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Resolving ambiguities of the mode truncation in cavity quantum electrodynamics

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This work provides the fundamental theoretical framework for few-mode cavity quantum electrodynamics by resolving the gauge ambiguities between the Coulomb gauge and the dipole gauge Hamiltonians under the photonic mode truncation. We first propose a general framework to resolve ambiguities for an arbitrary truncation in a given gauge. Then, we specifically consider the case of mode truncation, deriving gauge invariant expressions for both the Coulomb and dipole gauge Hamiltonians that naturally reduce to the commonly used single-mode Hamiltonians when considering a single-mode truncation. We finally provide the analytical and numerical results of both atomic and molecular model systems coupled to the cavity to demonstrate the validity of our theory.

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In cavity quantum electrodynamics (cQED), the single-mode approximation has been ubiquitously applied across multitudes of studies [1–11]. As this approximation breaks down, increasingly more modes must be considered [12–19]. However, the choice of gauge for a proper truncation of the photonic modes leads to a potential ambiguity [13], as one arrives at two different dipole gauge Hamiltonians when performing mode truncation before or after performing the Power–Zienau–Wooley (PZW) gauge transformation. In this Letter, we provide a theoretical framework to address this ambiguity and demonstrate how to accurately perform mode truncation in both the Coulomb and dipole gauges.

We begin by introducing the PZW gauge transformation operator [20,21] in the long-wavelength approximation as

$$\hat{U} = e^{-\frac{i}{\hbar}\hat{\boldsymbol{\mu}}\cdot\hat{\mathbf{A}}} = \exp[-\frac{i}{\hbar}\hat{\boldsymbol{\mu}}\cdot\sum_{k=0}^{\infty}\mathbf{A}_k(\hat{a}_k^\dagger + \hat{a}_k)], \tag{1}$$

where $\hat{\boldsymbol{\mu}} = \sum_j z_j \boldsymbol{q}_j$ is the total dipole operator, $\boldsymbol{q}_j$ is the position of the charged particle j (including all electrons and nuclei) with a charge of z_j, and $\hat{\mathbf{A}} = \sum_{k=0}^{\infty}\mathbf{A}_k(\hat{a}_k^\dagger + \hat{a}_k)$ is the purely transverse vector potential under the Coulomb gauge ($\nabla\cdot\boldsymbol{A} = 0$) and long-wavelength approximation. The vector potential amplitude is $\mathbf{A}_k = \sqrt{\hbar/2\omega_k\varepsilon\mathcal{V}}\cdot\hat{\mathbf{e}}$, with $\mathcal{V}$ as the quantization volume inside the cavity, ε as the vacuum permittivity, and $\hat{\mathbf{e}}$ as the unit vector of the field polarization. The PZW transformation operator can also be expressed as $\hat{U} = \exp\left[-\frac{i}{\hbar}\sqrt{2\omega_c/\hbar}\hat{\boldsymbol{\mu}}\cdot\sum_{k=0}^{\infty}\mathbf{A}_k\hat{q}_{c,k}\right] = \exp\left[-\frac{i}{\hbar}(\sum_j z_j\hat{\mathbf{A}}\boldsymbol{q}_j)\right]$. Recall that a momentum boost operator of the form $\hat{U}_p = e^{-\frac{i}{\hbar}p_0\hat{q}}$ displaces $\hat{p}$ by the amount p_0, such that $\hat{U}_p\hat{O}(\hat{p})\hat{U}_p^\dagger = \hat{O}(\hat{p}+p_0)$. The PZW can then be thought of simultaneously as a boost operator for both the matter momentum and the photonic momentum.

The dipole gauge (or the d · E form) and Coulomb gauge (or the p · A form) Hamiltonians for describing light–matter interactions can then be written [8,22,23] in terms of the PZW operator $\hat{U}$ as

$$\hat{H}_{\mathrm{p\cdot A}} = \hat{U}^\dagger\hat{H}_{\mathrm{M}}\hat{U} + \sum_{k=0}^{\infty}\hbar\omega_k\hat{a}_k^\dagger\hat{a}_k, \tag{2}$$

$$\hat{H}_{\mathrm{d\cdot E}} = \hat{H}_{\mathrm{M}} + \hat{U}\Big(\sum_{k=0}^{\infty}\hbar\omega_k\hat{a}_k^\dagger\hat{a}_k\Big)\hat{U}^\dagger, \tag{3}$$

where the matter Hamiltonian is $\hat{H}_{\mathrm{M}} = \sum_j \frac{p_j^2}{2m_j} + \hat{V}(\{\boldsymbol{q}_j\})$, the photonic Hamiltonian is $\hat{H}_{\mathrm{ph}} = \sum_{k=0}^{\infty}\hbar\omega_k\hat{a}_k^\dagger\hat{a}_k$, and $\hbar\omega_k\hat{a}_k^\dagger\hat{a}_k$ is the photonic Hamiltonian for the k_{th} mode. Note that in $\hat{H}_{\mathrm{d\cdot E}}$ (dipole gauge), $\hat{U}$ boosts the photonic momentum, and in $\hat{H}_{\mathrm{p\cdot A}}$ (Coulomb gauge), $\hat{U}$ boosts the matter momentum. More specifically, one has the well-known minimum coupling form for the p · A Hamiltonian as $\hat{H}_{\mathrm{p\cdot A}} = \sum_j \frac{1}{2m_j}(\hat{\mathbf{p}}_j - z_j\hat{\mathbf{A}})^2 + \hat{V}(\hat{\mathbf{x}}) + \hat{H}_{\mathrm{ph}}$, ensuring gauge invariance under any gauge transformation (such as the PZW transformation). The d · E Hamiltonian [Eq. (3)] can also be explicitly expressed as $\hat{H}_{\mathrm{d\cdot E}} = \hat{H}_{\mathrm{M}} + \hat{H}_{\mathrm{ph}} + \sum_{k=0}^{\infty}[i\omega_k\mathbf{A}_k\cdot\hat{\boldsymbol{\mu}}(\hat{a}_k^\dagger - \hat{a}_k) + \frac{\omega_k}{\hbar}(\mathbf{A}_k\cdot\hat{\boldsymbol{\mu}})^2]$.

Exact QED theory should be gauge invariant, which can be easily seen by noticing that $\hat{H}_{\mathrm{d\cdot E}} = \hat{U}\hat{H}_{\mathrm{p\cdot A}}\hat{U}^\dagger$. Gauge ambiguities, meaning different values for a quantum observable obtained from different gauges, could arise due to either matter state truncation (such as two-level approximations) [7,8] or the mode truncation [13]. In efforts to resolve these, it is commonly asked whether it is most appropriate to truncate the Hamiltonian in the Coulomb gauge or the dipole gauge [7,12,13]. We propose a new theoretical framework to consider truncations in light–matter interaction Hamiltonians by performing the truncation further upstream.

Whether the light–matter coupling is expressed in the Coulomb gauge or the dipole gauge, the total Hamiltonian is composed of a pure matter Hamiltonian operator, $\hat{H}_{\mathrm{M}}$, and a pure photonic operator, $\hat{H}_{\mathrm{ph}}$, where one of these operators' momentum is boosted by a unitary PZW gauge transformation operator

$\hat{U}$ [see Eqs. (2) and (3)]. Since one typically truncates the Hilbert space in the tensor product of the eigenstates of $\hat{H}_{\rm M}$ or $\hat{H}_{\rm ph}$ to form a properly truncated light–matter Hamiltonian, one must first project $\hat{H}_{\rm M}$ and $\hat{H}_{\rm ph}$ in that subspace and then evaluate the coupling terms by using the PZW operator which is also properly contained within the same subspace [8,23–25]. It should be noted that for a given truncation using the projection operator $\hat{\mathcal{P}}$, a simple truncation of the form $\hat{\mathcal{P}}\hat{U}\hat{\mathcal{P}}$ is not properly contained in the subspace $\hat{\mathcal{P}}$ [12,23,24], leading to the well-known gauge ambiguity for the matter state truncation [23]. To remedy this, one can construct a new PZW operator [23] in the same subspace as

$$\hat{\mathcal{U}} = \exp[-\frac{i}{\hbar}\hat{\mathcal{P}}(\hat{\boldsymbol{\mu}}\cdot\hat{\boldsymbol{A}})\hat{\mathcal{P}}], \tag{4}$$

such that it is properly confined in the subspace $\hat{\mathcal{P}}$, which is apparent because $\hat{\mathcal{U}} = \sum_{n=0}^{\infty}\frac{1}{n!}(\frac{i}{\hbar}\hat{\mathcal{P}}(\hat{\boldsymbol{\mu}}\cdot\hat{\boldsymbol{A}})\hat{\mathcal{P}})^n$. Note that in general, $\hat{\mathcal{P}} = \hat{\mathcal{P}}_{\rm M}\otimes\hat{\mathcal{P}}_{\rm ph}$, where $\hat{\mathcal{P}}_{\rm M}$ and $\hat{\mathcal{P}}_{\rm ph}$ are projection operators for the matter and the photonic degrees of freedom (DOFs), respectively, and $\hat{\mu}$ and $\hat{\mathbf{A}}$ are operators in the matter and photonic Hilbert spaces, respectively. We can then truncate the Coulomb and dipole gauge Hamiltonians within an arbitrary subspace $\hat{\mathcal{P}}$ (of the combined matter and photonic Hilbert space) as follows [23]:

$$\hat{\mathcal{H}}_{\rm p\cdot A} = \hat{\mathcal{U}}^{\dagger}\hat{\mathcal{P}}\hat{H}_{\rm M}\hat{\mathcal{P}}\hat{\mathcal{U}} + \hat{\mathcal{P}}\hat{H}_{\rm ph}\hat{\mathcal{P}}, \tag{5}$$

$$\hat{\mathcal{H}}_{\rm d\cdot E} = \hat{\mathcal{P}}\hat{H}_{\rm M}\hat{\mathcal{P}} + \hat{\mathcal{U}}\hat{\mathcal{P}}\hat{H}_{\rm ph}\hat{\mathcal{P}}\hat{\mathcal{U}}^{\dagger}, \tag{6}$$

which always guarantees gauge invariant results through $\hat{\mathcal{H}}_{\rm d\cdot E} = \hat{\mathcal{U}}\hat{\mathcal{H}}_{\rm p\cdot A}\hat{\mathcal{U}}^{\dagger}$. The special case of this approach is recently used to resolve the gauge ambiguity due to the matter state truncation [8,23].

With this generalized theory outlined in Eqs. (4)–(6), we turn our focus to a specific problem in cavity QED: the mode truncation. To begin our discussion, we define the projection operator to the first m photonic modes (labeled as $k\in[0, m-1]$) as

$$\hat{\mathcal{P}}^{(m)} = \hat{\mathcal{I}}_{\rm M}\otimes\Big(\bigotimes_{k=0}^{m-1}\sum_{n=0}^{\infty}|n_k\rangle\langle n_k|\bigotimes_{k'=m}^{\infty}|0_{k'}\rangle\langle 0_{k'}|\Big), \tag{7}$$

where $|n_k\rangle$ is the $n_{\rm th}$ Fock state in the $k_{\rm th}$ mode and $\hat{\mathcal{I}}_M$ is the identity operator for the matter Hilbert space, thus we do not explicitly consider the matter subspace truncation in this study. Note that $|0_{k'}\rangle\langle 0_{k'}|$ confines the Hilbert space's $k'_{\rm th}$ photonic mode to its vacuum state, such that this mode neither participates in the light–matter interactions [this is apparent by looking at the light–matter interaction Hamiltonian in Eq. (11), as well as in the PZW operator in Eq. (4), because $\langle 0_k|(\hat{a}_k^{\dagger}+\hat{a}_k)|0_k\rangle = 0$] nor explicitly shows up in $\hat{\mathcal{P}}^{(m)}\hat{H}_{\rm ph}\hat{\mathcal{P}}^{(m)}$. The physical device to effectively accomplish this mode truncation is the long-pass filter [25], which prevents the photonic excitations in those high-frequency modes $k\ge m$, or a simple dielectric Bragg reflector (DBR) cavity which has a mode-dependent quality factor [26], such that the lifetime of the photonic excitation goes to 0 for those higher frequency modes.

With our general procedure outlined in Eq. (4), we introduce the PZW operator that is properly confined in the m modes subspace (for $k\in[0, m-1]$) as

$$\hat{\mathcal{U}}^{(m)} = e^{-\frac{i}{\hbar}\hat{\mathcal{P}}^{(m)}(\hat{\boldsymbol{\mu}}\cdot\hat{\boldsymbol{A}})\hat{\mathcal{P}}^{(m)}} = \exp[-\frac{i}{\hbar}\hat{\boldsymbol{\mu}}\cdot\sum_{k=0}^{m-1}\boldsymbol{A}_k(\hat{a}_k^{\dagger}+\hat{a}_k)]. \tag{8}$$

Using Eq. (5), the Coulomb gauge Hamiltonian under an n-mode truncation is expressed as

$$\begin{aligned}\hat{\mathcal{H}}^{(m)}_{\rm p\cdot A} &= \hat{\mathcal{U}}^{(m)\dagger}\hat{\mathcal{P}}^{(m)}\hat{H}_{\rm M}\hat{\mathcal{P}}^{(m)}\hat{\mathcal{U}}^{(m)} + \hat{\mathcal{P}}^{(m)}\sum_{k=0}^{\infty}\hbar\omega_k\hat{a}_k^{\dagger}\hat{a}_k\hat{\mathcal{P}}^{(m)}\\ &= \hat{H}_{\rm M} + \sum_{k=0}^{m-1}\Big[\hbar\omega_k\hat{a}_k^{\dagger}\hat{a}_k + \frac{\hat{\boldsymbol{p}}\cdot\boldsymbol{A}_k}{m}(\hat{a}_k^{\dagger}+\hat{a}_k)\Big] + \frac{1}{2m}\Big[\sum_{k=0}^{m-1}|\boldsymbol{A}_k|(\hat{a}_k^{\dagger}+\hat{a}_k)\Big]^2.\end{aligned} \tag{9}$$

As $\hat{H}_{\rm M}$ is a pure matter operator, it is invariant upon mode truncation and therefore commutes with $\hat{\mathcal{P}}$, $\hat{\mathcal{P}}^{(m)}\hat{H}_{\rm M}\hat{\mathcal{P}}^{(m)} = \hat{H}_{\rm M}\hat{\mathcal{P}}^{(m)}$. In the case of a single mode $n=1$, Eq. (9) reduces to the well-known single-mode minimal coupling Hamiltonian. Interestingly, if we apply a simple mode truncation, $\hat{\mathcal{H}}'^{(m)}_{\rm p\cdot A} = \hat{\mathcal{P}}^{(m)}\hat{H}_{\rm p\cdot A}\hat{\mathcal{P}}^{(m)}$ has the same form of Eq. (9) up to a constant that represents the zero-point energy of all modes (see Supplement 1, Section 1.).

If we apply a simple truncation procedure to the dipole gauge Hamiltonian, we have

$$\begin{aligned}\hat{\mathcal{H}}'^{(m)}_{\rm d\cdot E} &= \hat{\mathcal{P}}^{(m)}\hat{H}_{\rm d\cdot E}\hat{\mathcal{P}}^{(m)} = \hat{H}_{\rm M} + \hat{\mathcal{P}}^{(m)}\hat{U}\Big(\sum_{k=0}^{\infty}\hbar\omega_k\hat{a}_k^{\dagger}\hat{a}_k\Big)\hat{U}^{\dagger}\hat{\mathcal{P}}^{(m)}\\ &= \hat{H}_{\rm M} + \sum_{k=0}^{m-1}\Big[\hbar\omega_k\hat{a}_k^{\dagger}\hat{a}_k + i\omega_k\boldsymbol{A}_k\cdot\hat{\boldsymbol{\mu}}(\hat{a}_k^{\dagger}-\hat{a}_k)\Big] + \sum_{k=0}^{\infty}\frac{\omega_k}{\hbar}(\boldsymbol{A}_k\cdot\hat{\boldsymbol{\mu}})^2.\end{aligned} \tag{10}$$

This procedure breaks the gauge invariance and generates different results from $\hat{\mathcal{H}}^{(m)}_{\rm p\cdot A}$, because the dipole self-energies for all the modes are still explicitly present, even for the modes $k\in[n,\infty]$ which are supposed to be truncated in the d · E term. The above

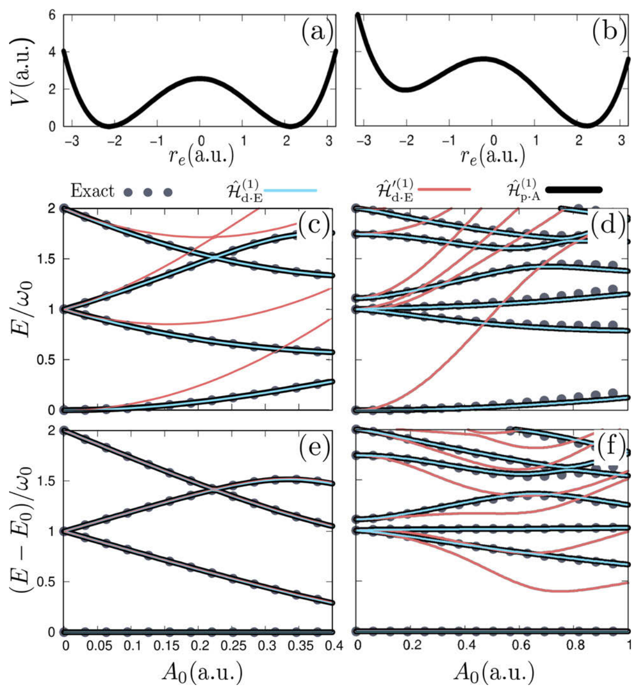

Fig. 1. Particle in a double-well potential coupled to the cavity. (a) Symmetric and (b) asymmetric double-well potential for the matter. (c) Polariton eigenspectra of different Hamiltonians as a function of coupling strength, A_0, for the symmetric double-well potential in panel (a). (d) Polariton eigenspectra of different Hamiltonians as a function of coupling strength for the asymmetric double well potential in panel (b). (e) and (f) Polariton eigenspectra relative to E_0.

inappropriate mode truncation was mentioned in the procedure outlined in Ref. [13], where the mode truncation is done after the PZW transformation of the minimal coupling Hamiltonian, and only on for the light–matter coupling term, but not for the dipole self-energy term. The problem with Eq. (10) arises from the fact that $\hat{\mathcal{P}}^{(m)}\hat{U}$ is not a unitary transformation [12] of $\sum_{k=0}^{\infty}\hbar\omega_k\hat{a}_k^{\dagger}\hat{a}_k$, which leads to operators outside the $\hat{\mathcal{P}}^{(m)}$ subspace [i.e., the dipole self energy (DSE) term $\sum_{k=m}^{\infty}\frac{\omega_k}{\hbar}(\boldsymbol{A}_k\cdot\hat{\boldsymbol{\mu}})^2$ in Eq. (10)]. This DSE term in $\hat{\mathcal{H}}'^{(m)}_{\mathrm{d\cdot E}}$ contains an infinite summation of dipole self-energy terms that does not converge.

With the procedure outlined in Eq. (6), we then obtain a properly truncated d · E Hamiltonian as follows:

$$\hat{\mathcal{H}}^{(m)}_{\mathrm{d.E}} = \hat{H}_{\mathrm{M}} + \hat{\mathcal{U}}^{(m)}\hat{\mathcal{P}}^{(m)}\Big(\sum_{k=0}^{\infty}\hbar\omega_k\hat{a}_k^{\dagger}\hat{a}_k\Big)\hat{\mathcal{P}}^{(m)}\hat{\mathcal{U}}^{(m)\dagger}$$
$$=\hat{H}_{\mathrm{M}} + \sum_{k=0}^{n-1}\Big[\hbar\omega_k\hat{a}_k^{\dagger}\hat{a}_k + i\omega_k\boldsymbol{A}_k\cdot\hat{\boldsymbol{\mu}}(\hat{a}_k^{\dagger}-\hat{a}_k) + \frac{\omega_k}{\hbar}(\boldsymbol{A}_k\cdot\hat{\boldsymbol{\mu}})^2\Big]. \quad (11)$$

The truncated Hamiltonian in Eq. (11) preserves the same form of Eq. (3), where the pure photonic operator in the truncated subspace is acted on by a unitary boost operator. This Hamiltonian does not have the non-converging infinite sum for DSE that is (incorrectly) shown in Eq. (10).

To illuminate the effects of these different mode truncation schemes on the energy eigenspectra of polaritonic systems, let us consider a simple model system that contains only two cavity modes in the p · A form as follows:

$$\hat{\mathcal{H}}^{(2)}_{\mathrm{p\cdot A}} = \hat{H}_{\mathrm{M}} + \sum_{k=0}^{1}\Big[\hbar\omega_k\hat{a}_k^{\dagger}\hat{a}_k + \frac{\hat{\boldsymbol{p}}\cdot\boldsymbol{A}_k}{m}(\hat{a}_k^{\dagger}+\hat{a}_k)\Big] + \frac{1}{2m}\Big[\sum_{k=0}^{1}|\boldsymbol{A}_k|(\hat{a}_k^{\dagger}+\hat{a}_k)\Big]^2, \quad (12)$$

whereas the equivalent d · E form is $\hat{\mathcal{H}}^{(2)}_{\mathrm{d.E}} = \hat{H}_{\mathrm{M}} + \sum_{k=0}^{1}[\hbar\omega_k\hat{a}_k^{\dagger}\hat{a}_k + i\omega_k\boldsymbol{A}_k\cdot\hat{\boldsymbol{\mu}}(\hat{a}_k^{\dagger}-\hat{a}_k) + \frac{\omega_k}{\hbar}(\boldsymbol{A}_k\cdot\hat{\boldsymbol{\mu}})^2]$. We then consider three different possible Hamiltonians truncated to a single cavity mode: (a) a single-mode Coulomb gauge Hamiltonian; (b) an inappropriately truncated single-mode dipole gauge Hamiltonian; and (c) a properly truncated single-mode dipole gauge Hamiltonian, as follows:

$$\hat{\mathcal{H}}^{(1)}_{\mathrm{p\cdot A}} = \hat{H}_{\mathrm{M}} + \hat{H}^{0}_{\mathrm{ph}} + \frac{\hat{\boldsymbol{p}}\cdot\boldsymbol{A}_0}{m}(\hat{a}_0^{\dagger}+\hat{a}_0) + \frac{|\boldsymbol{A}_0|^2}{2m}(\hat{a}_0^{\dagger}+\hat{a}_0)^2, \quad (13a)$$
$$\hat{\mathcal{H}}'^{(1)}_{\mathrm{d\cdot E}} = \hat{H}_{\mathrm{M}} + \hat{H}^{0}_{\mathrm{ph}} + i\omega_0\boldsymbol{A}_0\cdot\hat{\boldsymbol{\mu}}(\hat{a}_0^{\dagger}-\hat{a}_0) + \sum_{k=0,1}\frac{\omega_k}{\hbar}(\boldsymbol{A}_k\cdot\hat{\boldsymbol{\mu}})^2, \quad (13b)$$
$$\hat{\mathcal{H}}^{(1)}_{\mathrm{d\cdot E}} = \hat{H}_{\mathrm{M}} + \hat{H}^{0}_{\mathrm{ph}} + i\omega_0\boldsymbol{A}_0\cdot\hat{\boldsymbol{\mu}}(\hat{a}_0^{\dagger}-\hat{a}_0) + \frac{\omega_0}{\hbar}(\boldsymbol{A}_0\cdot\hat{\boldsymbol{\mu}})^2, \quad (13c)$$

where $\hat{H}^{0}_{\mathrm{ph}} = \hbar\omega_0\hat{a}_0^{\dagger}\hat{a}_0$.

Figure 1 presents the energy eigenspectra of a single particle experiencing symmetrical and asymmetrical double-well potential [8] $\hat{V}(\hat{r}) = \frac{a}{4}\hat{r}^4 - \frac{b}{2}(\hat{r}-\gamma)^2$ which is then coupled to the optical cavity. Here, $a, b>0$ are parameters used to for the double-well shape, and γ is an asymmetry factor. In this work, we use the values $a = 0.5$ and $b = 2.27$. The results from various truncated Hamiltonians are compared relative to the exact results. Figure 1(a) presents the symmetrical potential of the matter by setting $\gamma = 0$. Due to no permanent dipole in this symmetric potential, under the two-level approximation, $\hat{\mu} \approx \mu_{01}(|\phi_1\rangle\langle\phi_0| + |\phi_0\rangle\langle\phi_1|)$, $\hat{\mu}^2 \approx |\mu_{01}|^2(|\phi_0\rangle\langle\phi_0| + |\phi_1\rangle\langle\phi_1|)$. For $\hat{\mathcal{H}}'^{(1)}_{\mathrm{d\cdot E}}$ in Eq. (13b), the constant transition dipole moment causes the extra constant DSE term (for the $k = 1$ mode) to be a zero-point energy shift for all polariton states, weighted by a quadratic coupling strength $|\boldsymbol{A}_1|^2$ for mode $n = 1$ (note that $|\boldsymbol{A}_1|^2 = (\omega_0/\omega_1)\cdot|\boldsymbol{A}_0|^2$). We emphasize that this two-level approximation is not used in any of our numerical results presented in this work, but only used to intuitively understand the numerical results provided in Figs. 1(c) and 1(e).

Figure 1(c) presents the polariton eigenspectra obtained by diagonalizing $\hat{H}$, $\hat{\mathcal{H}}^{(1)}_{\mathrm{p\cdot A}}$, $\hat{\mathcal{H}}'^{(1)}_{\mathrm{d\cdot E}}$, and $\hat{\mathcal{H}}^{(1)}_{\mathrm{d\cdot E}}$ in a basis of $\{|\phi_i\rangle\otimes|n\rangle\}$, where $|\phi_i\rangle$ is the i_{th} electronic state of $\hat{H}_{\mathrm{M}}$, and $|n\rangle$ is the Fock state of $\hat{H}_{\mathrm{ph}}$. The numerical details are provided in Section 4 of Supplement 1. Figure 1(c) displays this quadratic deviation (with respect to the coupling strength) of all of the polariton eigenenergies of the improperly truncated d · E Hamiltonian $\hat{\mathcal{H}}'^{(1)}_{\mathrm{d.E}}$ from the properly truncated Hamiltonians of both $\hat{\mathcal{H}}^{(1)}_{\mathrm{d\cdot E}}$ and $\hat{\mathcal{H}}^{(1)}_{\mathrm{p\cdot A}}$. This departure, however, is almost perfectly constant for the first few polariton energy levels based on our simple analysis of the two-level approximation. When the ground polariton state energy, E_0, is subtracted from all states, the eigenspectrum of $\hat{\mathcal{H}}'^{(1)}_{\mathrm{d\cdot E}}$ almost matches those of $\hat{\mathcal{H}}^{(1)}_{\mathrm{d\cdot E}}$ and $\hat{\mathcal{H}}^{(1)}_{\mathrm{p\cdot A}}$, as shown in Fig. 1(e).

When a permanent dipole is added to the model system (by using $\gamma = 0.2$ for the double-well potential), significant off-diagonal terms appear for the $\hat{\boldsymbol{\mu}}^2$ operator [such as $\langle g|\hat{\boldsymbol{\mu}}(|g\rangle\langle g| + |e\rangle\langle e|)\hat{\boldsymbol{\mu}}|e\rangle$ due to the presence of both transition and the permanent dipoles]. For such a case, even the relative energies of the improperly truncated dipole gauge Hamiltonian $\hat{\mathcal{H}}'^{(1)}_{\mathrm{d\cdot E}}$ start to disagree with the exact results. This asymmetric potential is shown in Fig. 1(b). Figure 1(d) presents the energy eigenspectra for this asymmetric potential. As shown in this panel, the results obtained from $\hat{\mathcal{H}}'^{(1)}_{\mathrm{d\cdot E}}$ no longer have a quadratic departure from the exact results. However, the results obtained from $\hat{\mathcal{H}}^{(1)}_{\mathrm{p\cdot A}}$ and $\hat{\mathcal{H}}^{(1)}_{\mathrm{d\cdot E}}$ follow the results of the full Hamiltonian in Eq. (12) almost perfectly. Figure 1(f) further accentuates the breakdown of $\hat{\mathcal{H}}'^{(1)}_{\mathrm{d\cdot E}}$ in this model. The eigenspectrum produced does not qualitatively match the exact results. We emphasize that for the non-resonant mode ($m = 1$ mode in $\hat{\mathcal{H}}^{(2)}_{\mathrm{d\cdot E}}$), the polariton energy influenced by the dipole self-energy term almost perfectly cancels with that influenced by the light–matter coupling d · E term, such that the energy eigenspectrum is almost identical to the one for $\hat{\mathcal{H}}^{(1)}_{\mathrm{d\cdot E}}$). Using second-order perturbation theory, we provide the analytic expressions of the energy and demonstrate this cancellation explicitly (see Section 2 of Supplement 1).

To further demonstrate these results, we also compute the polaritonic potential energy surfaces of a molecule coupled to a cavity [27], defined as $(\hat{\mathcal{H}}^{(m)} - \hat{T}_{\mathbf{R}})|\Phi(R)\rangle = E(R)|\Phi(R)\rangle$, where the $|\Phi(R)\rangle$ is the polaritonic state for the hybrid system and $E(R)$ is the polariton potential energy surface [2,27]. Here, $\hat{\mathcal{H}}^{(m)} = \hat{\mathcal{H}}^{(2)}_{\mathrm{d\cdot E}}$ for the "exact" result, and $\hat{\mathcal{H}}^{(m)}$ with $\hat{\mathcal{H}}^{(1)}_{\mathrm{p\cdot A}}$ [Eq. (13a)], $\hat{\mathcal{H}}'^{(1)}_{\mathrm{d\cdot E}}$ [Eq. (13b)], and $\hat{\mathcal{H}}^{(1)}_{\mathrm{d\cdot E}}$ [Eq. (13c)] for the mode truncation results. We use the Shin–Metiu model molecular system [28], which contains two fixed ions, and one moving electron and proton (whose position is R), all interacting with each other through modified Coulombic potentials. The details for the model as well as numerical simulations are provided in Sections 3 and 4 of Supplement 1, respectively.

Figure 2 displays the various potential energy surfaces generated by various Hamiltonians, plotted as a function of the proton coordinate, R. The upper panels of this figure also depict the lowest electronic wavefunctions for the isolated matter (without

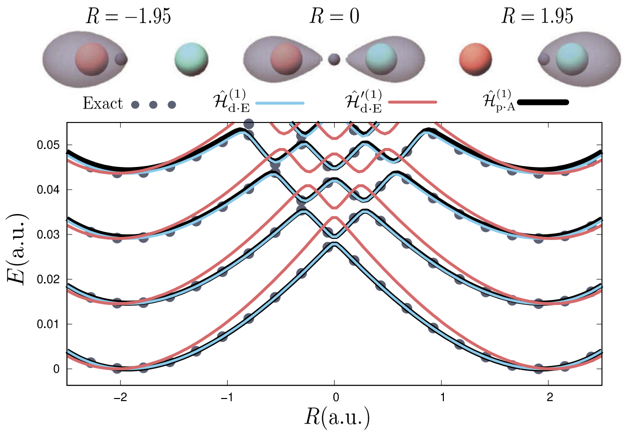

Fig. 2. Polariton potential energy surfaces at different level of theories, for a Shin–Metiu molecular system coupled to a two-modes optical cavity, with $\omega_0 = 0.4$eV and $A_0 = 0.15$. The upper panels visualize the lowest energy wavefunctions at three different proton positions: the donor configuration ($R = -1.95$); the equidistant position ($R = 0$); and the acceptor configuration ($R = 1.95$). "Exact" results refer to the eigenspectrum of $\hat{\mathcal{H}}^{(2)}_{\text{d}\cdot\text{E}}$.

coupling to the cavity) at three different locations of the proton: the donor configuration minimum energy position ($R = -1.95$); the equidistant position ($R = 0$); and the acceptor configuration minimum energy position ($R = 1.95$). The results in Fig. 2 again demonstrate the importance of a proper truncation of the DSE in the dipole gauge Hamiltonian. With a coupling strength of $A_0 = 0.15$, the relative polaritonic energies (i.e., energy differences between states) at a given R is very close to the exact results for $\hat{\mathcal{H}}'^{(1)}_{\text{d}\cdot\text{E}}$. However, to accurately reproduce the exact polaritonic potential energy surface, an R-dependent term would have to be subtracted from the $\hat{\mathcal{H}}'^{(1)}_{\text{d}\cdot\text{E}}$ potential energy surfaces. For this reason, it is especially pertinent to properly truncate the Hamiltonian as done in $\hat{\mathcal{H}}^{(m)}_{\text{d}.\text{E}}$ [Eq. (11)].

In conclusion, we present a new theoretical framework to properly truncate the photonic degrees of freedom in cavity-QED under the long-wavelength approximation. By first truncating the photonic Hamiltonian and matter Hamiltonian in their own Hilbert subspaces, and then performing unitary transformations using a properly projected PZW gauge transformation operator [as outlined in Eqs. (5) and (6)], any gauge ambiguities between the Coulomb and dipole gauges are resolved. This principle allows us to construct a proper procedure for mode truncation without introducing any ambiguities.

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Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

Supplemental document. See Supplement 1 for supporting content.

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