent Hamiltonian H_α for $t > 0$, and therefore yields the same spectrum $S_\alpha(\omega)$ as the time-independent theory. The gauge relativity of $S_\alpha(\omega)$ can now instead be understood by noting that introducing $\mu(t)$ constitutes a different physical assumption in each different gauge α , such that the resulting naive models $H_\alpha(t)$ are not equivalent and describe different experiments [38,39].

Lamb *et al.* [64] (see also Refs. [65–67]) argue that the dipole gauge provides the relevant subsystem definitions, particularly with regard to early spectroscopic experiments [68–70]. Essentially the same argument was originally made by Power and Zienau, who noted that the Coulomb gauge photonic operator $a_\lambda^0(t, \mathbf{k})$ suffers from static contributions characterized by a pole at $\omega = 0$. This infrared divergent behavior is obviously important in the far-field limit [71], and is needed to exactly cancel the nonlocal Fourier components of instantaneous interatomic Coulomb interactions that are explicit within the Coulomb gauge [52,56,62]. The Power-Zienau-Woolley (PZW) transformation R_{01} yields a photonic operator $a_\lambda^1(t, \mathbf{k})$ (dipole gauge) that by design does not possess static admixtures [62]. As a result, in the dipole (and more generally multipolar) gauge, all interactions occur through a local and properly retarded field $\mathbf{\Pi} = -\mathbf{E} - \mathbf{P}_1$ and there are no direct interatomic Coulomb interactions [52,56].

If, when introducing a modulation $\mu(t)$, the naive α -gauge Hamiltonian $H_\alpha(t)$ is found to be correct, which is to say, the fixed gauge α is found to be irrotational, then the correct Hamiltonian of any other gauge α' is $H_{\alpha'}^\alpha(t) = R_{\alpha\alpha'}(t)H_\alpha(t)R_{\alpha\alpha'}^\dagger(t) + i\dot{R}_{\alpha\alpha'}(t)R_{\alpha\alpha'}^\dagger(t)$. The equivalence class $\mathcal{S}_\alpha = \{H_{\alpha'}^\alpha(t)\}$ generated in this way is as in Sec. II B 2. The Hamiltonian $H_{\alpha'}^\alpha(t)$ can be written as

$$H_{\alpha'}^\alpha(t) = H_{\alpha'}(t) + X_{\alpha'}(t, \alpha), \quad (74)$$

where $H_{\alpha'}(t)$ is obtained by modulating the light-matter coupling within $H_{\alpha'}$ through $\mu(t)$, and

$$\begin{aligned} X_{\alpha'}(t, \alpha) &:= i\dot{R}_{\alpha\alpha'}(t)R_{\alpha\alpha'}^\dagger(t)^\dagger \\ &= \dot{\mu}(t) \int d^3x [\mathbf{P}_{\alpha'T}(\mathbf{x}) - \mathbf{P}_{\alpha'T}(\mathbf{x})] \cdot \mathbf{A}_T(\mathbf{x}) \\ &= \tilde{X}_{\alpha'}(t) - \tilde{X}_\alpha(t) \end{aligned} \quad (75)$$

such that $X_\alpha(t, 0) = \tilde{X}_\alpha(t)$. The prescription defined by Eqs. (71) and (72) is obtained by letting $\alpha = 0$ in Eqs. (74) and (75), which constitutes the assumption that the irrotational gauge is always the Coulomb gauge.

If by comparison with experiments so far one takes the spectrum $S_1(\omega)$ as correct, then one concludes that the dipole gauge is irrotational. The assumption that $\mu(t)$ should be introduced in the Coulomb gauge via $\mathbf{A}_T \rightarrow \theta(t)\mathbf{A}_T$ as in Eq. (71), that is, that the Coulomb gauge is irrotational, is then invalidated [63]. Use of any member of the Coulomb gauge equivalence class $\mathcal{S}_0$ defined by Eq. (71) results in the spectrum $S_0(\omega)$. More generally, using any member of the equivalence class $\mathcal{S}_\alpha$ defined by Eq. (74) results in the spectrum $S_\alpha(\omega)$. Details are given in the Appendix.

It is important to note that, conceivably, different experiments could be constructed that grant access to different spectra $S_\alpha(\omega)$, $\alpha \neq 1$. This aspect of the topic is somewhat nuanced and more detailed discussions can be found in, for example, Refs. [38,72,73]. What is clear, however, is that specifying a single gauge as the universally correct one within which to introduce a time-dependent modulation $\mu(t)$ is unsustainable. For describing an experiment that measures the spectrum $S_\alpha(\omega)$, the gauge α is irrotational and the correct equivalence class is $\mathcal{S}_\alpha$. The line shape example demonstrates that the Coulomb gauge light and matter subsystems possess characteristics that would seem to render them particularly poorly suited for representing the physical subsystem preparation, control, and measurement protocols that are realized in actual experiments. When described properly then, simple physical examples show that time-dependent light-matter interactions are indeed gauge relative [39].

V. CONCLUSIONS

We have presented a general framework for the construction of time-dependent Hamiltonian and Lagrangian theories with holonomic constraints. A main aim in doing so has been to fully understand the recently debated subject of introducing time dependence in quantum light-matter systems [38–42]. We have shown that, in general, a unique canonical theory is obtained only if time dependence is present within the constraints from the outset.

We have provided a general definition of the irrotational gauge as that within which a correct theory is also obtained when introducing time dependence later at the Hamiltonian level. The existence of such a gauge cannot be guaranteed, but several important examples certainly can be found. The idea is useful because it provides a generally simpler theory that is nevertheless correct. We have presented diverse examples within a unified framework [39,43], showing, in particular, that the Coulomb and irrotational gauges do not in general coincide. We have explained precisely how our results relate to those in existing literature on external time dependence of light-matter systems [39–42].

Since external control is necessary for next-generation quantum technologies, explicit time dependence is an increasingly prevalent feature of theoretical descriptions. In addition to the atom-cavity and superconducting circuit systems we have considered here, our general framework may apply to various other contexts. For example, in guiding further developments in the quantum theory of time-varying media, for which a simple, consistent Lagrangian formulation has recently been presented [7], but which is yet to be extended to include a description of guest quantum emitters.

DATA AVAILABILITY

No data were created or analyzed in this study.

APPENDIX

We here provide a more detailed description of the natural line shape problem. When adopting a passive view of gauge-fixing transformations, the photon annihilation operators of different gauges are related using

$$a_\lambda^\alpha(t, \mathbf{k}) = R_{0\alpha}^\dagger a_\lambda^0(t, \mathbf{k}) R_{0\alpha} = a_\lambda^0(t, \mathbf{k}) - \frac{i\alpha \mathbf{e}_\lambda(\mathbf{k}) \cdot \mathbf{d}(t)}{\sqrt{2\omega(2\pi)^3}}, \quad (\text{A1})$$

where $\mathbf{e}_\lambda(\mathbf{k})$ is a unit polarization vector orthogonal to $\mathbf{k}$. The line shape can be defined as the total spectral density of photons, $n_\lambda^\alpha(t, \mathbf{k}) := a_\lambda^\alpha(t, \mathbf{k})^\dagger a_\lambda^\alpha(t, \mathbf{k})$, in the long-time limit:

$$S_\alpha(\omega) = \lim_{t \rightarrow \infty} \int d^3k' \sum_\lambda \langle n_\lambda^\alpha(t, \mathbf{k}') \rangle_{e,0} \delta(\omega - \omega') \quad (\text{A2})$$

$$= \frac{\Gamma}{2\pi} \frac{(\omega/\omega_{eg}^3)[(1-\alpha)\omega_{eg} + \alpha\omega]^2}{(\omega - \omega_{eg})^2 + \Gamma^2/4} \quad (\text{A3})$$

$$= \left[\alpha + (1-\alpha) \frac{\omega_{eg}}{\omega} \right]^2 S_1(\omega), \quad (\text{A4})$$

where the integration is over all photon momenta and the summation is over polarizations. For simplicity, we have assumed that e is the first excited level of the dipole and we have denoted the transition frequency to the bare ground level by ω_{eg} . The result coincides with that obtained using the formal theory of radiation damping [38] and it generalizes to an arbitrary gauge α the well-known dipole and Coulomb gauge spectra found by various authors [63,64,74]. Equation (A3) is obtained by making the Markov and rotating-wave (Weisskopf-Wigner) approximations, within which the dipole moment decays exponentially and only the number-conserving emission of a single real photon is permitted.

In any gauge, the photon operator can be partitioned into vacuum and source parts as $a_\lambda^\alpha(t, \mathbf{k}) = a_{\lambda,\text{vac}}^\alpha(t, \mathbf{k}) + a_{\lambda,s}^\alpha(t, \mathbf{k})$, where $a_{\lambda,\text{vac}}^\alpha(t, \mathbf{k}) = a_\lambda^\alpha(0, \mathbf{k})e^{-i\omega t}$, and the vacuum component does not contribute to the α -gauge vacuum average of the normally ordered product appearing in Eq. (A2). Crucially, however, vacuum-source partitions are gauge relative [38]. Equation (A1) implies that

$$a_{\lambda,\text{vac}}^\alpha(t, \mathbf{k}) - a_{\lambda,\text{vac}}^1(t, \mathbf{k}) = i(1-\alpha) \frac{\mathbf{e}_\lambda(\mathbf{k}) \cdot \mathbf{d}(0)}{\sqrt{2\omega(2\pi)^3}} e^{-i\omega t} \\ =: (1-\alpha)\delta_\lambda(t, \mathbf{k}), \quad (\text{A5})$$

and in turn that

$$a_{\lambda,s}^\alpha(t, \mathbf{k}) - a_{\lambda,s}^1(t, \mathbf{k}) = (1-\alpha) \left[\frac{i\mathbf{e}_\lambda(\mathbf{k}) \cdot \mathbf{d}(t)}{\sqrt{2\omega(2\pi)^3}} - \delta_\lambda(t, \mathbf{k}) \right]. \quad (\text{A6})$$

Since in the Weisskopf-Wigner approximation the dipole moment decays exponentially, the $\mathbf{d}(t)$ -dependent term in Eq. (A6) vanishes for $t \rightarrow \infty$. The vacuum difference $\delta_\lambda(\infty, \mathbf{k})$ is nonvanishing, however, so the line shape $S_1(\omega)$ is only obtained for $\alpha \neq 1$ if $\delta_\lambda(t, \mathbf{k})$ is set to zero by

hand [38,63]. It is important to note that $\delta_\lambda(t, \mathbf{k})$ is time dependent and nonvanishing for all times, so it is not merely a difference in *initial* conditions for the $a_\lambda^\alpha(t, \mathbf{k})$. Moreover, since even very early experiments are able to resolve the difference between $S_0(\omega)$ and $S_1(\omega)$, removing $\delta_\lambda^0(t, \mathbf{k})$ from $a_{\lambda,s}^0(t, \mathbf{k})$ cannot be justified as an approximation. Indeed, in Eq. (A5), the initial dipole moment operator $\mathbf{d}(0)$, which is nothing but the Schrödinger picture operator, is necessarily nonvanishing. This is true classically as well; the charges comprising the dipole cannot be taken as identically coincident, despite their separation typically being small compared with resonant transition wavelengths (EDA). Imposing $\delta_\lambda^\alpha(t, \mathbf{k}) = 0$ within Eq. (A6) is simply an *ad hoc* means by which to force the equality $a_{\lambda,s}^\alpha(\infty, \mathbf{k}) = a_{\lambda,s}^1(\infty, \mathbf{k})$, thereby implementing within the gauge α the assumption that the dipole gauge subsystems are those most physically relevant [62,64–67].

The equivalent treatment of the problem that instead assumes a sudden interaction switch-on, $\mu(t) = \theta(t)$ (Heaviside step), reveals that using any member of the Coulomb gauge equivalence class $\mathcal{S}_0$ defined by Eq. (71) results in the incorrect spectrum $S_0(\omega)$. To see this, first note that for $\mu(t) = \theta(t)$, the additional term $\tilde{X}_\alpha(t)$ in Eq. (72) is for a dipole at $\mathbf{0}$ given by $\tilde{X}_\alpha(t) = \alpha\delta(t)\mathbf{d} \cdot \mathbf{A}_T(\mathbf{0})$, where $\dot{\mu}(t) = \delta(t)$. For a smoother switching function $\dot{\mu}(t)$ would be concurrently more smoothly peaked around $t = 0$. The equations of mo-

tion found using the arbitrary member $H_0^\alpha(t)$ of $\mathcal{S}_0$ can be integrated from any initial time $t_0 < 0$ at which μ vanishes, and for the case $\mu(t) = \theta(t)$, one obtains the photonic source operator

$$a_{0,\lambda,s}^\alpha(t, \mathbf{k}) = a_{\lambda,s}^\alpha(t, \mathbf{k}) - \alpha\delta_\lambda(t, \mathbf{k}), \quad (\text{A7})$$

where the second term is due to $\tilde{X}_\alpha(t)$. It is easily shown [75] that $a_{\lambda,s}^\alpha(t, \mathbf{k}) = (1 - \alpha)a_{\lambda,s}^0(t, \mathbf{k}) + \alpha a_{\lambda,s}^1(t, \mathbf{k})$ from which it follows using Eq. (A6) that

$$a_{0,\lambda,s}^\alpha(t, \mathbf{k}) = a_{\lambda,s}^0(t, \mathbf{k}) - i\alpha \frac{\mathbf{e}_\lambda(\mathbf{k}) \cdot \mathbf{d}(t)}{\sqrt{2\omega(2\pi)^3}}. \quad (\text{A8})$$

Exponential decay implies that the $\mathbf{d}(t)$ -dependent second term does not contribute to the spectrum, which is therefore the same incorrect result $S_0(\omega)$ found when using the naive Coulomb gauge Hamiltonian $H_0(t) = H_0^0(t)$ [63]. This calculation is easily extended to show more generally that the photonic source operator found using the EDA of the Hamiltonian $H_\alpha^\alpha(t)$ in Eq. (74) is

$$a_{\alpha,\lambda,s}^\alpha(t, \mathbf{k}) = a_{\lambda,s}^\alpha(t, \mathbf{k}) + i(\alpha - \alpha') \frac{\mathbf{e}_\lambda(\mathbf{k}) \cdot \mathbf{d}(t)}{\sqrt{2\omega(2\pi)^3}}. \quad (\text{A9})$$

Therefore, using any member of the equivalence class $\mathcal{S}_\alpha$ given by Eq. (74) results in the spectrum $S_\alpha(\omega)$.

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