Avoiding gauge ambiguities in cavity QED

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Systems of interacting charges and fields are ubiquitous in physics. Their mathematical description has an inbuilt flexibility known as gauge freedom, or gauge invariance, and all gauges must correspond to the same physical laws. Recently however, it has been shown that Hamiltonians derived using different gauges can yield different physical results when matter degrees of freedom are truncated to a few low-lying energy eigenstates; an effect that is particularly prominent in the ultra-strong coupling regime. This is because regular gauge transformations, those that alter the longitudinal part of the vector potential, can reshuffle the partition between light and matter degrees of freedom. From this it follows that these gauge transformations do not commute with level truncations. To avoid gauge ambiguity, we redefine the electromagnetic fields in terms of potentials for which the resulting canonical momenta and Hamiltonian are explicitly unchanged by the gauge choice of this theory. In our theory, gauge transformations hence commute with the light/matter partitioning, and instead this partition is assigned by the intuitive choice of separating an electric field between displacement and polarisation contributions, which naturally must be done in the context of the physical situation. This approach is an attractive choice in the typical cavity QED situation that has no free currents and where a few-level truncation is useful.

Introduction. — The gauge invariance of quantum electrodynamics (QED) is fundamental to the theory and can be used to greatly simplify calculations [1–7]. Of course, gauge invariance implies that physical observables are the same in all gauges despite superficial differences in the mathematics. However, it has recently been shown that the invariance is lost in the strong light/matter coupling regime if the matter degrees of freedom are treated as quantum systems with a fixed number of energy levels [4, 8–11], including the commonly used two-level truncation (2LT). At the origin of this is the role of gauge transformations (GTs) in deciding the partition between the light and matter degrees of freedom, even if the primary role of gauge freedom is to enforce Gauss’s law. Despite its long history [12], this has led to new questions about which gauge most accurately describes the physics.

Two common gauge choices of non-relativistic QED [8, 9, 11] are the Coulomb gauge, which has the advantage of describing photons as purely transverse radiation modes, and the multipolar gauge, which is most useful when the leading order (dipole) terms are dominant in a multipole expansion of the fields [13]. Interestingly, within the 2LT the multipolar gauge is usually found to agree more closely with exact, gauge invariant calculations than the Coulomb gauge [8, 9]. This has been attributed to the fact that in the Coulomb gauge the light/matter interaction strength scales with the transition frequency between the relevant matter levels, while in the multipolar gauge the coupling instead scales with the energy of the radiation mode. Therefore, transitions between well-separated matter levels can be non-negligible in the Coulomb gauge [8, 9]. Further, Ref. [11] suggested that the 2LT in the Coulomb gauge converts the local potential into a non-local one, which no longer only depends on position but now also on the gauge-dependent canonical momentum. This is further discussed in Refs. [14, 15] for a variety of physical settings.

The implication of these results is that the multipolar gauge is usually more accurate when the matter system is quantized and truncated to two levels. However, it has been emphasised [4, 10] that there exist a continuum of possible GTs, each with a unique light/matter partition and therefore also 2LT; depending on the physical setting, gauges other than the common choices can offer...
more accurate 2LTs [4, 10, 16]. Recently, it has also been reported that time-dependent light/matter couplings can lead to gauge ambiguities [17].

In this Letter, we reformulate QED such that gauge ambiguities do not manifest, see Fig. 1, and separate gauge issues from the choice of light/matter partition, for which we now offer an intuitive interpretation. Our reformulation builds on previous work on the dual representation of QED [18–23]. We shall find completely gauge invariant results, and for systems typical of QED, e.g. dipoles in a cavity, the Hamiltonian we provide has the same form as the multipolar gauge in the conventional approach.

Conventional approach. — We first outline the conventional approach, and how gauge ambiguities arise in it (see Supplemental Information (SI)-1 [24] for detail). Let us consider a generic system of charges \( q_\mu \) at positions \( r_\mu \) described by a charge density \( \rho \) and current density \( J \). Their dynamics are governed by the Maxwell equations and Lorentz force, see SI-1, Eqns. (SI 1).

Conventionally, the electric and magnetic fields are parametrized in terms of vector and scalar potentials \( A \) and \( A_0 \) as \( E = -\nabla A_0 - A \) and \( B = \nabla \times A \) respectively, leading immediately to Faraday’s law and sourceless magnetic fields [25]. The remaining equations are derived by minimizing the action of the \textit{minimal-coupling Lagrangian} [Eq. (SI 3)], from which the general Hamiltonian

\[
H_m' = \sum_\mu \frac{1}{2m_\mu} [P_\mu - q_\mu A(r_\mu) + q_\mu \nabla \chi(r_\mu)]^2
\]

\[
+ \int d^3x \left( \frac{1}{2\varepsilon_0} [\Pi'(x) + \phi'(x)]^2 + \frac{B^2(x)}{2\mu_0} \right)
\]

can also be obtained. Throughout, we denote gauge dependent quantities with a prime and all fields are time dependent. In the above,

\[
P_\mu' = m_\mu \dot{r}_\mu + q_\mu A(r_\mu) - q_\mu \nabla \chi(r_\mu),
\]

\[
\Pi'(x) = -\varepsilon_0 E(x) - \phi'(x),
\]

are the matter and field canonical momenta with \( \phi'(x) = \int d^3x' \rho(x') [\partial \chi(x,x')/\partial A(x)] \) and where \( \chi(x) = \int d^3x' \chi(x,x') \) is the generic scalar field resulting from the gauge freedom [2].

Gauge ambiguities can occur, particularly in the strong light/matter coupling regime, when approximations to the Hamiltonian are introduced. A prominent example of this is expressing the matter Hamiltonian using a truncated number of energy levels; this approximation has different meanings in each gauge. When quantizing, the gauge dependent classical momentum \( \mathbf{p}_\mu' \) is promoted to its quantum counterpart \( \hat{\mathbf{p}}_\mu' \) (along with the position \( r_\mu \to \hat{r}_\mu \)). The truncation to \( N+1 \) discrete energy levels follows next:

\[
\hat{T}'_\mu = \frac{\hat{\mathbf{p}}'^2_\mu}{2m_\mu} + \hat{U}_{\text{ext}}(\hat{r}_\mu) + \sum_{n=0}^N \epsilon_{n,\mu} |e_n,\mu\rangle \langle e'_{n,\mu}|, \tag{3}
\]

for each charge, where \( \hat{U}_{\text{ext}} \) is the external electrostatic interaction binding the charges. Each ‘matter’ eigenstate in (3) refers to a different physical system in each gauge and so truncation means losing different information. Formally, only when \( N \to \infty \) do all observables agree in different gauges, though for weak light/matter coupling a low-level truncation is usually sufficient for good agreement.

New approach. — The canonical momenta in the theory outlined above inherit their gauge-dependency from the minimal-coupling Lagrangian [Eq. (SI 3)], as the vector and scalar potentials are only defined up to the scalar function \( \chi \). To remove gauge ambiguities, we will therefore derive a theory which is described by a Lagrangian depending only on the physical fields.

The total charge and current densities of any system can be partitioned into \textit{bound} and \textit{free} contributions as \( \rho = \rho_b + \rho_f \) and \( J = J_b + J_f \) [25]. This naturally allows one to distinguish two contributions to the electric and magnetic fields: \( E = (D - P)/\varepsilon_0 \) and \( B = \mu_0 (H + M) \) with \( P \) and \( M \) being the polarisation and magnetisation fields. Our aim now is to parametrize the displacement and magnetic fields \( D \) and \( H \) using a dual vector potential \( C \) and scalar potential \( C_0 \) such that

\[
D(x) = \nabla \times C(x), \tag{4a}
\]

\[
H(x) = \nabla C_0(x) + \hat{C}(x). \tag{4b}
\]

This is the crucial point of this Letter, and as we will show, it avoids gauge ambiguities in the formulation of cavity QED. The parametrization in terms of \( C \)-fields relies on the absence of free currents \( J_f \), a common cavity QED setting [4, 8–11]. Other examples of defining the physical fields in this way can be found in [18–23], although here we extend the formulation to include the magnetization field and therefore move beyond the standard electric dipole approximation. The polarization field \( P \) and the magnetisation field \( M \) are sourced by the bound charge and currents, respectively:

\[
\nabla \cdot P(x, r) = -\rho_b(x, r), \tag{5a}
\]

\[
\nabla \times M(x, r) = J_b(x, r) - \hat{P}(x, r), \tag{5b}
\]

and we note that Maxwell’s equations Eqns. (SI 1c) and (SI 1d) become:

\[
\nabla \cdot D(x) = \rho_f(x), \tag{6a}
\]

\[
\nabla \times H(x) = J_f(x) + \hat{D}(x), \tag{6b}
\]

when written in terms of the displacement field \( D \) and magnetic field \( H \).

We now specify a system to illustrate the theory, and for simplicity we will choose a single dipole formed of an electron at position \( r \) and a hole at the origin. The bound charge density and current of this dipole are described by \( \rho_b(x, r) = -e\delta(x - r) + e\delta(x) \) and \( J_b(x, r) = e\delta(x) \).
We find that the Lagrangian is for general fields $V$ and $C$.

Maxwell [Eqns. (SI 1a) and (SI 1b)] and Lorentz force write a Lagrangian that reproduces the remaining by gauge freedom.

We emphasize that, in contrast, for the conventional partitioning. We emphasize that, in contrast, for the conventional -field theory this partitioning is encompassed within the system. There are no free charges or currents $(\rho f = J_f = 0)$ and so a symmetry emerges when comparing Eqns. (6) to Maxwell’s equations Eqns. (SI 1a) and (SI 1b) [13, 18–23, 25]. We will exploit this symmetry to parametrize the displacement and magnetic fields according to Eqns. (4).

The restrictions on $\mathbf{P}$ and $\mathbf{M}$ given by Eqns. (5) produce the correct bound charge density and current if $\mathbf{P}(x, r) = -\varepsilon_0 \int d\lambda \, \mathbf{r} \delta(\mathbf{x} - \lambda \mathbf{r})$, and $\mathbf{M}(x, r) = -\mathbf{\dot{r}} \times \mathbf{\theta}(x, r)$ where $\mathbf{\theta}(x, r) = -\varepsilon_0 \int d\lambda \, \lambda \mathbf{r} \delta(\mathbf{x} - \lambda \mathbf{r})$ [2, 4, 17]. These are however not unique, and Eqns. (5) are also satisfied by $\mathbf{P} \rightarrow \tilde{\mathbf{P}} = \mathbf{P} + \nabla \mathbf{V}_0(x, r)$, and $\mathbf{M} \rightarrow \tilde{\mathbf{M}} = \mathbf{M} - \nabla \mathbf{V}_0(x, r)$, for general fields $\mathbf{V}$ and $\mathbf{V}_0$ and quantities dependent on these are denoted with a tilde. Such a transformation does not change the physics, but alters the light/matter partitioning. We emphasize that, in contrast, for the conventional -field theory this partitioning is encompassed by gauge freedom.

All that remains to complete the theory is to write a Lagrangian that reproduces the remaining Maxwell [Eqns. (SI 1a) and (SI 1b)], and Lorentz force [Eqn. (SI 1e)] equations, when minimised with respect to the mechanical degrees of freedom $C_0$, $C$ and $\mathbf{r}$ respectively [26]. We find that the required Lagrangian is

$$L = \frac{1}{2} m \mathbf{r}^2 + \int d^3 x \frac{\varepsilon_0}{2} \left[ \mathbf{E}^2(x) - \mathbf{B}^2(x) \right],$$

which for $\mathbf{M} \rightarrow 0$ agrees with Refs. [23, 27] and we prove in SI-2 [24] that this Lagrangian satisfies all the necessary equations of motion.

Eqns. (4) are invariant under gauge transformations $C_0 \rightarrow C_0 + \xi$ and $C \rightarrow C - \nabla \xi$ as they must be, but importantly so is the Lagrangian in Eq. (8). This is because the Lagrangian is written only in terms of the physical fields. Additionally, this means that the Lagrangian is invariant under the transformations in Eqns. (7). For $\mathbf{B}$ and $\mathbf{E}$ to be invariant under this transformation, there must be an implicit change to the fields $C$ and $C_0$, which we write explicitly as $\mathbf{D} \rightarrow \tilde{\mathbf{D}}$ and $\mathbf{H} \rightarrow \tilde{\mathbf{H}}$ where

$$\tilde{\mathbf{D}}(x) = \nabla \times \tilde{\mathbf{C}}(x) = \mathbf{E}(x) + \tilde{\mathbf{P}}(x, r),$$

$$\tilde{\mathbf{H}}(x) = \tilde{\mathbf{C}}(x) + \nabla \tilde{C}_0(x) = \mathbf{B}(x) - \tilde{\mathbf{M}}(x, r).$$

This is a direct consequence of the transformation in Eqns. (7) changing the light/matter partition; a redistribution of the contributions of $\mathbf{D}^\perp$ and $\mathbf{P}^\perp$ to $\mathbf{E}^\perp$, and likewise $\mathbf{H}$ and $\mathbf{M}$ to $\mathbf{B}$. This leads to the crucial result that the canonical momenta in the new theory are no longer gauge dependent, although they do depend on the light/matter partition. We find that the canonical momenta are

$$\tilde{\mathbf{p}} = \frac{\partial L}{\partial \dot{\mathbf{r}}} = m \dot{\mathbf{r}} - \Phi_0(\mathbf{r}) - \tilde{\Phi}(\mathbf{r}),$$

$$\tilde{\Pi}(x) = \frac{\delta L}{\delta \dot{\mathbf{C}}(x)} = \mathbf{B}(x),$$

where $\Phi_0(\mathbf{r}) = \int d^3 x \, \mathbf{B}(x, r) \times \mathbf{M}_V(x, r)$. We derive Eqns. (11) in SI-3 [24]. We see that the field canonical momentum is always the magnetic field whilst the matter canonical momentum is dependent on the light/matter partition. There is a subtlety that arises in that we must be able to invert Eqn. (11a) to write $\mathbf{r}$ as a function of $\mathbf{p}$. This puts a constraint on the allowed $\mathbf{V}$ and $V_0$ fields in the transformations in Eqns. (7) which we discuss in SI-3 [24]. Assuming this constraint is met, we find that the Hamiltonian is

$$\tilde{H} = \frac{1}{2m} \left[ \mathbf{p} + \Phi_0(\mathbf{r}) + \tilde{\Phi}(\mathbf{r}) \right]^2 + \tilde{U}_\text{ext}$$

$$+ \int d^3 x \, \left( \frac{\mathbf{B}^2(x)}{\varepsilon_0} + \frac{1}{\varepsilon_0} \left[ \mathbf{D}^\perp(x) - \tilde{\mathbf{P}}(x) \right]^2 \right),$$

where we have introduced an external potential $\tilde{U}_\text{ext}$. Eqn. (13) is derived explicitly in SI-3 [24]. The gauge independence of Eq. (13) follows from the absence of magnetic monopoles, and as such the primary constraint $\nabla \cdot \mathbf{B} = 0$ can be satisfied without altering the light/matter partition [28].

Before quantising the fields, we must choose a light/matter partition. The benefit of the new theory is that the light/matter partition now has the physical motivation of defining $\mathbf{P}^\perp$ and $\mathbf{M}$, as opposed to the more esoteric gauge field. In this example, $\mathbf{P}$ should be chosen to wholly contain the dipole, as we are treating a small dipole and the polarisation field is thus a well-defined quantity. Therefore we choose $\mathbf{V} = V_0 = 0$ resulting in $\tilde{\Phi} = 0$. After making this choice, we can now remove the tildes on the fields. We must then also choose the gauge. Here we pick the Coulomb-gauge analogue of $\nabla \cdot C = 0$ and $C_0 = 0$, but we note that the gauge does not affect the light/matter partition nor the form of the Hamiltonian. We quantise the fields by enforcing

$$[\tilde{C}^\perp_{ij}(x), \tilde{\Pi}_{ij}(x')] = i \delta_{ij}(x-x'),$$

where

$$\tilde{C}^\perp(x) = \sum_{k\lambda} \epsilon_{k\lambda} f_k \left( i \tilde{a}^\dagger_{k\lambda} e^{-ik\cdot x} + \tilde{a}_{k\lambda} e^{ik\cdot x} \right),$$

and $f_k = (2\nu_k V)^{-1/2}$ is the coupling strength to the mode with frequency $\nu_k = k |v|$ in volume $V$, $\epsilon_{k\lambda}$ are polarisation vectors orthonormal to $\mathbf{k}$, and $a_{k\lambda}$ is the
plotted on the diagonal where \( g^\hat{V} \) for different choices of photon annihilation (creation) operator. We note that in the plot, with markers indicating individual experiments given in SI-5 [24].

\[ \eta \text{ ultra-strong coupling.} \]

We also show approximate regions where different types of QED experiments sit with respect to be written as of the Hamiltonian are also quantized and expanded into quantization process is given in more detail in SI-2 [24].

After a suitable definition of polarisation vectors, the \( \mathbf{C} \) quantization process is given in more detail in SI-2 [24].

In Figure 2 we display the ac-

\[ \mathbf{m} \text{ magnetisation interactions governed through} \]

\[ \Phi \text{ (for this example equivalent to the} \]

\[ \mathbf{C} \text{ matter system is possible.} \]

\[ (\text{EDA}) \]

At Eq. (15), we make the electric dipole approximation (EDA) \( (\mathbf{k} \cdot \mathbf{r} \ll 1) \) which allows us to evaluate the fields at the origin, set \( \mathbf{P} \sim -\mathbf{e} \mathbf{r} \delta(\mathbf{x}) \) and ignore the smaller magnetisation interactions governed through \( \mathbf{\Phi}_0 \). The quantization process is given in more detail in SI-2 [24].

After a suitable definition of polarisation vectors, the \( \mathbf{C} \) field Hamiltonian in Eqn. (15) has the same mathematical form as the multipolar gauge Hamiltonian for the \( \mathbf{A} \) field, which is a reflection of the light/matter partitions being identical.

We now turn to the question of whether a 2LT for the matter system is possible. In Figure 2 we display the accuracy of the \( \mathbf{C} \)-field Hamiltonian in an arbitrary gauge (for this example equivalent to the \( \mathbf{A} \)-field in the multipolar gauge) and the conventional \( \mathbf{A} \)-field in the Coulomb gauge when truncated to two or three dipole levels. See SI-1 [24] for details on these Hamiltonians. Here, we use only a single radiation mode that is resonant with the transition between the two lowest dipole levels, and \( \tilde{U}_{\text{ext}} \) is an infinite square well potential whose anharmonicity makes it amenable to few-level expansion. In the strong coupling limit, \( \tilde{g} \rightarrow 1 \), both truncated Hamiltonians become inaccurate, importantly for different reasons: All physical assumptions are well justified for the \( \mathbf{C} \)-fields, and the main source of error stems from the few-level truncation, so accuracy is much improved by going from two to three levels. In contrast, the Coulomb gauge should be inaccurate for small dipoles, as any small change in the dipole position results in a proportionally large change in momentum, which in the Coulomb gauge leads to a large light/matter coupling. Indeed, we only see limited improvement by going from two to three levels for the Coulomb gauge. This is further discussed in SI-6 [24]. Here we should also note that in both cases we must keep counter-rotating terms, as they contribute significantly in the strong-coupling regime [29].

\[ \text{Discussion.} \]

We should stress that we are free to work in any \( \mathbf{C} \)-field gauge without affecting the light/matter partitioning. The gauge should also be chosen to reflect the physical situation: for instance if the system centre-of-mass is moving a Lorenz gauge is appropriate, whereas a Coulomb gauge is a good choice for
static systems. The latter may be useful also if boundaries between different regions are considered, which is not the case for A-field Coulomb gauge, where a generalisation is required to make the problem tractable [30]. Since this choice aligns with the multipolar gauge in the A-field representation, we agree with the conclusion of Refs. [8, 9, 11] that this A-field gauge choice most accurately represents the physics of small, bound dipoles.

In conclusion, we find that for systems without free currents and where a truncation of the matter system to few levels is desirable – typical of cavity QED situations – the C-field representation is an attractive choice: it completely removes the dependence of physical predictions after a level truncation on the choice of gauge. Instead this choice is moved to the more intuitive consideration of how to find a description which appropriately partitions between matter and radiation, i.e. into the choice of $\mathbf{P}^\perp$ and $\mathbf{M}$. In any case, the accuracy of results obtained in the (matter-truncated) C-representation are independent of the choice of gauge and limited only by the validity of the few-level truncation.

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SUPPLEMENTARY NOTE 1: THE COULOMB AND THE MULTIPOLAR GAUGES FOR THE SIMPLE DIPOLE SYSTEM

For the generic charge distribution discussed when summarising the conventional A-field approach to QED, the Maxwell and Lorentz equations are as usual

\[ \nabla \cdot \mathbf{B}(x) = 0, \quad \text{(SI 1a)} \]
\[ \nabla \times \mathbf{E}(x) = -\dot{\mathbf{B}}(x), \quad \text{(SI 1b)} \]
\[ \nabla \cdot \mathbf{E}(x) = \rho(x)/\varepsilon_0, \quad \text{(SI 1c)} \]
\[ \nabla \times \mathbf{B}(x) = \mu_0 \mathbf{J}(x) + \varepsilon_0 \mu_0 \ddot{\mathbf{E}}(x), \quad \text{(SI 1d)} \]
\[ m_\mu \ddot{r}_\mu = q_\mu [\mathbf{E}(r_\mu) + \dot{r}_\mu \times \mathbf{B}(r_\mu)] \quad \text{(SI 1e)} \]

Any Lagrangian which can reproduce Maxwell’s equations and the Lorentz force [Eqns. (SI 1)] from the Euler-Lagrange equations is suitable for describing this system in an electromagnetic (EM) field. Maxwell’s equations Eqns. (SI 1a) and (SI 1b) can be satisfied by defining a vector potential \( \mathbf{A} \) and scalar potential \( A_0 \) such that

\[ \mathbf{E}(x) = -\nabla A_0(x) - \dot{\mathbf{A}}(x), \quad \text{(SI 2a)} \]
\[ \mathbf{B}(x) = \nabla \times \mathbf{A}(x), \quad \text{(SI 2b)} \]

The remaining Maxwell’s equations, Eqns. (SI 1c) and (SI 1d), along with the equations of motion for the charges Eq. (SI 1e), can be obtained from the \textit{minimal-coupling Lagrangian} \cite{2–6}

\[ L_m = \sum_\mu \frac{1}{2} m_\mu \ddot{r}_\mu^2 + \int d^3x \ L_m(x), \quad \text{(SI 3)} \]

with the Lagrange density

\[ L_m(x) = \frac{\varepsilon_0}{2} [\mathbf{E}^2(x) - e^2 \mathbf{B}^2(x)] + [\mathbf{J}(x) \cdot \mathbf{A}(x) - \rho(x) A_0(x)], \quad \text{(SI 4)} \]

where the mechanical degrees of freedom are \( A_0, \mathbf{A} \) and \( r_\mu \), respectively. The physical fields in Eq. (SI 2) are unchanged by the introduction of a scalar field

\[ \chi(x) = \int d^3x' \chi(x, x'), \quad \text{(SI 5)} \]

so long as

\[ A'_0(x) = A_0(x) + \dot{\chi}(x), \quad \text{(SI 6a)} \]
\[ \mathbf{A}'(x) = \mathbf{A}(x) - \nabla \chi(x), \quad \text{(SI 6b)} \]

where a primed variable indicates one transformed by the \( \chi \)-field. Under this transformation the Lagrangian \( L'_m \) has a modified Lagrange density given by

\[ L'_m(x) = L_m(x) - \mathbf{J}(x) \cdot \nabla \chi(x) - \rho(x) \int d^3x' \dot{\mathbf{A}}(x') \cdot \frac{\partial \chi(x, x')}{\partial \mathbf{A}(x')} \quad \text{(SI 7)} \]

The canonical momenta of this arbitrary-gauge Lagrangian, \( p'_\mu = \partial L'_m/\partial \dot{r}_\mu \) and \( \Pi' = \delta L'_m/\delta \dot{\mathbf{A}} \), can be found as \cite{2}

\[ p'_\mu = m_\mu \dot{r}_\mu + q_\mu \mathbf{A}(r_\mu) - q_\mu \nabla \chi(r_\mu), \quad \text{(SI 8a)} \]
\[ \Pi'(x) = -\varepsilon_0 \mathbf{E}(x) - \phi'(x) \quad \text{(SI 8b)} \]

where

\[ \phi'(x) = \int d^3x' \rho(x') \frac{\partial \chi(x, x')}{\partial \mathbf{A}(x)} \quad \text{(SI 9)} \]

Importantly, \( p'_\mu \) and \( \Pi' \) are explicitly gauge dependent and so correspond to different canonical momenta in every gauge \cite{2, 4}. This can lead to gauge ambiguities when the matter parts of the Hamiltonian are quantised and expanded.
into a finite number of energy eigenstates. After eliminating $A_0$ using the continuity equation, the arbitrary-gauge Hamiltonian is found as [2]

$$H' = \sum_\mu \frac{1}{2m_\mu} \left[ p_\mu - q_\mu A_\mu + q_\mu \nabla \chi (r_\mu) \right]^2 + \int d^3x \left( \frac{1}{2\varepsilon_0} [\Pi (x) + \phi (x)]^2 + \frac{B^2(x)}{2\mu_0} \right). \quad (SI 10)$$

Depending on the gauge choice, the photon field $A$ has different physical meanings. Only in Coulomb gauge, where $\nabla \chi = A^\parallel$, is the photon field purely transverse, with $A_C = A^\perp$ and $\Pi_C = -\varepsilon_0 E^\perp$ where the ‘prime’ is replaced with subscript ‘C’ to indicate the gauge. This also means that the Coulomb gauge canonical momentum is

$$p_{C,\mu} = m_\mu \dot{r}_\mu - q_\mu A^{\perp} (r_\mu). \quad (SI 11)$$

In other gauges, for example the multipolar gauge, the radiation field contains longitudinal, dipolar degrees of freedom. We will discuss these in detail later in this section.

Despite superficial differences, the theory is still gauge invariant and so, irrespective of the choice of $\chi$-field, $H'_{\text{eq}}$ will describe the same physics. From Eq. (SI 6b) it is clear that the transverse part of the vector potential is gauge invariant. This is quantised in the usual way by satisfying the commutation relation $[\hat{A}^\perp_i (x), \hat{\Pi}^\perp_j (x')] = i\delta^\perp_{ij} (x - x')$, where $\delta^\perp_{ij} (x - x')$ is the transverse $\delta$-function, yielding

$$\hat{\mathbf{A}}^\perp (x) = \sum_{k\lambda} \epsilon_{k\lambda} f_k \left( \hat{a}_{k\lambda} e^{-i \mathbf{k} \cdot \mathbf{x}} + \hat{a}^\dagger_{k\lambda} e^{i \mathbf{k} \cdot \mathbf{x}} \right), \quad (SI 12)$$

where $f_k = (2\pi \hbar^2 V)^{-1/2}$ is the coupling strength to the mode with frequency $\nu_k = c |\mathbf{k}|$ in volume $V$, $\epsilon_{k\lambda}$ are polarisation unit vectors orthonormal to $\mathbf{k}$, with $\hat{a}_{k\lambda}$ ($\hat{a}^\dagger_{k\lambda}$) being a gauge-dependent photon annihilation (creation) operator [4–6, 31]. In the following we will henceforth derive equations for the simple dipole system in the main text instead of the generic charge distribution.

**Coulomb gauge**

The Coulomb gauge Hamiltonian is defined by choosing $\nabla \chi = A^\parallel$ and results in $\phi_C = -\varepsilon_0 E^\parallel$ and so $\Pi_C = -\varepsilon_0 E^\perp$ where we have replaced superscript ‘prime’ with subscript ‘C’ to denote the gauge [2]. On substituting these into the arbitrary-gauge Hamiltonian Eq. (2) for the simple system described in the main text, one finds that

$$H_C = \frac{1}{2m} \left[ p_C + e A^{\perp} (r) \right]^2 + U_{\text{ext}} + U_b + \frac{1}{2} \int d^3x \left( \frac{1}{\varepsilon_0} |\Pi^\perp_C (x)|^2 + \frac{1}{\mu_0} |\mathbf{B} (x)|^2 \right), \quad (SI 13)$$

where $U_{\text{ext}}$ is an externally applied potential and $U_b = \frac{e^2}{2} \int d^3x |E^{\parallel} (x)|^2$ is the electron-hole electrostatic potential which contains the divergent self-energies and interaction between the particles. This potential follows from Gauss’s law Eq. (SI 1c) and the lack of free charge, which leads to

$$E^\parallel (x) = -\nabla \int d^3x' \frac{\rho_h (x')}{4\pi \varepsilon_0 |x - x'|} = -\frac{1}{\varepsilon_0} \nabla V_b (x), \quad (SI 14)$$

where $V_b = V_e + V_h$ is the sum of electron and hole potentials. Integration by parts, assuming fields vanish at infinities, and again using Gauss’s law leads to

$$U_b = \frac{1}{2\varepsilon_0} \int d^3x \rho_h (x) V_b (x) \equiv U_{e-h} + U_{e-e} + U_{h-h}, \quad (SI 15)$$

where $U_{e-e}$ and $U_{h-h}$ are the divergent self energies of the electron and hole and $U_{e-h}$ is the electrostatic energy due to the mutual attraction. The $A^{\perp}$-field is quantised as in Eqn. (SI 12) and from this the magnetic field follows as

$$\hat{\mathbf{B}} (x) = \nabla \times \hat{\mathbf{A}} (x) = -i \sum_{k\lambda} (\mathbf{k} \times \epsilon_{k\lambda}) f_k \left( \hat{a}^\dagger_{k\lambda} e^{-i \mathbf{k} \cdot \mathbf{x}} - \hat{a}_{k\lambda} e^{i \mathbf{k} \cdot \mathbf{x}} \right). \quad (SI 16)$$

The canonical momentum of the field is gauge dependent, but has the same expression in each gauge found through the commutator $[\hat{A}^\perp_i (x), \hat{\Pi}^\perp_j (x')] = i\delta^\perp_{ij} (x - x')$, where $\delta^\perp_{ij} (x - x')$, and is given by

$$\hat{\Pi}^\perp (x) = i \sum_{k\lambda} \nu_k \epsilon_{k\lambda} f_k \left( \hat{a}^\dagger_{k\lambda} e^{-i \mathbf{k} \cdot \mathbf{x}} - \hat{a}_{k\lambda} e^{i \mathbf{k} \cdot \mathbf{x}} \right). \quad (SI 17)$$
Using these quantised fields, the terms inside the integral in Eq. (SI 13) becomes the usual bosonic field energy term and so the Coulomb-gauge Hamiltonian becomes

\[ H_C = \frac{1}{2m} \left[ \hat{p}_C + e \hat{A}^\perp (\mathbf{r}) \right]^2 + \hat{U}_{\text{ext}} + \hat{U}_b + \sum_{k\lambda} \nu_k \left( a^\dagger_{k\lambda} a_{k\lambda} + \frac{1}{2} \right). \]  

(SI 18)

In Coulomb gauge, the light/matter coupling is between the dipole canonical momentum and the gauge invariant transverse vector potential, of the form \( \hat{p}_C \cdot \hat{A}^\perp \). From Eq. (SI 6b), we see that the field canonical momentum is proportional to the transverse electric field, \( \Pi_C = -\varepsilon_0 \mathbf{E}^\perp \). We now expand in the basis of \( N + 1 \) energy eigenstates. The system Hamiltonian is formed from

\[ \frac{\hat{p}^2}{2m} + \hat{U}_{\text{ext}} \to \sum_{n=0}^N \varepsilon_n |\epsilon^+_n \rangle \langle \epsilon^+_n | \equiv H_0'. \]  

(SI 19)

Finally, the Coulomb-gauge light matter interaction is written in this basis as follows

\[ \frac{e}{m} \hat{p} \cdot \hat{A}^\perp = ie [H_{0, C}, \hat{r}] \cdot \hat{A}^\perp \]

\[ = -\sum_{n=0}^N \sum_{n' \geq n} \varepsilon_{nn'} \hat{d}_{nn'} \cdot \hat{A}^\perp \hat{\sigma}^y_{nn'}, \]  

(SI 20)

where \( \varepsilon_{nn'} \equiv \varepsilon_{n' - \varepsilon_n}, \hat{d}_{nn'} = -e \langle n | \hat{r} | n' \rangle \) and \( \hat{\sigma}^y_{nn'} = -i \langle n' | \langle n | - | n \rangle \langle n' | \rangle \). In the second line of Eq. (SI 20) we have written \( \hat{p} = m \hat{v}_r \), which is only true in Coulomb gauge in the absence of a vector potential, and is why the unperturbed Hamiltonian is used in the expectation value \( \hat{r} = \hat{v}_0, \mathbf{r} \). Finally, the Coulomb-gauge Hamiltonian written in the energy eigenbasis is

\[ H_C = \sum_{n=0}^N \varepsilon_n |\epsilon^+_n \rangle \langle \epsilon^+_n | - \sum_{n=0}^N \sum_{n' > n} \varepsilon_{nn'} \hat{d}_{nn'} \cdot \hat{A}^\perp \hat{\sigma}^y_{nn'} + \frac{1}{2m} \left[ e \hat{A}^\perp \right]^2 + \sum_{k\lambda} \nu_k \hat{a}^\dagger_{k\lambda} \hat{a}_{k\lambda}, \]  

(SI 21)

where the ladder operators are implicitly in Coulomb gauge. We have made the electric dipole approximation which allows us to evaluate the vector potential at the centre of the dipole at the origin at the coordinate system, i.e. \( \mathbf{A}^\perp \equiv \hat{A}^\perp (0) \). Additionally, we have ignored the vacuum energy of the photon field, and the divergent self energies of the electron and hole, \( \hat{U}_{e-h} \), and also the electron-hole interaction \( \hat{U}_{e-h} \), which is itself divergent in the electric dipole approximation.

**Multipolar gauge**

The derivation of the multipolar gauge Hamiltonian is more involved and the reader is pointed to [2, 7] for the details. It can be summarised by the choice \( \nabla \chi = \mathbf{A}^\perp - \mathbf{P}/e \). This leads to \( \mathbf{P}_m = \mathbf{P} \) and therefore \( \Pi_m = -\mathbf{D} = -\mathbf{D}^\perp \) where the lack of longitudinal component follows from Eq. (6a) because there are no free charges. By substitution into Eq. (2), one finds that the Hamiltonian in the multipolar gauge is

\[ H_m = \frac{1}{2m} [\mathbf{p}_m + \Phi(x)]^2 + U_{\text{ext}} + \frac{1}{2} \int d^3x \left( \frac{1}{\varepsilon_0} \left[ \Pi_m^\perp (\mathbf{x}) + \mathbf{P} (\mathbf{x}, \mathbf{r}) \right]^2 + \frac{1}{\mu_0} \mathbf{B}^2 (\mathbf{x}) \right). \]  

(SI 22)

The EM fields are quantised in the same way as in Coulomb-gauge, despite \( \Pi_m^\perp \) and \( \Pi_C^\perp \) corresponding to different physical fields. In the multipolar gauge, the electrostatic interaction is governed by the longitudinal component of the polarisation field \( \hat{U}_b = \frac{1}{2\varepsilon_0} \int d^3x |\mathbf{P}(x)|^2 \), which is clear from Eq. (5a) and leads to the same expression given in Eq. (SI 15). In the multipolar gauge, the photon field is much more complicated, \( \Pi_m = -\mathbf{D}^\perp = -\mathbf{E}^\perp - \mathbf{P}^\perp \), and contains both dipole and transverse field degrees of freedom. The light/matter coupling is between the mechanical dipole position and the canonical momentum of the field, \( \int d^3x \mathbf{P}^\perp (\mathbf{x}) \cdot \mathbf{D}^\perp (\mathbf{x}) \). Now expanding the matter into the energy basis, the interaction becomes

\[ \int d^3x \mathbf{P}^\perp (\mathbf{x}, \mathbf{r}) \cdot \mathbf{\Pi}_m^\perp (\mathbf{x}) = -e \int d^3x \int_0^1 d\lambda \hat{r} \cdot \mathbf{\Pi}_m^\perp (\mathbf{x}) \delta (\mathbf{x} - \lambda \mathbf{r}) \]

\[ = -e \hat{r} \cdot \int_0^1 d\lambda \mathbf{\Pi}_m^\perp (\lambda \mathbf{r}) \]

\[ \approx \hat{d} \cdot \mathbf{\Pi}_m^\perp (0), \]  

(SI 23)
where we used the dipole operator $\hat{d} = -e \hat{r}$ and in the last line we have made the electric dipole approximation. Within the EDA, the dipolar self-energy term can be rewritten using the transverse delta function as [4]

$$\frac{1}{2} \int d^3x \left[ \hat{P}^\perp (x, r) \right]^2 \approx \sum_k f^2_k \nu_k \left( \hat{d} \cdot \epsilon_{k}\lambda \right)^2.$$  

(Appendix 24)

Applying these, expanding the kinetic energy terms of the eigenbasis as in Eqn. (SI 19) and ignoring the magnetic contribution in $\Phi$, we find the standard multipolar Hamiltonian

$$H_m = \sum_{n=0}^{N} \epsilon_n |\epsilon_{n,M}\rangle \langle \epsilon_{n,M}| - \frac{1}{\varepsilon_0} \hat{d} \cdot \hat{D}^\perp (0) + \frac{1}{\varepsilon_0} \sum_k f^2_k \nu_k \left( \hat{d} \cdot \epsilon_{k}\lambda \right)^2 + \sum_{k\lambda} \nu_k \hat{a}_{k\lambda} \hat{a}_{k\lambda}^\dagger,$$  

(Appendix 25)

where the ladder operators are implicitly in multipolar gauge. The similarities between the multipolar Hamiltonian derived here in the $A$-field approach and the gauge-invariant Hamiltonian we have derived in the new $C$-field approach in Eqn. (15) should be noted (discussed further in the main text). Furthermore, the expansion of the dipole operator into the $N + 1$ energy eigenstates is

$$\hat{d} = \sum_{n=0}^{N} \sum_{n' > n} d_{nn'} \hat{a}_{nn'}^\dagger,$$  

(Appendix 26)

where we have assumed that the binding potential gives rise to eigenstates for which the diagonal elements of the dipole transition matrix are zero and $d_{nn'} = -e \langle n | \hat{r} | n' \rangle$. Note that the coupling strength between levels $n$ and $n'$ scales like $d_{nn'} \nu$ in the multipolar gauge and $d_{nn'} \epsilon_{nn'}$ in Coulomb gauge. The increasing coupling strength with energy level separation in Coulomb gauge is related to the breakdown of gauge invariance for finite dipole level truncation, as reported by Refs. [8, 9].
SUPPLEMENTARY NOTE 2: DERIVING THE EQUATIONS OF MOTION AND HAMILTONIAN FROM THE V = V₀ = 0 THEORY

In this section, we will derive the equations of motion and the Hamiltonian of the C-field theory from the Lagrangian given in the main text by Eqn. (8). For ease of reading, this Lagrangian is

\[ L = \frac{1}{2} m \dot{r}^2 + \int d^3 x \left( \frac{1}{2} \left( \mu_0 |H(x) + M(x, r)|^2 - \frac{1}{\varepsilon_0} |D(x) - P(x, r)|^2 \right) \right), \]  

(SI 27)

where \( D = \nabla \times C \) and \( H = \nabla C₀ + \dot{C} \), which automatically satisfy Gauss’s law \( \nabla \cdot D = 0 \) and Maxwell-Ampere’s law \( \nabla \times H = \dot{D} \). Note that the equations of motion for the Lagrangian transformed by Eqns. (7) must be identical because the transformation preserves the equations of motion by construction.

No magnetic monopoles

The non-existence of magnetic monopoles means that \( B \) is sourceless and hence divergenceless: \( \nabla \cdot B = 0 \). This equation of motion is found from the Euler-Lagrange equation

\[ \frac{d}{dt} \frac{\partial L}{\partial \dot{C}_0(x)} - \frac{\partial L}{\partial C_0(x)} = 0. \]  

(SI 28)

Since \( L \) does not depend on \( \dot{C}_0 \) we can immediately write that \( \partial L / \partial \dot{C}_0 = 0 \). We can also show that

\[ \frac{\partial L}{\partial C_0(x)} = \frac{\partial}{\partial C_0(x)} \int d^3 x' \mu_0 \left[ \frac{1}{2} H^2(x') + H(x') \cdot M(x', r) \right] = \int d^3 x' \mu_0 \left[ H(x') + M(x', r) \right] \cdot \frac{\partial}{\partial C_0(x)} \nabla C_0(x') = - \int d^3 x' \left[ \nabla \cdot B(x') \right] \frac{\partial C_0(x')}{\partial C_0(x)} = - \nabla \cdot B(x), \]

where, to arrive at the third line, we have integrated by parts and, to arrive at the final line, we have used that \( \partial C_0(x') / \partial C_0(x) = \delta(x - x') \). This completes the proof. Note that treating \( C_0 \) as an independent coordinate results in a singular Hessian and so a non-invertible Lagrangian. It is better to not treat \( C_0 \) as a coordinate, and instead impose \( \nabla \cdot B = 0 \) as a primary constraint on the Lagrangian. This is identical to Gauss’s law \( \nabla \cdot E = \rho \) and \( A₀ \) in the conventional theory.

Faraday’s equation

Faraday’s equation, \( \nabla \times E = -\dot{B} \), is found from the Euler-Lagrange equation

\[ \frac{d}{dt} \frac{\partial L}{\partial C(x)} - \frac{\partial L}{\partial C(x)} = 0. \]  

(SI 30)

We start with the derivative with respect to \( C \), for which we find that

\[ \frac{\partial L}{\partial C(x)} = \frac{\partial}{\partial C(x)} \int d^3 x' \mu_0 \left[ \frac{1}{2} H^2(x') + H(x') \cdot M(x', r) \right] = \int d^3 x' B(x') \cdot \frac{\partial C(x')}{\partial C(x)} = B(x), \]

(SI 31)

and note that this is also the conjugate momentum of the field. The other derivative is

\[ \frac{\partial L}{\partial C(x)} = \frac{\partial}{\partial C(x)} \int d^3 x' \frac{1}{\varepsilon_0} \left[ -\frac{1}{2} D^2(x') + D(x') \cdot P(x', r) \right]. \]  

(SI 32)
This is slightly more complicated so we will take each term individually. The $D^2$ term is evaluated using $D = \nabla \times C$ and the identity $\partial / \partial \mathbf{V} (\nabla \times \mathbf{V})^2 = 2 \nabla \times \nabla \times \mathbf{V}$ for any vector field $\mathbf{V}$. The $D \cdot P$ term is evaluated as

$$\frac{\partial}{\partial C(x)} \int d^3x' D(x') \cdot P(x', r) =$$

$$\int d^3x' \left[ \left( P(x', r) \cdot \frac{\partial}{\partial C(x)} \right) D(x') + P(x', r) \times \left( \frac{\partial}{\partial C(x')} \times D(x') \right) \right]. \quad (SI \ 33)$$

For the first term of Eqn. (SI 33) we find that

$$\left( P(x', r) \cdot \frac{\partial}{\partial C(x)} \right) D(x') = P_i(x', r) \frac{\partial}{\partial C_i(x)} D(x')$$

$$= P_i(x', r) \nabla \times \frac{\partial C(x')}{\partial C_i(x)}$$

$$= P_i(x', r) \nabla \times \left[ \hat{r}_i \delta(x - x') \right]$$

$$= \nabla \times \left[ P(x', r) \delta(x - x') \right], \quad (SI \ 34)$$

where $\hat{r}_i$ is the unit vector along $i^{th}$ cartesian direction. The second term in Eqn. (SI 33) is zero because $\partial / \partial C \times (\nabla \times C) = \nabla \times [\partial / \partial C \times C] = 0$. Therefore, we find that

$$\frac{\partial L}{\partial C(x)} = -\nabla \times \int d^3x' \frac{1}{\varepsilon_0} [D(x') - P(x', r)] \delta(x - x') = -\nabla \times E(x). \quad (SI \ 35)$$

Together, Eqns. (SI 31) and (SI 35) complete the proof.

**Lorentz force**

The proceeding derivation of the Lorentz force is very lengthy. It can be bypassed by noting that our Hamiltonian in Eqn. (13) is almost identical to the multipolar Hamiltonian in [2] when written in terms of the physical fields. Since the multipolar gauge Hamiltonian is equivalent to the Coulomb gauge Hamiltonian, and it is readily proven that the Coulomb gauge Hamiltonian reproduces the Lorentz force through Heisenberg’s equation, so must the C-field Hamiltonian. This argument can only be made for the Lorentz force derivation because this does not depend on the choice of potentials to characterize the physical fields. Nevertheless, for concreteness here we will derive the Lorentz force explicitly from the C-field Lagrangian. For simplicity, we will choose here use $\mathbf{V} = 0$ and $V_0 = 0$.

The Lorentz force of the electron [32], $m \mathbf{\ddot{r}} = -e [\mathbf{E}(r) + \hat{r} \times \mathbf{B}(r)]$, is found from the Euler-Lagrange equation

$$\frac{d}{dt} \frac{\partial L}{\partial \dot{r}} - \frac{\partial L}{\partial r} = 0. \quad (SI \ 36)$$

First consider the velocity derivative,

$$\frac{\partial L}{\partial \dot{r}} = m \ddot{r} + \frac{\partial}{\partial \dot{r}} \int d^3x \mu_0 \left[ \mathbf{H}(x) \cdot \mathbf{M}(x, r) + \frac{1}{2} \mathbf{M}^2(x, r) \right]. \quad (SI \ 37)$$

It can be shown that

$$\frac{\partial}{\partial \dot{r}} \mathbf{H}(x) \cdot \mathbf{M}(x, r) = -\theta(x, r) \times \mathbf{H}(x), \quad (SI \ 38)$$

$$\frac{\partial}{\partial \dot{r}} \frac{1}{2} \mathbf{M}^2(x, r) = -\theta(x, r) \times \mathbf{M}(x, r). \quad (SI \ 39)$$

Therefore,

$$\frac{\partial L}{\partial \dot{r}} = m \ddot{r} - \int d^3x \theta(x, r) \times \mathbf{B}(x)$$

$$= m \ddot{r} + e \int_0^1 d\lambda \lambda \mathbf{r} \times \mathbf{B}(\lambda \mathbf{r}), \quad (SI \ 40)$$
which is also equal to the conjugate momentum $p$ in Eq. (11a) if $V = V_0 = 0$. We then take the total time derivative of this to find
\[
\frac{d}{dt} \frac{\partial L}{\partial \dot{r}} = m \ddot{r} + e \int_0^1 d\lambda \lambda \left( \dot{r} \times B(\lambda r) + r \times \dot{B}(\lambda r) + r \times [(\dot{r} \cdot \nabla_r) B(\lambda r)] \right). \tag{SI 41}
\]

The position derivative separates into electric and magnetic parts
\[
\frac{\partial L}{\partial r} = \int d^3x \nabla_r (L_{\text{elec}} + L_{\text{mag}}), \tag{SI 42}
\]
where
\[
\nabla_r L_{\text{elec}} = \varepsilon^{-1} \nabla_r \left( D(x) \cdot P(x, r) - \frac{1}{2} P^2(x, r) \right)
= [E(x) \cdot \nabla_r] P(x, r) + E(x) \times [\nabla_r \times P(x, r)]
= \nabla_p [E(x) \cdot P(x, r)], \tag{SI 43}
\]
and the terms involving partial derivatives of $D$ with respect to $r$ are zero. To arrive at the last line we have used the identity $a \times (\nabla_r \times b) = \nabla_b (a \cdot b) - (a \cdot \nabla_r) b$ where the gradient operator with a tilde is the Feynman subscript notation, i.e.
\[
\nabla_b (a \cdot b) = \left( a \cdot \frac{\partial b}{\partial r_x}, a \cdot \frac{\partial b}{\partial r_y}, a \cdot \frac{\partial b}{\partial r_z} \right). \tag{SI 44}
\]

Likewise the magnetic part is
\[
\nabla_r L_{\text{mag}} = \nabla_r \left( H(x) \cdot M(x, r) + \frac{1}{2} M^2(x, r) \right)
= [B(x) \cdot \nabla_r] M(x, r) + B(x) \times [\nabla_r \times M(x, r)]
= \nabla_M [B(x) \cdot M(x, r)], \tag{SI 45}
\]
and partial derivatives of $H$ with respect to $r$ are zero. The electric part evaluates to
\[
\int d^3x \nabla_r L_{\text{elec}} = \int d^3x \nabla_p [E(x) \cdot P(x, r)]
= -e \int_0^1 d\lambda \int d^3x \ [E(x) + (E(x) \cdot r) \nabla_r] \delta(x - \lambda r). \tag{SI 46}
\]
and the magnetic part is
\[
\int d^3x \nabla_r L_{\text{mag}} = \int d^3x \nabla_M [B(x) \cdot M(x, r)]
= -e \int_0^1 d\lambda \int d^3x \ \{ \dot{r} \times B(x) + (r \cdot [\dot{r} \times B(x)]) \nabla_r \} \delta(x - \lambda r). \tag{SI 47}
\]

Before substituting into the Euler-Lagrange equation we note that by using Faraday’s law we can rewrite the second term in Eqn. (SI 41) as
\[
e \int_0^1 d\lambda \lambda \dot{r} \times \dot{B}(\lambda r) = -e \int_0^1 d\lambda \ r \times [\nabla_r \times E(\lambda r)]
= -e \int_0^1 d\lambda \ (\nabla_r \times E(\lambda r) - [r \cdot \nabla_r] E(\lambda r))
= -e \int_0^1 d\lambda \ \int d^3x \ \{ [r \cdot E(x)] \nabla_r - E(x) \cdot [r \cdot \nabla_r] \} \delta(x - \lambda r), \tag{SI 48}
\]
where to arrive at the last line we have written \( E(\lambda r) = \int d^3x \, E(x) \delta(x - \lambda r) \). Substituting Eqns. (SI 41), (SI 46), (SI 47) and (SI 48) into the Euler-Lagrange equation results in

\[
m\ddot{r} = -e \int_0^1 d\lambda \int d^3x \, E(x) \left[ 1 + (r \cdot \nabla_r) \right] \delta(x - \lambda r)
- e \int_0^1 d\lambda \int d^3x \, \lambda \left\{ [r \times B(x)] \left( \dot{r} \cdot \nabla_x \right) + 2\dot{r} \times B(x) + (r \cdot [\dot{r} \times B(x)]) \nabla_x \right\} \delta(x - \lambda r),
\]

where we have also written \( B(\lambda r) = \int d^3x \, B(x) \delta(x - \lambda r) \). After some algebra we can then prove that

\[
[r \times B(x)] \left( \dot{r} \cdot \nabla_x \right) = \dot{r} \times [B(x) \nabla_x] = (r \cdot \nabla_x) [\dot{r} \times B(x)] + (r \times \dot{r}) [B(x) \cdot \nabla],
\]

which when substituted into Eqn. (SI 49) along with \( \nabla_x \delta(x - \lambda r) = -\lambda \nabla_x \delta(x - \lambda r) \) we find

\[
m\ddot{r} = -e \int d^3x \, E(x) \int_0^1 d\lambda \left[ 1 - \lambda (r \cdot \nabla_x) \right] \delta(x - \lambda r)
- e \int d^3x \, \left[ \dot{r} \times B(x) \right] \int_0^1 d\lambda \lambda (2 - \lambda r \cdot \nabla_x) \delta(x - \lambda r)
- \int_0^1 d\lambda \lambda \dot{r} \times \ddot{r} \int d^3x \, [B(x) \cdot \nabla_x] \delta(x - \lambda r).
\]

From here the Lorentz force is derived by noting that because of the non-existence of magnetic monopoles

\[
\int d^3x \, [B(x) \cdot \nabla_x] \delta(x - \lambda r) = -\int d^3x \, [\nabla_x \cdot B(x)] \delta(x - \lambda r) = 0,
\]

and that the following identities hold

\[
\int_0^1 d\lambda \, (1 - \lambda r \cdot \nabla_x) \delta(x - \lambda r) = \delta(x - r),
\]
\[
\int_0^1 d\lambda \, \lambda (2 - \lambda r \cdot \nabla_x) \delta(x - \lambda r) = \delta(x - r).
\]

These identities can be proven using the Fourier representation of the delta function [2]. Using these we find that \( m\ddot{r} = -e [E(r) + \dot{r} \times B(r)] \) as required.

### Hamiltonian

The Hamiltonian is derived as is standard by

\[
H = \sum_i p_i \cdot q_i - L,
\]

where \( q_i \) and \( p_i \) are the generalised coordinates and canonical momenta. For this theory, the canonical momenta of the matter and \( C \)-field are given in Eqn. (11). We find that

\[
H = p \cdot \dot{r} - \frac{1}{2} m \dot{r}^2 + \int d^3x \left( B(x) \cdot \dot{C}(x) - \frac{\mu_0}{2} B^2(x) + \frac{\varepsilon_0}{2} E^2(x) \right).
\]

Focusing first on the terms within the integral, we can rewrite \( B \cdot \dot{C} = B \cdot H - B \cdot \nabla C_0 \). The term \( B \cdot \nabla C_0 \) then vanishes after integration by parts and enforcing \( \nabla \cdot B = 0 \). which follow from \( \nabla \cdot B = 0 \). Since the canonical field momentum is \( B \), we want to eliminate \( H \) in favour of \( B \) to make the quantisation process easier. Therefore, writing \( H + M = B/\mu_0 \) we arrive at

\[
H = p \cdot \dot{r} - \frac{1}{2} m \dot{r}^2 + \frac{1}{2} \int d^3x \left( \varepsilon_0 E^2(x) + \frac{1}{\mu_0} B^2(x) - 2B(x) \cdot M(x, r) \right).
\]
Using the definitions of $\theta$ and $\Phi_0$ from the main text we are then able to show that
\[
\int d^3x \, \mathbf{B}(x) \cdot \mathbf{M}(x, r) = -\frac{1}{m} \left[ \mathbf{p} \cdot \Phi_0(r) + \Phi_0^2(r) \right].
\] (SI 58)

Finally, after combining this with the terms outside the integral using $m\hat{r} = \mathbf{p} + \Phi_0$, we have
\[
H = \frac{1}{2m} [\mathbf{p} + \Phi_0(r)]^2 + \int d^3x \left( \frac{\mathbf{B}^2(x)}{2\mu_0} + \frac{1}{2\varepsilon_0} \left[ \mathbf{D}^\perp(x) - \mathbf{P}(x, r) \right]^2 \right),
\] (SI 59)
as in Eqn. (13) of the main text. Note that here we have explicitly used that the longitudinal component of $\mathbf{D}$ is zero because there is no free charge. To align with the Hamiltonians in SI-1 for the conventional theory we will ignore the magnetic interactions governed by $\Phi_0$. Quantising the radiation fields is completely analogous as in the conventional theory described in SI-1; we express the transverse vector potential as Eqn. (14). The displacement field, analogous to the magnetic field in the conventional theory, follows from this
\[
\hat{\mathbf{D}}(x) = \nabla \times \hat{\mathbf{C}}(x) = -i \sum_{k\lambda} (\mathbf{k} \times \epsilon_{k\lambda}) f_k \left( \hat{a}^\dagger_{k\lambda} e^{-ik \cdot x} - \hat{a}_{k\lambda} e^{ik \cdot x} \right).
\] (SI 60)

Using the canonical commutation relation $[\hat{\mathbf{C}}_i(x), \hat{\mathbf{C}}_j(x')] = [\hat{\mathbf{C}}_j(x), \hat{\mathbf{C}}_j^\dagger(x')] = i\delta_{ij}(x - x')$, where the second equality follows from $\nabla \cdot \mathbf{B} = 0$, implies that
\[
\hat{\mathbf{B}}(x) = i \sum_{k\lambda} \nu_k \epsilon_{k\lambda} f_k \left( \hat{a}^\dagger_{k\lambda} e^{-ik \cdot x} - \hat{a}_{k\lambda} e^{ik \cdot x} \right).
\] (SI 61)

Identically to multipolar gauge of the conventional theory (see SI-1), the longitudinal component of the polarisation field leads to the electron and hole self energies and interaction energy. Making the electric dipole approximation, ignoring these divergent energies and quantising the matter part of the Hamiltonian by introducing an external potential $U_{\text{ext}}$ (see SI-1 for the details) results in
\[
\hat{\mathcal{H}} = \sum_{n=0}^N \epsilon_n \langle \epsilon_n \rangle \Phi_n^2 \sum_{k\lambda} \nu_k \epsilon_{k\lambda} \hat{a}^\dagger_{k\lambda} \hat{a}_{k\lambda} - \frac{1}{\varepsilon_0} \hat{\mathbf{d}} \cdot \left[ \nabla \times \hat{\mathbf{C}}^\dagger(0) \right] + \frac{1}{\varepsilon_0} \sum_{k\lambda} f_k^2 \nu_k \left( \hat{\mathbf{d}} \cdot \epsilon_{k\lambda} \right)^2,
\] (SI 62)
which is Eqn. 15 in the main text.

**Comparison between C-field and A-field**

| A-field approach | C-field approach |
|------------------|------------------|
| $\mathbf{B} = \nabla \times \mathbf{A}$ | $\mathbf{H} = \nabla \Phi_0 + \mathbf{C}$ |
| $\mathbf{E} = -\nabla A_0 - \hat{\mathbf{A}}$ | $\mathbf{D} = \nabla \times \mathbf{C}$ |
| $\mathbf{A}' = \begin{cases} \mathbf{A}^\perp C \end{cases}$ | $\mathbf{C}' = \begin{cases} \mathbf{C}^\perp \Phi_0 \Phi & \text{"C"} \end{cases}$ |
| $\Pi' = \begin{cases} -\varepsilon_0 \mathbf{E}^\perp C \\
\mathbf{D} \end{cases}$ | $\Pi = \mathbf{B}$ |
| $\mathbf{p}' = \begin{cases} m \hat{r} + e \mathbf{A}^\perp C \\
\hat{r} - \mathbf{p} \end{cases}$ | $\tilde{\mathbf{p}} = m \hat{r} - \Phi_0 - \Phi$ |

**TABLE I.** Comparison of $\mathbf{A}$-field and $\mathbf{C}$-field representations. Gauge dependent parameters are denoted with a prime. $C/m$ ("C/m") denotes the Coulomb/multipolar gauge (-analogues) in the $\mathbf{A}$-field ($\mathbf{C}$-field) representation respectively, however the choice of gauge is inconsequential for predictions of the $\mathbf{C}$-theory. We define $\Phi_D(r) = \int d^3x \, \theta(x, r) \times \mathbf{D}(x)$. 

SUPPLEMENTARY NOTE 3: DERIVING THE CANONICAL MOMENTA AND HAMILTONIAN WITH NONZERO $V$ AND $V_0$

Field and matter canonical momenta

After transformation by Equations (7) the Lagrangian is

$$\tilde{L} = \frac{1}{2} m \tilde{\dot{x}}^2 + \int d^3 x \, \tilde{\mathcal{L}}(x),$$

where

$$\tilde{\mathcal{L}}(x) = \frac{1}{2} \left( \frac{1}{\mu_0} \mathbf{B}^2(x) - \varepsilon_0 \mathbf{E}^2(x) \right)$$

$$= \frac{1}{2} \left( \mu_0 \left[ \tilde{\mathbf{H}}(x) + \tilde{\mathbf{M}}(x, r) \right]^2 - \frac{1}{\varepsilon_0} \left[ \tilde{\mathbf{D}}(x) - \tilde{\mathbf{P}}(x, r) \right]^2 \right).$$

Also, recall the definitions $\tilde{\mathbf{P}} = \mathbf{P} + \tilde{\mathbf{P}}_V$, $\tilde{\mathbf{M}} = \mathbf{M} - \tilde{\mathbf{M}}_V$

$$\tilde{\mathbf{P}}_V (x, r) = \nabla \times \mathbf{V} (x, r),$$

$$\tilde{\mathbf{M}}_V (x, r) = \tilde{\mathbf{V}} (x, r) + \nabla V_0 (x, r),$$

$$\tilde{\mathbf{D}}(x) = \nabla \times \tilde{\mathbf{C}}(x),$$

$$\tilde{\mathbf{H}}(x) = \tilde{\mathbf{C}}(x) + \nabla \tilde{\mathbf{C}}_0(x),$$

where we have written the implicit change in $\mathbf{C}$ ($C_0$) due to the change in light/matter partition explicitly as $\mathbf{C} \rightarrow \tilde{\mathbf{C}}$ ($C_0 \rightarrow \tilde{\mathbf{C}}_0$). It is clear that the field canonical momentum will not change from the $V = V_0 = 0$ case, because the field $\mathbf{C}$ only changes implicitly. Therefore we trivially find that

$$\tilde{\mathbf{H}}(x) = \frac{\delta \tilde{\mathcal{L}}}{\delta \tilde{\mathbf{C}}(x)} = \mu_0 \left[ \tilde{\mathbf{H}}(x) + \tilde{\mathbf{M}}(x, r) \right] = \mathbf{B}(x).$$

For the matter canonical momentum we must evaluate

$$\tilde{\mathbf{p}} = m \tilde{\dot{x}} + \mu_0 \nabla_x \int d^3 x \left( \tilde{\mathbf{H}}(x) \cdot \tilde{\mathbf{M}}(x, r) + \frac{1}{2} \tilde{\mathbf{M}}^2(x, r) \right).$$

To do so we recall some algebraic relations used in the $V = V_0 = 0$ case:

$$\nabla_x [\mathbf{F}(x) \cdot \mathbf{M}(x, r)] = -\theta (x, r) \cdot \mathbf{F}(x) \forall \mathbf{F}(x),$$

$$\nabla_x \frac{1}{2} \mathbf{M}^2(x, r) = -\theta (x, r) \cdot \mathbf{M}(x, r).$$

We can expand the integral as

$$\tilde{\mathbf{p}} = m \tilde{\dot{x}} + \mu_0 \nabla_x \int d^3 x \left( \frac{1}{2} \tilde{\mathbf{M}}^2(x) - \tilde{\mathbf{H}}(x) \cdot \mathbf{M}(x, r) \right)$$

$$+ \mu_0 \nabla_x \int d^3 x \left( \frac{1}{2} \tilde{\mathbf{M}}^2_V(x, r) - \left[ \mathbf{M}(x, r) + \tilde{\mathbf{H}}(x) \right] \cdot \tilde{\mathbf{M}}_V(x, r) \right),$$

where $\mathbf{M}_V \equiv \tilde{\mathbf{V}} + \nabla V_0$. Using Eqns. (SI 72) we can show that

$$\mu_0 \nabla_x \int d^3 x \left( \frac{1}{2} \tilde{\mathbf{M}}^2_V(x, r) - \left[ \mathbf{M}(x, r) + \tilde{\mathbf{H}}(x) \right] \cdot \tilde{\mathbf{M}}_V(x, r) \right) = -\mu_0 \int d^3 x \, \theta(r) \times \left[ \mathbf{M}(x, r) + \tilde{\mathbf{H}}(x) \right].$$

Then, using standard vector calculus identities we find that the second integral can be written as

$$\mu_0 \nabla_x \int d^3 x \left( \frac{1}{2} \tilde{\mathbf{M}}^2_V(x, r) - \left[ \mathbf{M}(x, r) + \tilde{\mathbf{H}}(x) \right] \cdot \tilde{\mathbf{M}}_V(x, r) \right)$$

$$= \int d^3 x \left( \mu_0 \left( \left[ \tilde{\mathbf{M}}_V \cdot \nabla_x \right] \mathbf{M} + \tilde{\mathbf{M}}_V \times [\nabla_x \times \mathbf{M}] \right) - \left\{ \left[ \mathbf{B} \cdot \nabla_x \right] \tilde{\mathbf{M}}_V + \mathbf{B} \times [\nabla_x \times \tilde{\mathbf{M}}_V] \right\} \right)$$

$$= \int d^3 x \mu_0 \left[ \theta(x, r) \times \tilde{\mathbf{M}}_V(x, r) \right] - \nabla_x \int d^3 x \, \mathbf{B}(x) \cdot \tilde{\mathbf{M}}_V(x, r).$$
Combining these, we find that

\[
\tilde{p} = m\tilde{r} - \int d^3x \, \theta(r) \times B(x) - \nabla_{\tilde{r}} \int d^3x \, B(x) \cdot \vec{M}_V(x, r)
\]

\[
\equiv m\tilde{r} - \Phi_0(r) - \Phi(r)
\]  

(SI 79)

as in Eqn. (11a) in the main text.

**Hamiltonian**

As usual we write the Hamiltonian in terms of the coordinates and momenta as in Eqn. (SI 55)

\[
\tilde{H} = \tilde{p} \cdot \tilde{r} - \frac{1}{2} m\tilde{r}^2 + \int d^3x \, \left[ B(x) \cdot \tilde{C}(x) - \frac{1}{2\mu_0} B^2(x) + \frac{\varepsilon_0}{2} E^2(x) \right].
\]

(SI 81)

We then define \( \tilde{X} = \Phi_0 + \tilde{\Phi} \) and note the following three relations:

- Using the definition \( \tilde{C} + \nabla \tilde{C} = \mu_0^{-1} B - \vec{M} \) and that \( \nabla \cdot B = 0 \) leads to

\[
\int d^3x \, B(x) \cdot \tilde{C}(x) = \int d^3x \, \left( \frac{1}{\mu_0} B^2(x) - B(x) \cdot \vec{M}(x, r) \right).
\]

(SI 82)

- The terms outside the volume integral can be written as

\[
\tilde{p} \cdot \tilde{r} - \frac{1}{2} m\tilde{r}^2 = \frac{1}{2m} \left( \tilde{p}^2 - \tilde{X}^2(r) \right).
\]

(SI 83)

- We can rewrite

\[
- \int d^3x \, B(x) \cdot M(x, r) = \frac{1}{m} \left( \tilde{p} \cdot \tilde{X}(r) + \tilde{X}^2(r) \right) - \tilde{r} \cdot \tilde{\Phi}(r).
\]

(SI 84)

Inserting these three relations into Eqn. (SI 81) leads to

\[
\tilde{H} = \frac{1}{2m} \left( \tilde{p} + \tilde{X}(r) \right)^2 + \frac{1}{2} \int d^3x \, \left[ \frac{1}{\mu_0} B^2(x) + \varepsilon_0 E^2(x) \right] + (1 - \tilde{r} \cdot \nabla_{\tilde{r}}) \int d^3x \, B(x) \cdot \vec{M}_V(x, r).
\]

(SI 85)

The third term in Eqn. (SI 85) is a result of not being able to invert the matter canonical momentum for an expression for \( \tilde{r} \) in terms of \( \tilde{p} \). We therefore impose a constraint on the choice of \( V \) and \( V_0 \) such that:

\[
(1 - \tilde{r} \cdot \nabla_{\tilde{r}}) \int d^3x \, B(x) \cdot \vec{M}_V(x, r) = 0.
\]

(SI 86)

This extra constraint on the transformation follows from

\[
\tilde{\Phi}(r) = \frac{\partial}{\partial \tilde{r}} \int d^3x \, B(x) \cdot \vec{M}_V(x, r),
\]

(SI 87)

being in general a complicated function of \( \tilde{r} \). Therefore, it is not possible obtain for \( \tilde{r} \) in terms of \( \tilde{p} \) by inverting the canonical momentum Eqn. (11a). As an example of an allowed transformation, consider \( V = \frac{1}{2} r \times x \) and \( V_0 = 0 \). For this transformation, we find that \( \tilde{\Phi} = \frac{1}{2} \int d^3x \, x \times B \) which means that we can write \( \tilde{r} \) as a function of \( \tilde{p} \) by inverting Eqn. (11a). Importantly, this also means that the last term of Eq. (SI 85) vanishes, and we arrive at Eq. (13) in the main text.
SUPPLEMENTARY NOTE 4: ADDING AN IONIC LATTICE IN THE NEW APPROACH

An ionic lattice can be added to the new approach consistently under the assumption that it does not contribute any macroscopic current. This is justified given that ions vibrate about a mean position. To do so, we note that the partitioning of $\rho$ and $\mathbf{J}$ into free and bound charges was arbitrary. We could just as well make the partition $\rho = \rho_d + \rho_i$ and $\mathbf{J} = \mathbf{J}_d + \mathbf{J}_i$ where subscripts ‘d’ and ‘i’ denote charges that are bound in dipoles, and charges that are ions, respectively. The lattice is described by ions of charges $Q_k$ at positions $\mathbf{R}_k$ with charge density

$$\rho_i (\mathbf{x}, \mathbf{R}_k) = \sum_k Q_k \delta (\mathbf{x} - \mathbf{R}_k), \quad (\text{SI } 88)$$

and $\mathbf{J}_i = 0$. Note that to conserve charge without a current density, the charge density must not be an explicit function of time.

A field $\mathbf{P}$, analogous to the polarisation field $\mathbf{P}$, is then defined to be sourced by the dipoles: $\nabla \cdot \mathbf{P} = -\rho_d$ and a field $\mathbf{D}$ analogous to the displacement field $\mathbf{D}$ to be sourced by the ions: $\nabla \cdot \mathbf{D} = \rho_i$. If there are no ions, then the new $\mathbf{C}$-field theory outlined in the main text is unchanged so long as $\mathbf{P} \to \mathbf{P}$ and $\mathbf{D} \to \mathbf{D}$ because $\nabla \cdot \mathbf{E} = \rho$ is still satisfied for $\mathbf{E} = (\mathbf{D} - \mathbf{P})/\varepsilon_0$. Including the ions, the Maxwell equations Eqns. (6) become

$$\nabla \cdot \mathbf{D}^\parallel (\mathbf{x}, \mathbf{R}_k) = \rho_i (\mathbf{x}, \mathbf{R}_k), \quad (\text{SI } 89a)$$

$$\nabla \times \mathbf{H}(\mathbf{x}) = \mathbf{D}^\perp (\mathbf{x}), \quad (\text{SI } 89b)$$

where we have set $\mathbf{J}_i = 0$. Analogously to the main text, we write the fields in terms of vector potentials to satisfy Eqns. (SI 89). As before, $\mathbf{H} = \nabla \mathbf{C}_0 + \mathbf{C}$, however, the displacement field analogue requires a longitudinal component

$$\mathbf{D} (\mathbf{x}, \mathbf{R}_k) = \nabla \times \mathbf{C}(\mathbf{x}) + \mathbf{D}^\parallel (\mathbf{x}, \mathbf{R}_k), \quad (\text{SI } 90)$$

with the restrictions that $\nabla \cdot \mathbf{D}^\parallel = \rho_i$ and $\mathbf{D}^\parallel = 0$. The latter restriction comes from Eq. (SI 89b). Note that as before $\mathbf{D}^\perp (\mathbf{x}) = \nabla \times \mathbf{C}(\mathbf{x})$. These restrictions are satisfied by choosing

$$\mathbf{D}^\parallel (\mathbf{x}, \mathbf{R}_k) = -\nabla \int d^3x' \frac{\rho_i (\mathbf{x}', \mathbf{R}_k)}{4\pi |\mathbf{x} - \mathbf{x}'|} \equiv -\nabla V_i (\mathbf{x}, \mathbf{R}_k), \quad (\text{SI } 91)$$

where $V_i$ is the electrostatic potential of the ionic lattice. The inclusion of an ionic lattice to the Lagrangian Eq. (8) brings an additional kinetic term

$$L \to \tilde{L} = L + \sum_k \frac{1}{2} M_k \ddot{\mathbf{R}}_k^2, \quad (\text{SI } 92)$$

along with the replacements $\mathbf{P} \to \mathbf{P}$ and $\mathbf{D}^\perp \to \mathbf{D}^\perp + \mathbf{D}^\parallel$ where $\mathbf{D}^\perp = \mathbf{D}^\parallel = \nabla \times \mathbf{C}$. In order for the lattice to have been added consistently we must be able to derive the Lorentz force acting on the ions by using the Euler-Lagrange equation with ion position $\mathbf{R}_k$. The velocity derivative clearly results in $\partial \tilde{L}/\partial \mathbf{R}_k = M_k \ddot{\mathbf{R}}_k$, though the position derivative is more difficult. To start we rewrite Eqn. (SI 91) as

$$\mathbf{D}^\parallel (\mathbf{x}, \mathbf{R}_k) = -\sum_k Q_k \nabla \frac{1}{4\pi |\mathbf{x} - \mathbf{R}_k|} = \sum_k Q_k \frac{\partial}{\partial \mathbf{R}_k} \frac{1}{4\pi |\mathbf{x} - \mathbf{R}_k|}. \quad (\text{SI } 93)$$

We can then write the ion position derivative of the Lagrangian as

$$\frac{\partial \tilde{L}}{\partial \mathbf{R}_k} = \frac{\partial}{\partial \mathbf{R}_k} \int d^3x \left( \frac{\mathbf{D}^2 (\mathbf{x}, \mathbf{R}_k)}{2} + \mathbf{D} (\mathbf{x}, \mathbf{R}_k) \cdot \mathbf{P} (\mathbf{x}, \mathbf{r}) \right) \quad (\text{SI } 94)$$

$$= -\int d^3x \left( \mathbf{E}(\mathbf{x}) \cdot \frac{\partial}{\partial \mathbf{R}_k} \right) \mathbf{D}^\parallel (\mathbf{x}, \mathbf{R}_k), \quad (\text{SI } 95)$$

where we have enforced that $\mathbf{P}$ and $\mathbf{D}^\perp$ are not functions of $\mathbf{R}_k$ and that $\partial/\partial \mathbf{R}_k \times \mathbf{D}^\parallel = 0$ because $\mathbf{D}^\parallel$ is a longitudinal field with respect to $\mathbf{R}_k$. The $j$-th component of the integrand is therefore

$$\left[ \left( \mathbf{E}(\mathbf{x}) \cdot \frac{\partial}{\partial \mathbf{R}_k} \right) \mathbf{D}^\parallel (\mathbf{x}, \mathbf{R}_k) \right]_j = Q_k \left( E_i (\mathbf{x}) \frac{\partial}{\partial \mathbf{R}_k,i} \right) \frac{\partial}{\partial \mathbf{R}_k,j} \frac{1}{4\pi |\mathbf{x} - \mathbf{R}_k|}. \quad (\text{SI } 96)$$
where the sum over $i$ is implied. The longitudinal and transverse delta functions are [33]

$$
\delta^\parallel_{ij}(x) = -\lim_{\epsilon \rightarrow 0} \frac{\partial}{\partial x_i} \frac{\partial}{\partial x_j} \frac{1}{4\pi \sqrt{x^2 + \epsilon^2}} = \frac{1}{3} \delta_{ij}(x) + \frac{1}{4\pi |x|^3} \left( \delta_{ij} - \frac{x_i x_j}{|x|^2} \right),
$$

(SI 97)

$$
\delta_{ij}(x) = \delta^\parallel_{ij}(x) + \delta^\perp_{ij}(x),
$$

(SI 98)

where $\delta_{ij}$ is a Kronecker-delta and $\delta_{ij}(x) \equiv \delta_{ij}(x)$.

Therefore,

$$
\left[ \left( \mathbf{E}(x) \cdot \frac{\partial}{\partial \mathbf{R}_k} \right) \mathbf{D}^\parallel (x, \mathbf{R}_k) \right]_j = -Q_k E_i(x) \delta^\parallel_{ij} (x - \mathbf{R}_k),
$$

(SI 99)

which gives the Lorentz force

$$
M_k \ddot{\mathbf{R}}_k = Q_k \mathbf{E}^\parallel(\mathbf{R}_k).
$$

(SI 100)

The lack of magnetic force and transverse electric force originates from assuming that the ions do not generate any current. Therefore, the ions are not affected by magnetic forces and the electrostatic force is conservative which means it has zero curl and so no transverse component.

Since $\mathbf{D}^\parallel = 0$, the mechanical degree of freedom $\mathbf{D}^\parallel$ does not have a canonical momentum. The ion position does, however, have a canonical momentum given by

$$
\mathbf{P}_k = \frac{\partial L}{\partial \dot{\mathbf{R}}_k} = M_k \dot{\mathbf{R}}_k,
$$

(SI 101)

which brings an additional kinetic energy term to the total Hamiltonian $\bar{H}$. This Hamiltonian is then found to be (using $\mathbf{V} = V_0 = 0$)

$$
\bar{H} = \frac{1}{2m} [\mathbf{p} + \Phi(\mathbf{r})]^2 + \frac{1}{2} \int d^3x \left( \frac{1}{\varepsilon_0} \left[ \mathbf{D}^\parallel(\mathbf{x}) - \mathbf{P}(\mathbf{x}, \mathbf{r}) \right]^2 + \frac{1}{\mu_0} \mathbf{B}^2(\mathbf{x}) \right) + \sum_k \frac{\mathbf{P}_k^2}{2M_k} + \frac{1}{\varepsilon_0} \int d^3x \left[ \frac{1}{2} \left[ \mathbf{D}^\parallel(\mathbf{x}, \mathbf{R}_k) \right]^2 + \mathbf{D}^\parallel(\mathbf{x}, \mathbf{R}_k) \cdot \mathbf{P}(\mathbf{x}, \mathbf{r}) \right].
$$

(SI 102)

As described in the main text, we can quantise the $\mathbf{C}^\perp$-field as in Eqn. (14). The radiation fields are quantised exactly as without the ionic lattice, described in SI-2. That is, for the $\mathbf{C}^\perp$-field is quantised as in Eqn. (14), $\mathbf{D}^\perp$ takes the form

$$
\hat{\mathbf{D}}^\perp(\mathbf{x}) = -i \sum_{k\lambda} (\mathbf{k} \times \mathbf{e}_{k\lambda}) f_k \left( \hat{a}_{k\lambda}^\dagger e^{-i\mathbf{k} \cdot \mathbf{x}} - \hat{a}_{k\lambda} e^{i\mathbf{k} \cdot \mathbf{x}} \right),
$$

(SI 103)

and the magnetic field is just the canonical momentum

$$
\hat{\mathbf{B}}^\perp(\mathbf{x}) = i \sum_{k\lambda} \mu_k \mathbf{e}_{k\lambda} f_k \left( \hat{a}_{k\lambda}^\dagger e^{-i\mathbf{k} \cdot \mathbf{x}} - \hat{a}_{k\lambda} e^{i\mathbf{k} \cdot \mathbf{x}} \right).
$$

(SI 104)

After quantising the radiation fields in this way, it is clear that the first line of Eqn. (SI 102) will lead to the Hamiltonian describing the dipole interacting with the light field that is given in the main text in Eqn. (15), so long as the magnetic effects in $\Phi$ are ignored and the electric dipole approximation is made to simplify $\mathbf{P}(\mathbf{x}) \approx -e \overline{\delta}(\mathbf{x})$. Quantising the second line of Eqn. (SI 102) leads to a description of the influence (on the dipole) of vibrations in the ionic lattice from their mean positions. From Eqn. (SI 91) we can see that the total energy of the phonon field is described by the terms

$$
\sum_k \frac{\mathbf{P}_k^2}{2M_k} + \int d^3x \left[ \frac{\mathbf{D}^\parallel(\mathbf{x}, \mathbf{R}_k)}{2} \right]^2 = \sum_{\mathbf{q}\mu} \omega_{\mathbf{q}\mu} \left( \hat{b}_{\mathbf{q}\mu}^\dagger \hat{b}_{\mathbf{q}\mu} + \frac{1}{2} \right),
$$

(SI 105)

where $b_{\mathbf{q}\mu}$ ($b_{\mathbf{q}\mu}^\dagger$) is the annihilation (creation) operator of a phonon of wavenumber $\mathbf{q}$ and polarisation $\mu$. Details of the derivation of Eqn. (SI 105) can be found in, for example, [5, 6, 31, 34]. The final term in the Hamiltonian, $\int d^3x \mathbf{D}^\parallel \cdot \mathbf{\dot{P}}$, describes the interaction between the phonons and the dipole. After linearising this interaction by
assuming that the ions are only displaced from equilibrium by small amounts, one can then show that in the EDA
[5, 6, 31, 34]

\[
\frac{1}{\varepsilon_0} \int d^3 x \, \mathbf{D}^\parallel (\mathbf{x}, \mathbf{R}_k) \cdot \hat{\mathbf{P}} (\mathbf{x}, \mathbf{r}) \approx \sum_{\mathbf{q}\mu} \left( \hat{b}_{\mathbf{q}\mu}^\dagger + \hat{b}_{\mathbf{q}\mu} \right) \sum_{n=1}^N g_{n\mathbf{q}\mu} |\epsilon_n\rangle \langle \epsilon_n|,
\tag{SI 106}
\]

which is the usual phonon interaction encountered throughout the literature where the coupling strength of dipole
level \( n \) to phonon mode with wavenumber \( \mathbf{q} \) and polarisation \( \mu \) is \( g_{n\mathbf{q}\mu} \). We can finally write the full, gauge-invariant Hamiltonian describing the dynamics of a dipole in an EM field interacting with a vibrating ionic lattice:

\[
\hat{H} = \sum_{n=0}^N \epsilon_n |\epsilon_n\rangle \langle \epsilon_n| - \frac{1}{\varepsilon_0} \hat{\mathbf{d}} \cdot \left( \nabla \times \mathbf{C}^\perp (0) \right) + \frac{1}{\varepsilon_0} \sum_{\mathbf{k}\nu} f_{\mathbf{k}\nu}^2 \mathbf{\nu}_\mathbf{k} \left( \hat{\mathbf{d}} \cdot \mathbf{\epsilon}_{\mathbf{k}\lambda} \right)^2 + \sum_{\mathbf{k}\lambda} \mathbf{\nu}_\mathbf{k} \hat{a}_{\mathbf{k}\lambda}^\dagger \hat{a}_{\mathbf{k}\lambda} \tag{SI 107}
\]

\[
+ \sum_{\mathbf{q}\mu} \left( \hat{b}_{\mathbf{q}\mu}^\dagger + \hat{b}_{\mathbf{q}\mu} \right) \sum_{n=1}^N g_{n\mathbf{q}\mu} |\epsilon_n\rangle \langle \epsilon_n| + \sum_{\mathbf{q}\mu} \omega_{\mathbf{q}\mu} \hat{b}_{\mathbf{q}\mu}^\dagger \hat{b}_{\mathbf{q}\mu}. \tag{SI 108}
\]
### SUPPLEMENTARY NOTE 5: TABLE OF EXPERIMENTAL DATA POINTS

| Experiment                           | \( \eta \)  | \( f_{\mu eV} \) | Dipole size / \( \mu m \) | \( g/\nu \)    | \( g/\nu_{10} \) | References |
|-------------------------------------|-------------|---------------|-----------------|----------|-------------|------------|
| Cs in optical cavity                | 3.1 \times 10^{-5} | 6.5 \times 10^{4} | 1.7 \times 10^{-4} | 2.2 \times 10^{-6} | 2.2 \times 10^{-6} | [35–37]   |
| Rb gas in optical cavity            | 2.0 \times 10^{-4} | 1.6 \times 10^{-4} | 2.48 \times 10^{-10} | 4.28 \times 10^{-10} | 4.28 \times 10^{-10} | [38–41]   |
| Quantum dot arrays                  | 0.012       | 0.023         | 0.01            | 5.3 \times 10^{-4} | 6 \times 10^{-4} | [42–46]   |
| Superconducting circuit             | -5.9 \times 10^{-4} | -0.029        | -60             | -1.3       | -15.3       | [47–51]   |
| Rare earth spins in microwave       | -5.2 \times 10^{-3} | 3.3 \times 10^{-4} | 299.4          | 0.15       | 0.051       | [52]       |
| resonator                           | 0.0091      | 0.11          | 0.0070         | 0.0012     | 0.0012      | [53, 54]   |
| polaritons                          | -0.0098     | -0.51         | -0.0076        | -0.0060    | -0.0060     | [55–58]   |
| Exciton polaritons in dyes          | 6.0 \times 10^{-4} | 44.94        | 5 \times 10^{-4} | 0.035      | 0.043       | [55–58]   |
| Inter-subband polaritons in quantum | 3.0 \times 10^{-4} | 4.7          | 0.0075         | 0.023      | 0.032       | [59–63]   |
| Electron cyclotron resonances       | 1.7 \times 10^{-6} | 0.76         | 0.073          | 0.17       | 0.058       | [64–67]   |

We have used various estimates when the dipole size has not been reported. In particular:

- The dipole size \( x_{10} \) is approximately a couple of Bohr radii \( a_0 \) for atomic species, whereas the effective Bohr radius \( a_{\text{eff}} \approx 1 \mu m \) is used for Rydberg atoms.
- For quantum dots and quantum wells, the physical extent of the device is a good estimate for the dipole size.
- The estimates become slightly more complicated for superconducting circuits, but an approximate value can be extracted in two different ways, depending on the values reported: First from the definition of the dipole moment

\[
x_{10} \approx |\mathbf{d}|/e \quad \text{with} \quad |\mathbf{d}| = \hbar g/E_{\text{vac}},
\]

\[
\Rightarrow x_{10} \approx \hbar g/(eE_{\text{vac}}),
\]

where \( g \) is the interaction strength, and \( E_{\text{vac}} \) is the electric field amplitude of the vacuum fluctuations. Electric field fluctuations can in turn be estimated by \( E_{\text{vac}} \approx (V_{\text{vac}}/\text{length of relevant region}) \), with \( V_{\text{vac}} \) being the fluctuations in the electric potential. Alternatively, it is possibly to define an analogous Bohr radius \( a_{\text{eff}} \) through the resonance frequency, as in

\[
\omega_{10} = 2\pi c/\lambda_{10} = 2\pi c \left( \frac{\alpha}{4\pi a_{\text{eff}}} \right),
\]

\[
\Rightarrow a_{\text{eff}} \approx \frac{\alpha c}{2\omega_{10}},
\]

where \( \alpha \) is the fine-structure constant and where we have interpreted the transition wavelength \( \lambda_{10} = 4\pi a_{\text{eff}}/\alpha \) as an effective Bohr wavelength.
- In dye-filled polariton cavities, the spatial extent of the dye molecule acts as a bound of the dipole size.
- In the case of electron cyclotron resonances, we can estimate the dipole size as

\[
x_{10} \approx l_0 \sqrt{\nu},
\]

where \( l_0 = \sqrt{\hbar/(eB)} \) is the magnetic length for a given magnetic field strength \( B \), and \( \nu = \rho_{2\text{deg}} 2\pi l_0^2 \) is the filling factor in the system with \( \rho_{2\text{deg}} \) being the electron density. Note that in some cases, the filling factor is directly reported.
SUPPLEMENTARY NOTE 6: ACCURACY OF FEW LEVEL TRUNCATION WITHOUT RESONANCE

Figure 3 is the same calculation as for Figure 2, however, the splitting between the two lowest lying dipole levels is no longer kept resonant with the single photon mode \( \nu \). Instead, we fix the length of the infinite square well such that the lowest lying dipole transition, \( \omega_{10} = \epsilon_1 - \epsilon_0 = 1 \text{ eV} \) is held constant. Outwith resonance, the light-matter coupling ratios with \( \omega_{10} \) and \( \nu \) in either gauge is not the same, and so the contours along the diagonal- and \( x \)-axes are different. Just as in Figure 2, even without resonance the C-field calculations yield more accurate results for a given coupling strength than the Coulomb gauge, and also converge onto the exact answer quicker as the number of dipole levels in the calculation is increased. Note also that it is for large dipoles in the strong-coupling limit that the C-field calculations become inaccurate, i.e. in the scenario when we expect the description of the polarisation field in terms of dipole degrees of freedom to become non-trivial.

![Relative error diagram](image)

**FIG. 3.** As in Figure 2, we plot the relative error in calculating the energy spacing between the two lowest eigenstates of the full Coulomb gauge and C-field Hamiltonians within the two and three dipole-level approximations, using an infinite square well potential as \( \hat{U}_{\text{ext}} \). The difference is that in this plot the lowest lying dipole transition is kept fixed at 1 eV, and so, is not on resonance with the mode frequency which is varied along the \( y \)-axes. For all other details, refer to Figure 2.