nature physics

ARTICLES

https://doi.org/10.1038/s41567-019-0534-4

Resolution of gauge ambiguities in ultrastrong-coupling cavity quantum electrodynamics

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In quantum electrodynamics, the choice of gauge influences the form of light–matter interactions. However, gauge invariance implies that all physical results should be independent of this formal choice. The Rabi model, a widespread description for the dipolar coupling between a two-level atom and a quantized electromagnetic field, seemingly violates this principle in the presence of ultrastrong light–matter coupling, a regime that is now experimentally accessible in many physical systems. This failure is attributed to the finite-level truncation of the matter system, an approximation that enters the derivation of the Rabi model. Here, we identify the source of gauge violation and provide a general method for the derivation of light–matter Hamiltonians in truncated Hilbert spaces that produces gauge-invariant physical results, even for extreme light–matter interaction regimes. This is achieved by compensating the non-localities introduced in the construction of the effective Hamiltonians. The resulting quantum Rabi Hamiltonian in the Coulomb gauge differs significantly in form from the standard one, but provides the same physical results obtained by using the dipole gauge. These results shed light on gauge invariance in the non-perturbative and extreme-interaction regimes, and solve long-lasting controversies arising from gauge ambiguities in the quantum Rabi and Dicke models.

The ultrastrong coupling (USC) between an effective two-level system (TLS) and the electromagnetic field has been realized in several solid-state systems[1,2]. In this regime of quantum light–matter interaction, going beyond weak and strong coupling, the coupling strength becomes comparable to the transition frequencies of the system. Recently, light–matter coupling strengths larger than the system transition frequencies have been achieved in circuit quantum electrodynamics (QED) experiments involving a single *LC*-oscillator mode coupled to a flux qubit superconducting quantum circuit[3,4]. This extreme interaction regime has been denoted as deep strong coupling (DSC). In these regimes[1,2], several properties of coupled light–matter systems change drastically, opening the way to a wealth of new intriguing physical effects (see, for example, refs. [5–18]), which offer opportunities for the development of new quantum technologies[19–26].

The form of the electron–photon interaction is gauge dependent (see, for example, ref. [27]). However, all physical results must be independent of this choice. Gauge invariance is a general guiding principle in building the theory of fundamental interactions (see, for example, ref. [28]). Let us consider, for example, a particle field whose action is invariant under a global phase change [$U(1)$ invariance]. If this phase is allowed to depend on the space–time coordinate x, its action is not invariant. The symmetry can be restored, replacing the four-momentum derivatives in the action with covariant derivatives: $D_\mu = (\partial_\mu + iqA_\mu)$, where q is the charge parameter and A_μ is the gauge potential.

It has been shown[29–33] that approximate models for light–matter interactions derived in different gauges may lead to different predictions, or can display different convergence properties[34]. When the light–matter interaction becomes very strong, different gauges can lead to drastically different predictions, giving rise to controversies[35–43]. For example, in the case of several TLSs interacting with a single mode of an optical resonator[44], different gauges may even lead to very conflicting predictions, such as the presence or the absence of a quantum phase transition. One important conclusion that can be drawn from these controversies is that, once the light–matter coupling becomes non-perturbative, the validity of the two-level approximation for the atomic dipoles depends explicitly on the choice of gauge[45,46].

In all of these previous studies[35–43,45,46], it is clear that approximations in the description of the matter system (for example, a finite-level truncation) seem to ruin the gauge invariance of the theory. In 1971, it was pointed out[31] that gauge ambiguities in the calculation of atomic oscillator strengths can originate from the occurrence of non-local potentials determined by the approximation procedures. Because a non-local potential in the position representation is an integral operator, it does not commute with the position operator. Indeed, it is easy to show that it can be expressed as a local momentum-dependent operator $V(\mathbf{r}, \hat{\mathbf{p}})$. This affects the interaction of light with quantum systems described by approximate Hamiltonians. Specifically, to introduce the coupling of the matter system with the electromagnetic field, the minimal replacement rule $\hat{\mathbf{p}} \rightarrow \hat{\mathbf{p}} - \hat{\mathbf{A}}(\mathbf{r}, t)$ ($\hat{\mathbf{A}}$ is the vector potential) has to be applied not only to the kinetic energy terms, but also to the non-local potentials in the effective Hamiltonian of the particles in the system. By applying such a procedure, approximate matrix elements for electric dipole transitions[31] and two-photon transition rates, involving Wannier excitons in semiconductors[32], become gauge invariant. Also the microscopic quantum theory of excitonic polaritons is affected by the presence of non-local potentials[47,48].

Here we investigate whether this strategy can work in the maximally truncated Hilbert space provided by a TLS, and in the non-perturbative regimes of cavity QED. This investigation is relevant not only to remove gauge ambiguities in quantum optical systems,

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which are attracting great interest, but also to provide a general insight into gauge invariance in extreme interaction regimes. We find that the usual strategy, which consists of taking into account the non-locality of the atomic potential, performing the minimal coupling replacement and developing the resulting interaction Hamiltonian up to second order in the vector potential, fails when the coupling strength reaches a significant fraction of the resonance frequencies of the system. We demonstrate that these gauge ambiguities can be eliminated for arbitrary coupling strengths only by taking into account the approximation-induced non-locality and keeping the resulting interaction Hamiltonian to all orders in the vector potential $\hat{\mathbf{A}}$. The results presented here solve all the long-lasting controversies arising from gauge ambiguities in the quantum Rabi and Dicke models.

The minimal coupling replacement

We consider a non-relativistic quantum particle of mass m with Hamiltonian $\hat{H}_0 = \hat{\mathbf{p}}^2/(2m) + V(x)$, where $V(x)$ is a local potential. According to the gauge principle, the corresponding gauge-invariant Hamiltonian $\hat{H}_0$ can be expressed as

$$\hat{H}_0 = q\phi + \frac{1}{2m}(\hat{\mathbf{p}} - q\mathbf{A})^2 + V(x) \tag{1}$$

where q is the charge, and $\phi(\mathbf{x})$ and $\mathbf{A}(\mathbf{x})$ are the scalar and vector potentials of the electromagnetic field. Of course, the total energy also has to include the energy of the free field. We observe that, if the particle potential V is non-local, that is, momentum-dependent, the gauge principle, implying the replacement $\hat{\mathbf{p}} \rightarrow \hat{\mathbf{p}} - q\mathbf{A}$, should also be applied to it. In most cases, when dealing with the quantization of the electromagnetic field, it is useful to adopt the Coulomb gauge, where the particle momentum is coupled only to the transverse part of the vector potential. For an effective quantum particle, focusing on a single-cavity mode and considering the electric-dipole approximation, the Hamiltonian in the Coulomb gauge[27] is

$$\hat{H}_C = \frac{1}{2m}(\hat{p} - q\hat{A})^2 + V(x) + \hat{H}_{ph} \tag{2}$$

where $\hat{A} = A_0(\hat{a} + \hat{a}^\dagger)$ is the vector potential calculated at the particle position with a zero-point-fluctuation amplitude A_0, and $\hat{H}_{ph} = \hbar\omega_c \hat{a}^\dagger \hat{a}$ is the cavity-field Hamiltonian. For a multimode resonator: $\hat{A} = A_n(\hat{a}_n + \hat{a}_n^\dagger)$ and $\hat{H}_{ph} = \sum_n \hbar\omega_n \hat{a}_n^\dagger \hat{a}_n$.

If the two lowest energy levels of the effective quantum particle are well separated from the higher energy levels, as in the case of flux qubits[3], and if the detuning $\Delta \equiv \omega_c - \omega_{10}$ (where ω_{10} is the transition frequency of the two lowest energy levels) is much smaller than the detunings of other transitions, the truncation of the Hilbert space to the two lowest energy levels is expected to be a good approximation. Projecting $\hat{H}_C$ in a two-level space, the standard quantum Rabi Hamiltonian in the Coulomb gauge is obtained

$$\hat{\mathcal{H}}_C' = \hat{H}_{ph} + \frac{\hbar\omega_{10}}{2}\hat{\sigma}_z + \hbar g_C \hat{\sigma}_y(\hat{a}^\dagger + \hat{a}) + D(\hat{a}^\dagger + \hat{a})^2 \tag{3}$$

where $g_C = \omega_{10}A_0 d_{10}/\hbar$, $D = q^2A_0^2/(2m)$, and $d_{10} \equiv q\langle 1|\hat{x}|0\rangle$ is the dipole matrix element. Throughout this Article we will use calligraphic symbols as, for example, $\hat{\mathcal{H}}_C'$, to indicate quantum operators in truncated Hilbert spaces. The diamagnetic term $q^2\hat{A}^2/(2m)$ can be absorbed by using a Bogoliubov transformation involving only the photon operators[3,45]. In contrast to the interaction term of first order in the charge, the $\hat{A}^2$ term is not affected by the truncation of the particle Hilbert space. Hence, considering a few-level description of the matter part can result in an over-estimation of the diamagnetic term. Using the Thomas–Reiche–Kuhn sum rule, $\sum_k \hbar\omega_{kj}|d_{jk}|^2 = (\hbar q)^2/(2m)$, the coefficient of the diamagnetic term can be written as $D = A_0^2 \sum_k \omega_{kj}|d_{jk}|^2/\hbar$. When a single transition is considered, this expression can be used to establish a lower bound: $D \geq \hbar g_C^2/\omega_{10}$.

We observe that, in contrast to the Hamiltonian in equation (2), equation (3) violates the gauge principle, because its derivation does not take into account that, in the presence of a truncated Hilbert space, the particle potential loses its locality: $V(x) \rightarrow V'(x, \hat{p})$. We will discuss this problem below, showing the correct procedure for solving it.

The dipole gauge

The Hamiltonian in the dipole gauge, $\hat{H}_D$, corresponds to the Power–Zienau–Woolley Hamiltonian after the dipole approximation[27]. It can be obtained directly from the Hamiltonian in the Coulomb gauge with the electric dipole approximation (2) by means of a gauge transformation, which is also a unitary trasformation: $\hat{H}_D = \hat{U}_1\hat{H}_C\hat{U}_1^\dagger$, where the unitary operator is $\hat{U}_1 = \exp[-iqx\hat{A}/\hbar]$.

The resulting Hamiltonian in the dipole gauge is

$$\hat{H}_D = \hat{H}_{ph} + \hat{H}_0 + \frac{q^2A_0^2\omega_c}{\hbar}x^2 + iq\omega_c x A_0(\hat{a}^\dagger - \hat{a}) \tag{4}$$

Projecting $\hat{H}_D$ to a two-level space, the quantum Rabi Hamiltonian in the dipole gauge is obtained:

$$\hat{\mathcal{H}}_D = \hat{H}_{ph} + \frac{\hbar\omega_{10}}{2}\hat{\sigma}_z + i\hbar g_D(\hat{a}^\dagger - \hat{a})\hat{\sigma}_x \tag{5}$$

where $g_D = \omega_c A_0 d_{10}/\hbar = g_C\omega_c/\omega_{10}$, and $d_{10} \equiv q\langle 1|x|0\rangle$ is the dipole matrix element. In equation (5) we neglected the term $\hat{C} = (A_0^2 d_{10}^2\omega_c/\hbar)\hat{P}$, where $\hat{P} = |0\rangle\langle 0| + |1\rangle\langle 1|$ is the TLS identity operator. This term is obtained by projecting x^2 in the two-dimensional Hilbert space, $\hat{P}x^2\hat{P} = \hat{P}x\hat{P}x\hat{P}$, and using parity symmetry, which implies $\langle n|x|n\rangle = 0$. A more accurate derivation can be carried out including this term in the particle potential before the diagonalization[45] or using perturbation theory. However, we made the choice of considering the interaction terms only after the Hilbert space truncation. In ref. [45] it is shown that, if the two lowest energy levels are well separated from the higher ones ($\omega_{21} \gg g_D$), the two-level approximation provides accurate results even for extreme coupling strengths ($g_D \gg \omega_{10}$).

Revisiting the quantum Rabi model in the Coulomb gauge

As observed above, the derivation of equation (3) does not take into account that, in the presence of a truncated Hilbert space, the particle potential can lose its locality: $V(x) \rightarrow V'(x, \hat{p})$. Thus, to preserve gauge invariance, one has to also apply the substitution $\hat{p} \rightarrow \hat{p} - q\hat{A}$ to the potential. In principle, this procedure can give rise to additional terms in the interaction Hamiltonian to all orders in the vector potential. Although these higher-order terms are expected to be negligible for small normalized couplings $\eta \equiv g_C/\omega_c$, they can become important at higher coupling strengths.

As shown in detail in the Methods, by using some general operator theorems it is possible to apply the minimal coupling replacement to both the kinetic energy and the non-local potential of the effective Hamiltonian of a quantum particle by employing a unitary transformation[47]. In particular, applying equation (20) (see Methods), we obtain

$$\hat{H}_C = \hat{U}\hat{H}_0\hat{U}^\dagger + \hat{H}_{ph} \tag{6}$$

where the unitary operator is $\hat{U}=\hat{U}_1^{\dagger}$, with $\hat{U}_1$ as defined above.

This alternative, although equivalent, minimal-coupling method allows us to understand precisely why the standard quantum Rabi Hamiltonian in the Coulomb gauge $\hat{\mathcal{H}}_{\rm C}^{'}$ violates the gauge principle. $\hat{\mathcal{H}}_{\rm C}^{'}$ can be obtained by applying the minimal coupling replacement to the full matter Hamiltonian, $\hat{H}_0 \rightarrow \hat{U}\hat{H}_0\hat{U}^{\dagger}$, and then projecting in the truncated Hilbert space. Therefore

$$\hat{\mathcal{H}}_{\rm C}^{'}=\hat{P}\hat{U}\hat{H}_0\hat{U}^{\dagger}\hat{P}+\hat{H}_{\rm ph}=\hat{P}\left[\hat{U}\frac{\hat{p}^2}{2m}\hat{U}^{\dagger}+\hat{V}\right]\hat{P}+\hat{H}_{\rm ph} \tag{7}$$

where $\hat{P}$ is the projection operator for the truncated Hilbert space, and we used the relation $\hat{U}\hat{V}\hat{U}^{\dagger}=\hat{V}$, valid when the potential $\hat{V}$ is local. Using Supplementary equation (6), it can easily be shown that equation (7) gives equation (3). Notice that equation (7) contains the non-local potential $\hat{P}\hat{V}\hat{P}$ to which the gauge principle has not been applied. Hence, we can conclude that $\hat{\mathcal{H}}_{\rm C}$ violates the gauge principle. This problem arises whenever the matter system is described within a truncated Hilbert space, and can be solved by first applying to the matter system Hamiltonian (in the absence of interaction) the projection operator, and then the unitary operator, $\hat{P}\hat{H}_0\hat{P}\rightarrow\hat{U}\hat{P}\hat{H}_0\hat{P}\hat{U}^{\dagger}$. Finally, if one desires the resulting Hamiltonian to be within the truncated Hilbert space, it can be done by projecting it at the end. Applying the projection operator and using $\hat{P}^2=\hat{P}$, we obtain

$$\hat{\mathcal{H}}_{\rm C}^{'}=\hat{\mathcal{U}}\hat{\mathcal{H}}_0\hat{\mathcal{U}}^{\dagger}+\hat{H}_{\rm ph} \tag{8}$$

where the projected unitary operator is $\hat{\mathcal{U}}=\hat{P}\hat{U}\hat{P}$ and $\hat{\mathcal{H}}_0=\hat{P}\hat{H}_0\hat{P}$. Equation (8) describes the total light–matter interaction Hamiltonian in the Coulomb gauge and in the electric dipole approximation, satisfying the gauge principle despite the, often unavoidable, truncation of the Hilbert space. We note that $\hat{\mathcal{U}}$ is a unitary operator, in contrast to the operator $\hat{P}\hat{U}$ used in equation (7). This feature is very important because, as we will discuss below, it ensures gauge invariance in truncated Hilbert spaces.

When the matter system is described by a single transition (TLS), we have $\hat{\mathcal{H}}_0=\hbar\omega_{10}\hat{\sigma}_z/2$ and

$$\hat{\mathcal{U}}=\exp\left[i\eta\hat{\sigma}_x(\hat{a}+\hat{a}^{\dagger})\right] \tag{9}$$

where $\eta=g_{\rm D}/\omega_{\rm c}$ is the normalized coupling strength. Therefore, in the Coulomb gauge

$$\hat{\mathcal{H}}_{\rm C}=\hbar\omega_{\rm c}\hat{a}^{\dagger}\hat{a}+\frac{\hbar\omega_{10}}{2}\{\hat{\sigma}_z\cos\left[2\eta(\hat{a}+\hat{a}^{\dagger})\right]+\hat{\sigma}_y\sin\left[2\eta(\hat{a}+\hat{a}^{\dagger})\right]\} \tag{10}$$

is the correct quantum Rabi Hamiltonian. The price one has to pay for preserving the gauge principle in such a truncated space is that the resulting Hamiltonian will contain field operators at all orders. This result shows that the occurrence of a non-local potential, arising from the truncation of the matter system Hilbert space, does not simply modify the dipole moment[31], but profoundly changes the structure of the interaction Hamiltonian. In Supplementary Section I, we show (for the case of TLSs) that, in contrast to equation (3), $\hat{\mathcal{U}}\hat{\mathcal{H}}_0\hat{\mathcal{U}}^{\dagger}$ is able to restore the $U(1)$ symmetry that is broken by coordinate-dependent phase transformations of the matter system wavefunctions.

In addition to the two-level approximation for the matter system, the quantum Rabi model also relies on the single-mode approximation. This further assumption does not result in a breakdown of gauge invariance and it is also largely satisfied for very strong coupling strengths when the electromagnetic resonator is an *LC* circuit[3]. It may fail for other kinds of resonator displaying propagation effects[49–53]. Multimode calculations accounting for the infinite set of cavity modes can lead to divergences unless a cutoff is imposed (see, for example, refs. [50,51]). Recently, it has been shown[50] that finite expressions can be obtained when gauge invariance is respected. The generalization of equation (10) to multimode fields is straightforward. It can be directly obtained from equation (10) by replacing the normalized coupling $\eta(\hat{a}+\hat{a}^{\dagger})$ with $d_{10}\hat{A}/\hbar$, where $\hat{A}=\sum_n A_n(\hat{a}_n+\hat{a}_n^{\dagger})$ is the total vector potential operator at the atom position. Replacing the discrete index n with a proper continuous parameter, this generalization can also be applied to a matter quantum system strongly interacting with a continuum of electromagnetic modes[1,2].

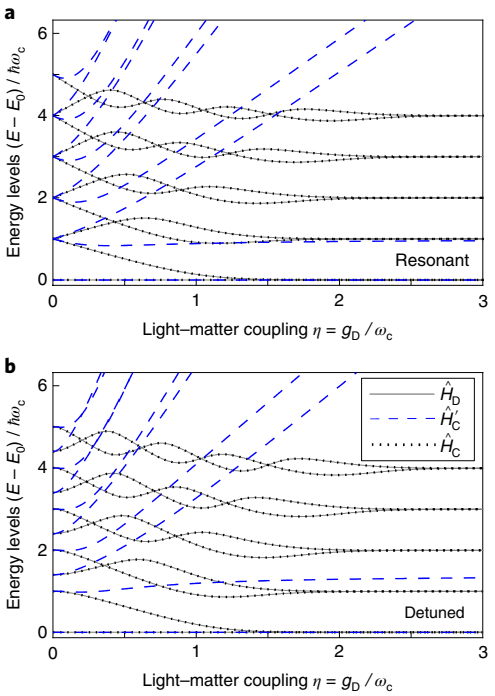

Fig. 1 | Numerical comparisons between different gauges. a,**b**, Comparison of the energy spectra as a function of the normalized coupling $\eta=g_{\rm D}/\omega_{\rm c}$, obtained from the quantum Rabi Hamiltonians in the dipole gauge ($\hat{\mathcal{H}}_{\rm D}$), in the standard Coulomb gauge ($\hat{\mathcal{H}}_{\rm D}^{'}$) and in the Coulomb gauge taking into account the presence of non-local potentials ($\hat{\mathcal{H}}_{\rm C}$): plots for zero detuning ($\Delta=0$) (**a**) and $\Delta=2\omega_{\rm c}/3$ (**b**).

The fulfilment of the gauge principle when considering a TLS interacting with a strong laser (classical) field[54–56] in the Coulomb gauge also requires us to take into account the effect of non-local potentials. In this case, the correct semiclassical Hamiltonian in the Coulomb gauge $\hat{\mathcal{H}}_{\rm C}^{\rm sc}$ is

$$\hat{\mathcal{H}}_{\rm C}^{\rm sc}=\frac{\hbar\omega_{10}}{2}\left\{\hat{\sigma}_z\cos\left[2\frac{d_{10}}{\hbar}A(t)\right]+\hat{\sigma}_y\sin\left[2\frac{d_{10}}{\hbar}A(t)\right]\right\} \tag{11}$$

where $A(t)$ is the classical time-dependent vector potential describing the applied field.

In Fig. 1, we plot the energy differences $((E-E_0)/\hbar\omega_{\rm c})$ for the lowest eigenstates of $H_{\rm D}$ (equation (5)), $\hat{\mathcal{H}}_{\rm C}^{'}$ (equation (3)) and $\hat{\mathcal{H}}_{\rm C}$ (equation (10)), as a function of the normalized coupling $\eta=g_{\rm D}/\omega_{\rm c}$. For $\hat{\mathcal{H}}_{\rm C}^{'}$ we used $D=g_{\rm C}^2/\omega_{10}$; however, the qualitative results do not

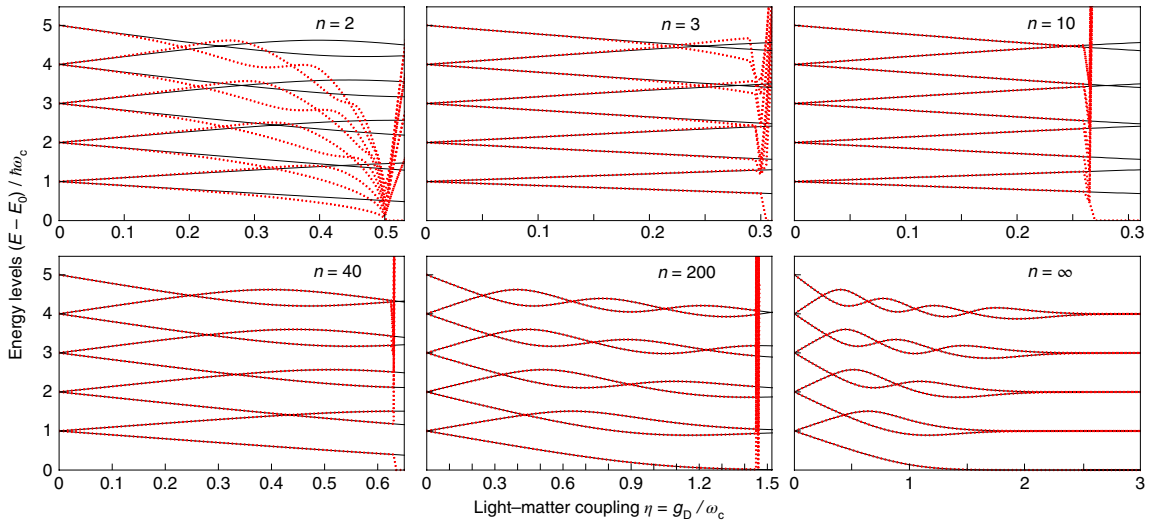

Fig. 2 | Breakdown of gauge invariance. Energy spectra versus normalized coupling η, obtained from the nth-order Taylor expansion $\hat{\mathcal{H}}_{\mathrm{C}}^{(n)}$ of the quantum Rabi Hamiltonian $\hat{\mathcal{H}}_{\mathrm{C}}$, taking into account the presence of non-local potentials. Each panel shows a comparison between the exact spectra, containing all terms (black continuous curves) and the approximated energy levels (red dotted curves) at different orders n of approximation. Note the breakdown of the $\hat{\mathcal{H}}_{\mathrm{C}}^{(n)}$ spectra on the right-hand side of the panels.

change for higher values of D. The comparison in Fig. 1 shows that, for very small values of the coupling, the eigenvalues of the different Hamiltonians reproduce the expected behaviour. However, already at moderate coupling strengths, $\eta \sim 0.1$, there are significant deviations in the predicted energies. For $\eta \gtrsim 0.5$, these differences become drastic. In particular, while the eigenvalues of $\hat{\mathcal{H}}_{\mathrm{D}}$ and $\hat{\mathcal{H}}_{\mathrm{C}}$ do coincide for all the coupling strengths, $\hat{\mathcal{H}}_{\mathrm{C}}'$ provides very different results. This anomalous behaviour of $\hat{\mathcal{H}}_{\mathrm{C}}'$ is a direct consequence of its violation of the gauge principle, demonstrated here. If a Lagrangian or a Hamiltonian does not satisfy the gauge principle, a gauge transformation will produce different physical results. Recently, values of $\eta > 0.1$ have been obtained by several groups[1,2]. In 2017, the record value $\eta = 1.34$ was achieved[3]. The plots in Fig. 1 also enforce the validity of the quantum Rabi Hamiltonian in the dipole gauge (equation (5)), as they show that it provides the same energy levels as the corresponding Hamiltonian in the Coulomb gauge, obtained according to the gauge principle.

The strong differences between the energy levels of $\hat{\mathcal{H}}_{\mathrm{C}}'$ and $\hat{\mathcal{H}}_{\mathrm{D}}$ agree with the results in ref. [45] They also show that, when the matter system displays a strong anharmonicity, the energy levels of $\hat{\mathcal{H}}_{\mathrm{D}}$ (in contrast to those of $\hat{\mathcal{H}}_{\mathrm{C}}$) agree very well with those obtained from the numerically calculated energy levels of the full light–matter Hamiltonian $\hat{H}_{\mathrm{D}}$ for a large range of coupling strengths. Indeed, the two-level approximation is expected to be robust for $\mu \gg \eta$, where $\mu \equiv (\omega_{21} - \omega_{10})/\omega_{10}$ is the anharmonicity. The results of ref. [45] confirm this robustness only for the dipole gauge. Surprisingly, this conclusion seems to be in contrast with the results of ref. [46], where they concluded that the flux gauge (analogous to the dipole gauge for a superconducting artificial atom) provides completely incorrect predictions in most cases. These contradictory results are discussed in Supplementary Section V. Here, we only observe that the degree of anharmonicity of the matter system considered in ref. [46] ($\mu \sim \eta$) is not enough to ensure the validity of the two-level approximation. However, a common feature of refs. [45,46] is that the energy levels of the quantum Rabi model are strongly gauge dependent. In contrast, our derivation of the Rabi Hamiltonian in the Coulomb gauge and the results in Fig. 1 clearly show that, as highly desired, the predictions of the quantum Rabi model are gauge invariant if the gauge principle is correctly applied.

To understand how many powers of the photon operators have to be included in $\hat{\mathcal{H}}_{\mathrm{C}}$ to obtain the correct spectra, in Fig. 2 we compare the approximate spectra, calculated from different n-order Taylor expansions $\hat{\mathcal{H}}_{\mathrm{C}}^{(n)}$ of $\hat{\mathcal{H}}_{\mathrm{C}}$, with the exact ones (the eigenvalues of $\hat{\mathcal{H}}_{\mathrm{C}}$). The results are interesting. For $n = 3$ there is already a significant improvement (with respect to $n = 2$), up to $\eta \lesssim 0.25$. However, the spectra become completely wrong at $\eta \lesssim 0.3$. Accuracy improves for $n = 10$, but only up to $\eta \lesssim 0.25$. For $n = 200$, there is an excellent agreement, but only for $\eta \lesssim 1.3$. These results show that for values of η larger than 1 (DSC), a very large n is needed to obtain the correct spectra. However, further increasing η requires the inclusion of more and more terms in the expansion. This shows that the procedure of taking into account the non-locality of the atomic potential and modifying the interaction Hamiltonian only up to second order in the vector potential[31,32,47,48] completely fails in the USC regime. Figure 2 clearly displays that, for arbitrary coupling strengths, the breakdown of gauge invariance can be avoided only by taking into account the approximation-induced non-locality and keeping the resulting interaction Hamiltonian to all orders in the vector potential. This confirms the non-perturbative spatial non-locality that occurs when heavily truncating the particle's Hilbert space. The results obtained here for a single two-level dipole (Rabi) can be extended to the multi-dipole case (Dicke) [35,43]. Supplementary Section III shows how to obtain the correct Dicke model in the Coulomb gauge. As addressed in ref. [46], gauge ambiguities also arise in circuit-QED systems. For example, the full Hamiltonian of a fluxonium capacitively coupled to an LC oscillator circuit[57] (corresponding to the charge gauge) can be obtained through an analogous minimal coupling replacement. As shown in Supplementary Section IV, the resulting correct total Hamiltonian for the two-level model in the charge gauge is very similar to equation (10).

Resolution of gauge ambiguities

Note that $\hat{H}_{\mathrm{D}}$ and $\hat{H}_{\mathrm{C}}$ are related by a gauge transformation[27,45], which can be expressed by $\hat{H}_{\mathrm{D}} = \hat{U}_1 \hat{H}_{\mathrm{C}} \hat{U}_1^{\dagger}$. As discussed above, $\hat{\mathcal{H}}_{\mathrm{C}}'$ (the standard two-level approximation of $\hat{H}_{\mathrm{C}}$) gives rise to wrong spectra, thus ruining gauge invariance. Instead, we have demonstrated that $\hat{\mathcal{H}}_{\mathrm{C}}$ in equation (10) is the correct quantum Rabi Hamiltonian in the Coulomb gauge. The numerical results in Fig. 1 show that $\hat{\mathcal{H}}_{\mathrm{C}}$

gives the same spectra as $\hat{\mathcal{H}}_{\rm D}$, thus providing clear evidence that the procedure developed here restores gauge invariance in TLSs.

We now present an analytical demonstration of the gauge invariance of a TLS coupled to the electromagnetic field. We start from $\hat{\mathcal{H}}_{\rm D}$, which, according to ref. [45], provides a very good approximation of the full Hamiltonian $\hat{H}_{\rm D}$, and apply the gauge transformation projecting $\hat{U}_1^\dagger = \hat{U}$ in the two-level space, $\hat{U}_1^\dagger \hat{\mathcal{H}}_{\rm D} \hat{U}_1$, where $\hat{U}_1^\dagger = \hat{U}$. The result of this unitary transformation should be $\hat{\mathcal{H}}_{\rm D} \to \hat{\mathcal{H}}_{\rm C}$. Noticing that $\hat{U}$ corresponds to a spin rotation along the x axis, and using the Baker–Campbell–Hausdorff lemma, it is easy to obtain

$$\begin{aligned}\hat{U}_1^\dagger \hat{\mathcal{H}}_{\rm D} \hat{U}_1 &= \hbar\omega_{\rm c}\hat{a}^\dagger\hat{a} + \frac{\hbar\omega_0}{2}\{\hat{\sigma}_z \cos[2\eta(\hat{a}+\hat{a}^\dagger)] \\ &\quad + \hat{\sigma}_y \sin[2\eta(\hat{a}+\hat{a}^\dagger)]\} = \hat{\mathcal{H}}_{\rm C}\end{aligned} \tag{12}$$

This result demonstrates that, if we use $\hat{\mathcal{H}}_{\rm C}$ instead of $\hat{\mathcal{H}}'_{\rm C}$ and apply the gauge transformation consistently, gauge invariance is preserved in a two-level truncated space.

Following ref. [46], it is possible to employ a formulation in which the gauge freedom is contained within a single real continuous parameter α, which determines the gauge through a function operator $\hat{X}_\alpha$. The general gauge transformation in the dipole approximation is generated by a unitary transformation determined by $\hat{U}_\alpha = \exp[-i\hat{X}_\alpha]$, where $\hat{X}_\alpha = \alpha q x \hat{A}/\hbar$. The values $\alpha = \{0, 1\}$ specify the Coulomb and the dipole gauge, respectively. According to the standard procedure (violating the gauge principle), the α-gauge quantum Rabi Hamiltonians can be expressed as[46] $\hat{\mathcal{H}}'^{(\alpha)} = \hat{P}\hat{U}_\alpha \hat{H}_{\rm C} \hat{U}_\alpha^\dagger \hat{P}$. Indeed, following the procedure described above, one finds that the corresponding correct two-level projected unitary operator is $\hat{\mathcal{U}}_\alpha = \exp[-i\hat{\chi}_\alpha]$, where $\hat{\chi}_\alpha = \alpha\eta(\hat{a}+\hat{a}^\dagger)\hat{\sigma}_x$ and the correct α-gauge Hamiltonian for a TLS is thus

$$\hat{\mathcal{H}}^{(\alpha)} = \hat{\mathcal{U}}_\alpha \hat{\mathcal{H}}_{\rm C} \hat{\mathcal{U}}_\alpha^\dagger \tag{13}$$

We obtain

$$\begin{aligned}\hat{\mathcal{H}}^{(\alpha)} &= \hbar\omega_{\rm c}\hat{a}^\dagger\hat{a} - i\alpha g_{\rm D}(\hat{a}-\hat{a}^\dagger)\hat{\sigma}_x \\ &\quad + \frac{\hbar\omega_0}{2}\{\hat{\sigma}_z \cos[2\eta(1-\alpha)(\hat{a}+\hat{a}^\dagger)] \\ &\quad + \hat{\sigma}_y \sin[2\eta(1-\alpha)(\hat{a}+\hat{a}^\dagger)]\}\end{aligned} \tag{14}$$

Because $\hat{\mathcal{U}}_\alpha$ is unitary, the Hamiltonians (14) will have the same energy spectra of $\hat{\mathcal{H}}_{\rm C} = \hat{\mathcal{H}}^{(\alpha=0)}$ and of $\hat{\mathcal{H}}_{\rm D} = \hat{\mathcal{H}}^{(\alpha=1)}$ for any value of α. This eliminates the gauge ambiguities of the quantum Rabi model.

We conclude this subsection with two remarks. First, when calculating expectation values in the various gauges, the unitary transformation in equation (13) also applies to the operators. For example, the photon destruction operator transforms as $\hat{a}_\alpha = \hat{\mathcal{U}}_\alpha \hat{a}_0 \hat{\mathcal{U}}_\alpha^\dagger = \hat{a}_0 + i\alpha\eta\hat{\sigma}_x$, where $\hat{a}_0$ is the photon operator in the Coulomb gauge. Second, different gauges give rise to different eigenstates (all related by unitary transformations), even when using the correct gauge transformations. This feature can lead to some apparent gauge ambiguities when considering time-dependent coupling strengths. In USC systems, the virtual photons in the ground state can be released if the interaction is suddenly switched off [16,53]. Because the different gauges give rise to different eigenstates, the number of emitted photons (proportional to the virtual photon population in the ground state) seems to be gauge dependent. We observe that, during and after the switch off of the interaction, only the $\alpha = 0$ (Coulomb) gauge is well defined. Indeed, in the $\alpha \neq 0$ gauges the field momenta depend on the interaction strength. According to this reasoning, the vacuum emission after the switch-off can be safely described only in the Coulomb gauge. This points out the relevance of obtaining the correct quantum Rabi Hamiltonian in the Coulomb gauge (14) for the design and analysis of these experiments.

Discussion

The method developed here is not limited to TLSs but can be applied to derive gauge-invariant Hamiltonians in arbitrary light–matter quantum systems. These results are also relevant for the study of systems with non-adiabatic time-dependent coupling strength[1,2], as the Coulomb gauge ($\alpha = 0$) is the only one where the field canonical operators are independent of the interaction.

Our results are also relevant for the study of open quantum systems. For example, it turns out that when the interaction of the light and matter components of a quantum system is very strong (USC), the correct gauge dependence of the subsystem operators appearing in the master equation cannot be neglected as usual. Moreover, if the coupling between a subsystem (for example, the matter system) and the environment is described by a gauge interaction and the system–bath coupling strength is not weak, the preservation of the gauge principle should be ensured despite any truncation procedure.

Finally, our investigation also applies to quantum matter systems under the effect of strong laser fields[56] and can be extended to study ultrastrong and deep strong light–matter interactions beyond the dipole approximation[33], where the multipolar gauge[27] is also affected by the presence of non-local potentials.

Online content

Any methods, additional references, Nature Research reporting summaries, source data, statements of code and data availability and associated accession codes are available at https://doi.org/10.1038/s41567-019-0534-4.

Received: 12 October 2018; Accepted: 12 April 2019;
Published online: 3 June 2019

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Acknowledgements

The authors acknowledge discussions with S. De Liberato, A. Nazir and P. Rabl. F.N. is supported in part by the MURI Center for Dynamic Magneto-Optics via the Air Force Office of Scientific Research (AFOSR) (FA9550-14-1-0040), the Army Research Office (ARO) (grant no. W911NF-18-1-0358), the Asian Office of Aerospace Research and Development (AOARD) (grant no. FA2386-18-1-4045), the Japan Science and Technology Agency (JST) (via the Q-LEAP programme and CREST grant no. JPMJCR1676), the Japan Society for the Promotion of Science (JSPS) (JSPS-RFBR grant no. 17-52-50023 and JSPSFWO grant no. VS.059.18N), the RIKEN-AIST Challenge Research Fund and the John Templeton Foundation. S.S. acknowledges support from the Army Research Office (ARO) (grant no. W911NF1910065).

Author contributions

S.S. conceived the main idea and F.N. supervised the work. S.S., O.D. and F.N. designed the study. O.D. and L.G. performed analytical calculations. O.D. and A.S. performed numerical calculations. V.M. and R.S. numerically studied the full Rabi model and the Dicke model. O.D., S.S., L.G. and F.N. contributed to writing the manuscript. All authors were involved in the preparation and discussion of the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

Supplementary information is available for this paper at https://doi.org/10.1038/s41567-019-0534-4.

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Methods

Non-local potentials. To understand why local potentials become non-local when the Hilbert space is truncated, let us consider a 1D potential $\hat{V}$. In the coordinate basis, it can be written as

$$\hat{V} = \iint_{-\infty} \mathrm{d}x\mathrm{d}x' \langle x|\hat{V}|x'\rangle |x\rangle\langle x'| \tag{15}$$

If the potential is local, its matrix elements can be written as $\langle x|\hat{V}|x'\rangle \equiv V(x,x') = W(x)\delta(x-x')$. Considering a complete orthonormal basis $\{|n\rangle\}$, these matrix elements can be expressed as $V(x,x') = W(x)\delta(x-x') = \sum_{n,n'} W_{n,n'}\psi_{n'}^*(x')\psi_n(x)$, where we define $\psi_n(x) \equiv \langle x|n\rangle$. Notice that the Dirac delta function can be reconstructed only by keeping all the infinite vectors of the basis. Hence, any truncation of the complete basis can transform a local potential into a non-local one. If only two states are included, for example, the two lowest energy levels, we obtain

$$V(x,x') = W_{1,0}\left[\psi_0^*(x')\psi_1(x) + \psi_1^*(x')\psi_0(x)\right] \tag{16}$$

where, for simplicity, we assume parity symmetry (which implies that the diagonal matrix elements $W_{n,n}$ are zero) and real matrix elements. It is evident that the sum of the two terms in equation (16), which are products of two smooth wavefunctions, cannot reproduce the Dirac-delta function, and this will result in a potential with a high degree of spatial non-locality. It has been shown by several authors[31–33] that a non-local potential can be expressed as a momentum-dependent operator $V(r,\hat{p})$. Indeed, by using the translation operator $\psi(x') = \exp[i(x'-x)\hat{p}]\psi(x)$, where $\hat{p}$ is the momentum operator, we obtain

$$\int V(x,x')\psi(x')\mathrm{d}x' = V(x,\hat{p})\psi(x) \tag{17}$$

Generalized minimal coupling replacement. In this section, using some general operator theorems[58], we show how to implement the minimal coupling replacement on a generic operator $O(x,\hat{p})$ by performing a unitary transformation[47]. Given two non-commuting operators $\hat{\alpha}$ and $\hat{\beta}$ and a parameter μ we want to calculate $\mathrm{e}^{\mu\hat{\beta}}O(\hat{\alpha})\mathrm{e}^{-\mu\hat{\beta}}$. The function $O(\hat{\alpha})$ can be expanded in a power series:

$$O(\hat{\alpha}) = \sum_n c_n\hat{\alpha}^n \tag{18}$$

Using equation (18), we have

$$\mathrm{e}^{\mu\hat{\beta}}O(\hat{\alpha})\mathrm{e}^{-\mu\hat{\beta}} = \sum_n c_n\mathrm{e}^{\mu\hat{\beta}}\hat{\alpha}^n\mathrm{e}^{-\mu\hat{\beta}} \tag{19}$$

Observing that

$$\mathrm{e}^{\mu\hat{\beta}}\hat{\alpha}^n\mathrm{e}^{-\mu\hat{\beta}} = \mathrm{e}^{\mu\hat{\beta}}\hat{\alpha}\mathrm{e}^{-\mu\hat{\beta}}\mathrm{e}^{\mu\hat{\beta}}\hat{\alpha}\mathrm{e}^{-\mu\hat{\beta}}\cdots\mathrm{e}^{\mu\hat{\beta}}\hat{\alpha}\mathrm{e}^{-\mu\hat{\beta}} = \left(\mathrm{e}^{\mu\hat{\beta}}\hat{\alpha}\mathrm{e}^{-\mu\hat{\beta}}\right)^n$$

we have

$$\mathrm{e}^{\mu\hat{\beta}}O(\hat{\alpha})\mathrm{e}^{-\mu\hat{\beta}} = \sum_n c_n\left(\mathrm{e}^{\mu\hat{\beta}}\hat{\alpha}\mathrm{e}^{-\mu\hat{\beta}}\right)^n = O\left(\mathrm{e}^{\mu\hat{\beta}}\hat{\alpha}\mathrm{e}^{-\mu\hat{\beta}}\right) \tag{20}$$

We now apply equation (20) to $\mathrm{e}^{i\hat{\chi}(x)/\hbar}O(x,\hat{p})\mathrm{e}^{-i\hat{\chi}(x)/\hbar}$. For the sake of simplicity, here we consider the 1D case. The generalization to 3D is straightforward. We obtain

$$\mathrm{e}^{i\hat{\chi}(x)/\hbar}O(x,\hat{p})\mathrm{e}^{-i\hat{\chi}(x)/\hbar} = O\left(x,\mathrm{e}^{i\hat{\chi}(x)/\hbar}\hat{p}\,\mathrm{e}^{-i\hat{\chi}(x)/\hbar}\right) \tag{21}$$

Then, by using the Baker–Campbell–Hausdorff formula, we obtain

$$\mathrm{e}^{i\hat{\chi}(x)/\hbar}\hat{p}\,\mathrm{e}^{-i\hat{\chi}(x)/\hbar} = \hat{p} + \frac{i}{\hbar}[\hat{\chi}(x),\hat{p}] + \frac{1}{2}\left(\frac{i}{\hbar}\right)^2[\hat{\chi}(x),[\hat{\chi}(x),\hat{p}]] + \cdots = \hat{p} - \partial_x\hat{\chi}(x) \tag{22}$$

where we used the result $[\hat{\chi}(x),\hat{p}] = i\hbar\partial_x\hat{\chi}(x)$. In conclusion, using equations (21) and (22), this becomes

$$\mathrm{e}^{i\hat{\chi}(x)/\hbar}O(x,\hat{p})\mathrm{e}^{-i\hat{\chi}(x)/\hbar} = O[x,\hat{p} - \partial_x\hat{\chi}(x)] \tag{23}$$

Considering now the special function

$$\hat{\chi}(x) = q\,x\hat{A}_0 \tag{24}$$

with $\hat{A}_0 \equiv \hat{A}(x_0)$ being the field potential calculated at atom position x_0, we obtain

$$\partial_x\hat{\chi}(x) = q\hat{A}_0 \tag{25}$$

If we plug this result into equation (23), we obtain

$$\mathrm{e}^{i\hat{\chi}(x)/\hbar}O(x,\hat{p})\mathrm{e}^{-i\hat{\chi}(x)/\hbar} = O(x,\hat{p} - q\hat{A}_0) \tag{26}$$

demonstrating that the unitary transformation in equation (23) corresponds to the application of the minimal coupling replacement in the dipole approximation.

Data availability

The data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

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