Cavity Quantum Electrodynamics at Arbitrary Light-Matter Coupling Strengths

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Understanding quantum systems with strong light-matter interaction has become a central problem in both fundamental physics and quantum technologies [1]. Recent experimental and theoretical advances in solid-state physics [2–36], quantum optics [37–68], and quantum chemistry [69–88] have made it possible to achieve strong coupling regimes in a variety of setups. In these systems, standard assumptions such as the rotating wave approximation can no longer be justified, and the inclusion of the diamagnetic term or multilevel structure of matter becomes crucial. Thus, quantized light and matter degrees of freedom must be treated on equal footing within the exact quantum electrodynamics (QED) Hamiltonian. Despite considerable theoretical efforts, a comprehensive formulation for analyzing such challenging problems at arbitrary coupling strengths is still lacking.

On another front, strongly correlated many-body systems have often been tackled by devising a unitary transformation that disentangles certain degrees of freedom, after which a simplified ansatz can be applied: a highly entangled quantum state in the original frame can then be expressed as a factorable state after the transformation. This general idea has been used in several contexts, such as analyzing quantum impurity systems [89–92], constructing low-energy effective models [93–96], and solving many-body localization [97] or electron-phonon problems [98].

The aim of this Letter is to extend this nonperturbative approach to strongly correlated light-matter systems, thus developing a consistent and versatile framework to seamlessly analyze arbitrary coupling regimes. Specifically, we propose to use a unitary transformation that asymptotically decouples light and matter in the strong-coupling limit. Our approach puts no limitations on the coupling strength and allows us to explore the full range of system parameters, including the regime where light-matter coupling dominates over all other relevant energy scales. Importantly, we construct a general way to systematically derive low-energy effective models by faithful level truncations, which remain valid at all coupling strengths. This in particular provides a solution to the long-standing controversy [99–115] about which frame is best suited for studying strong-coupling physics. We demonstrate the versatility of our formalism by applying it to specific models relevant to materials and atomic systems in cavity QED.

Asymptotic decoupling of light-matter interaction.—To illustrate the main idea, we first focus on a one-dimensional many-body system coupled to a single electromagnetic mode; a generalization to higher-dimensional systems with spatially varying electromagnetic modes will be given later. We start from the QED Hamiltonian in the Coulomb gauge:

\[
\hat{H}_C = \int dx \left[ \frac{-(\pi \hbar \partial_x - q \hat{A})^2}{2m} + V(x) \right] \hat{\psi}_x + \hbar \omega_c \hat{a}^\dagger \hat{a} + \hat{H}_\parallel, \tag{1}
\]

FIG. 1. (Top) Effective parameter \( \xi_g \) characterizing interaction strength in the asymptotically decoupled frame against the bare light-matter coupling \( g \). (Bottom) Exact spectrum obtained by diagonalizing Eq. (4), or equivalently (5), for an electron in periodic potential. In the extremely strong coupling (ESC) regime, it exhibits equally spaced flat bands narrowing as \( \propto 1/g^{1/2} \), corresponding to localized electrons with the large renormalized mass (inset). Numerical values are shown in the unit \( \omega_c = \hbar = m = 1 \) throughout this Letter. The potential depth and lattice constant are \( v = 5 \) and \( d = 4 \), respectively.
where $\hat{\psi}_x (\hat{\psi}_x^\dagger)$ is the annihilation (creation) operator of fermions of mass $m$ and charge $q$ at position $x$, and $V(x)$ is an arbitrary external potential. Equation (1) describes the coupling between electrons and a cavity electromagnetic mode with frequency $\omega_c$ and the vector potential operator $\hat{A} = \hat{A}(\hat{a} + \hat{a}^\dagger)$, where $\hat{A}$ is the mode amplitude and $\hat{a} (\hat{a}^\dagger)$ is the annihilation (creation) operator. The instantaneous Coulomb interaction is given by $\hat{H}_C = \int dx \hat{\psi}_x^\dagger (x) \hat{V} (x) \hat{\psi}_x + \hbar \Omega \hat{b}^\dagger \hat{b} + \hat{H}_{\parallel}$

where $\Omega = \sqrt{\omega_c^2 + 2N g^2}$ is the dressed photon frequency with the particle number $N$ and the coupling strength $g = qA/\hbar \omega_c$ is a characteristic length relevant both in weak and strong coupling regimes. Here, the photon part has been diagonalized by a Bogoliubov transformation: $\hat{b}^\dagger + \hat{b} = \sqrt{\Omega/\omega_c} (\hat{a} + \hat{a}^\dagger)$.

To asymptotically decouple light and matter degrees of freedom, we propose to use a unitary transformation

$$\hat{U} = \exp \left[ -i \xi_g \int_{-\infty}^{\infty} dx \hat{\psi}_x^\dagger (x) (i \hbar \partial_x) \hat{\psi}_x \pi \right],$$

where $\xi_g = g x\Omega/\Omega$ is the effective length scale characterized by the coupling strength $g$ and $\pi = i (\hat{b}^\dagger - \hat{b})$. The transformation (3) is reminiscent of the Lee-Low-Pines transformation used for polaronic systems [89], and leads to the Hamiltonian $H_U \equiv \hat{U}^\dagger \hat{H}_C \hat{U}$ given by

$$\hat{H}_U = \int dx \hat{\psi}_x^\dagger \left[ -\frac{\hbar^2 \partial_x^2}{2m} + V (x + \xi_g \pi) \right] \hat{\psi}_x + \hbar \Omega \hat{b}^\dagger \hat{b} + \hat{H}_{\parallel}$$

where the light-matter interaction is now absorbed by the potential term as the shift $\xi_g \pi$ of the electron coordinates. Physically, the unitary operator (3) changes a reference frame in such a way that quantum particles no longer interact with the electromagnetic mode through the usual minimal coupling, but through the gauge-field dependent shift of the electron coordinates and the associated quantum fluctuations in the external potential. Thus, in the transformed frame the effective strength of the light-matter interaction is characterized by $\xi_g$ instead of the original coupling $g$. Remarkably, as shown in the top panel of Fig. 1, $\xi_g$ remains small over the entire region of $g$ and, in particular, vanishes as $\xi_g \propto g^{-1/2}$ in the strong-coupling limit $g \to \infty$. For this reason, we shall call the present frame as the asymptotically decoupled (AD) frame; the identification of the AD Hamiltonian (4) is the first main result of this Letter.

Several remarks are in order. First, a specific form of $\xi_g$ can depend on polarization of an electromagnetic mode. For instance, when matter is coupled to a circularly polarized mode, $\xi_g$ vanishes as $\xi_g \propto g^{-1}$, which is faster than the above linearly polarized case, and the transverse part of the electron wavefunction is modified by the AD transformation [116]. Second, we note that the transformation (3) preserves the translational symmetry of the (bare) matter Hamiltonian. This should be compared to, e.g., the Power-Zienau-Woolley (PZW) frame in which such symmetry is broken due to the additional $x \cdot \vec{E}$ and $x^2$ terms in the transformed Hamiltonian [116–118]. Third, in view of our definition of the coupling strength $g$, the so-called ultrastrong (deep strong) coupling regime should approximately correspond to $g \gtrsim 0.3$ ($g \gtrsim 3$). Below we show that further increase of $g$ leads to the new regime, namely, the extremely strong coupling (ESC) regime. In the latter, truncation of matter/photon levels can no longer be justified in the conventional frames, but is asymptotically exact in the AD frame as discussed in detail below.

**General properties at extremely strong coupling.**—From now on, we focus on the single-electron problems and delineate common properties in the ESC regime; the role of electron interactions in many-electron problems will be discussed in a future publication. The AD-frame Hamiltonian is then simplified to

$$\hat{H}_U = \frac{\hat{p}^2}{2m_{\text{eff}}} + V (x + \xi_g \pi) + \hbar \Omega \hat{b}^\dagger \hat{b},$$

where renormalization of the effective mass $m_{\text{eff}} = m [1 + 2(g/\omega_c)^2]$ arises from the last term in Eq. (4). One can understand the key features of the spectrum of $\hat{H}_U$ in the ESC regime as follows. In the limit of large $g$, the renormalized photon frequency $\Omega$ becomes large, while the effective light-matter coupling, characterized by $\xi_g$, eventually decreases. Thus, in the strong-coupling limit, the lowest-energy eigenstates $|\Psi_U\rangle$ of $\hat{H}_U$ are well approximated by a product state:

$$|\Psi_U\rangle \simeq |\psi_U\rangle |0\rangle_\Omega,$$

where $|\psi_U\rangle$ is an eigenstate of $\hat{p}^2/2m_{\text{eff}} + V(x)$, and $|0\rangle_\Omega$ is the dressed-photon vacuum. Now, suppose that potential $V$ has well-defined local minima, around which it can be expanded as $\delta V \propto x^2$. Since the effective mass rapidly increases as $m_{\text{eff}} \propto g^2$, $|\psi_U\rangle$ is tightly localized around the potential minima. The low-lying spectrum of $\hat{H}_U$ thus reduces to that of the harmonic oscillator with narrowing level spacing $\delta E \propto 1/g$.

The above argument shows that, in the AD frame, an energy eigenstate can be well approximated by a product of light and matter states. Nevertheless, they are still strongly entangled in the original frame. To see this, we consider an eigenstate $|\Psi_C\rangle = \hat{U} |\Psi_U\rangle$ of the Coulomb-gauge Hamiltonian $\hat{H}_C$:

$$|\Psi_C\rangle = \hat{U} \int dp |\psi_p\rangle |0\rangle_\Omega = \int dp |\psi_p\rangle \hat{D}_{\xi g p} |0\rangle_\Omega,$$

where $\int dp |\psi_p\rangle = |\psi_U\rangle$ is the AD-frame eigenstate expressed in the momentum basis, and $\hat{D}_g = e^{i \theta \hat{b}^\dagger - \theta^* \hat{b}}$ is the displacement operator. In the ESC regime, $|\psi_U\rangle$ has vanishingly small width $\sigma_x \propto 1/g$; accordingly, the momentum distribution $|\psi_p|^2$ is very broad with variance $\sigma_p^2 \propto g^2$, showing
that the Coulomb-gauge eigenstate (7) is a highly entangled state consisting of superposition of coherent states with large photon occupancy determined by the particle momentum.

**Difficulties of level truncations in conventional frames.**—
The AD frame readily allows us to elucidate the origin of difficulties for level truncations in the Coulomb gauge [107, 112, 113, 115]. Namely, if we expand a tightly localized state |ψ_U⟩ in terms of eigenstates of \( \hat{p}^2/2m + V \) with the bare mass \( m \), we will find substantial contribution from high-energyelectron states. This can be seen from Eq. (7), which contains large-momentum eigenstates. This argument shows that any analysis performed in the Coulomb gauge, which uses a fixed UV cutoff for electron states, should become invalid at sufficiently strong coupling. The same applies to photons. As seen from Eq. (7), both the mean and fluctuation of the photon number in |Ψ_C⟩ increases as \( \delta n \propto g \). Thus, the number of photon states required to diagonalize \( \hat{H}_C \) diverges at large \( g \), making photon-level truncation (that is unavoidable in actual calculation) ill-justified in the ultra- or extremely-strong coupling regimes.

While the use of the PZW frame can partially mitigate the limitations on level truncations, it is ultimately constrained by the same restrictions, especially in the ESC regime. As we demonstrate below, this holds true even when high-lying states appear to be reasonably out of resonance. Altogether, as long as one relies on the conventional frames, we conclude that effective models derived by level truncations, such as tight-binding models or the quantum Rabi model, must inevitably break down when \( g \) becomes sufficiently large.

In contrast, the AD frame (5) introduced here provides a simple solution to this problem. Specifically, matter-level truncation, i.e., tight-binding approximation, is increasingly well-justified in \( H_U \) at larger \( g \), owing to tighter localization of the wavefunction |ψ_U⟩. Similarly, due to the photon dressing and asymptotic decoupling, one can always truncate high-lying photon levels because the mean photon number remains very small over the entire region of \( g \) and, in particular, vanishes in the ESC limit. Below we demonstrate such versatility of the AD frame by applying it to concrete models relevant to quantum electrodynamical materials and atomic dipoles.

**Application to solid-state systems.**— We first consider an electron in periodic potential and discuss the formation of electron-polariton band structures. To be concrete, we assume \( V = v[1 + \cos(2\pi x/d)] \) with \( d \) and \( v \) being the lattice constant and potential depth, respectively. Since the AD frame preserves the translational symmetry, Bloch’s theorem remains valid and every eigenvalue of \( H_U \) has a well-defined crystal wavevector \( k \in [-\pi/d, \pi/d] \). Figures 1 and 2a-d show the obtained exact eigenspectra at different coupling strengths \( g \), in the sense that matter/photon-energy cutoffs are taken to be large enough such that the results are converged. As \( g \) is increased, the bands become increasingly flat and form equally spaced spectra with energy spacing narrowing \( \propto 1/g \), which is fully in accord with the general properties discussed earlier.

To construct the effective low-energy Hamiltonian, we derive the tight-binding model by projecting the continuum system on the lowest-band Wannier orbitals. Specifically, we first expand a matter state in terms of the Wannier basis,

\[
\hat{\psi}_x = \sum_j w_j(x) \hat{c}_j,
\]

where \( w_j \) is the Wannier function at site \( j \) for the lowest band of \( \hat{p}^2/2m_{\text{eff}} + V \) with the effective mass, and \( \hat{c}_j \) is the corresponding annihilation operator. We then consider a manifold spanned by product states of these Wannier orbitals and an arbitrary photon state. Projecting \( \hat{H}_U \) on this manifold and considering the leading contributions, we obtain the tight-binding Hamiltonian in the AD frame as [116]

\[
\hat{H}_U^{\text{TB}} = \left( t_g + t_g' \delta_g \right) \sum_i (\hat{c}_i^\dagger \hat{c}_{i+1} + \text{h.c.}) + \left( \mu_g + \mu_g' \delta_g \right) \sum_i \hat{c}_i^\dagger \hat{c}_i + \hbar \Omega b^\dagger b,
\]

where \( t_g = \int \frac{dk}{K} \epsilon_{k,g} e^{ikd} \) is the effective hopping parameter with \( \epsilon_{k,g} \) being the lowest-band energy of \( \hat{p}^2/2m_{\text{eff}} + V \) and \( K = 2\pi/d \), and \( \mu_g = \int \frac{dk}{K} \epsilon_{k,g} \) is the effective chemical potential. The electromagnetically induced fluctuation of potential causes the terms with \( \delta_g = \cos(\tilde{K} \xi_g \tilde{r}) - 1 \), \( t_g' = v \int dx w_i^* \cos(Kx) w_{i+1} \), and \( \mu_g' = v \int dx w_i^* \cos(Kx) w_i \).

Figure 2e shows that this surprisingly simple tight-binding model (black dashed curve) accurately predicts the exact spectrum (solid curve) at any \( g \). In particular, it asymptotically becomes exact in the strong-coupling limit as expected. For the sake of comparison, we also show the tight-binding results in the Coulomb gauge (red dotted curve), which are obtained by projecting \( \hat{H}_C \) onto the lowest band of \( \hat{p}^2/2m + V \) with the bare mass [116]. While this naive tight-binding model is valid when the coupling is weak \( g \approx 0.1 \), it completely misses key features at larger \( g \), such as band flattening and narrowing. Physically, this drastic failure originates from ill-justified truncation of strongly entangled high-lying light-matter states in the original frame [cf. Eq. (7)].
These results clearly demonstrate that a choice of the frame is essential to construct an accurate tight-binding model in strong-coupling regimes. The AD frame solves this issue by performing the projection after the unitary transformation, which effectively realizes suitable nonlinear truncation in the Coulomb gauge. In general, the AD-frame tight-binding Hamiltonian for arbitrary periodic potential and internal degrees of freedom is given by

$$\hat{H}_U^{\text{TB}} = \sum_{ij \nu \lambda} t_{ij \nu \lambda} \hat{c}^\dagger_{ij \nu} \hat{c}_{ij \nu} + \sum_{ij \nu \lambda} t^{(l)}_{ij \nu \lambda} \hat{c}^\dagger_{ij \nu} \hat{c}_{ij \nu} + \hbar \Omega \hat{b}^\dagger \hat{b}, \quad (10)$$

where $\nu (\lambda)$ labels internal degrees of freedom in each unit cell $i (j)$, and $t_{ij \nu \lambda} = \int dx w^{*}_{\nu \lambda} [\hat{p}^2/2m_{\text{eff}} + V] w_{\nu \lambda}$, $t^{(l)}_{ij \nu \lambda} = \frac{\partial}{\partial x} \int dx w^{*}_{\nu \lambda} V^{(l)}(x) w_{\nu \lambda}$ with $w_{\nu \lambda}$ being the Wannier orbitals for the lowest isolated composite bands, and $V^{(l)}$ is the $l$-th derivative of $V$ with $l = 1, 2, \ldots$. The renormalized parameters $t, t^{(l)}$ nonperturbatively depend on $g$ through the nonlinear truncation. Higher-order terms with larger $l$ contribute less to eigenspectrum owing to smallness of $\xi_{\omega}$ (cf. Fig. 1), which enables a systematic approximation when necessary. The derivation of the minimal tight-binding Hamiltonian (10), which is valid at arbitrary coupling strengths and provides the material counterpart of the quantum Rabi model, is the second main result of this Letter.

**Application to atomic dipoles.** We next apply the AD frame to the case of a quantum particle in double-well potential $V = -\lambda x^2/2 + \mu x^4/4$, which is a standard model for the electrical dipole moment. Blue solid curves in Fig. 3a,b show the exact spectra obtained in the AD frame at different potential depths, where the results efficiently converge already at a low photon-number cutoff $n_c \approx 5-10$ [116]. The spectra at ESC exhibit the common features discussed above, i.e., energies become doubly degenerate corresponding to two wells and are equally spaced with narrowing $\propto 1/g$ due to tight localization around the minima (cf. insets).

As discussed earlier, truncation of high-lying photon states should eventually be invalid in conventional frames. We demonstrate this by comparing to results obtained in the PZW frame, $\hat{H}_{PZW} = \hat{U}_{PZW} \hat{H}_C \hat{U}_{PZW}$ with $\hat{U}_{PZW} = \exp(i qx \hat{A}/\hbar)$, at a large cutoff $n_c = 100$ (red dotted curves in Fig. 3). Notably, the PZW frame dramatically fails in the ESC regime, which has its root in the rapid increase of mean-photon number due to strong light-matter entanglement and sizable probability amplitudes of high-photon-number states [116]. We remark that matter-level cutoff is taken to be sufficiently large such that the results converge because the strong light-matter entanglement also invalidates matter-level truncation, leading, for instance, to the Rabi-model descriptions. Since any actual calculation must resort to finite cutoffs, these results indicate the fundamental difficulties of the conventional frames in the ESC regime.

**Beyond the single-mode description.** While the single-mode description can be justified in, e.g., an LC-circuit resonator [47], it may fail when more than one cavity mode must be included depending on the cavity geometry. The unitary transformation (3) can be generalized to such a case with spatially varying electromagnetic modes:

$$\hat{U} = \exp \left[ -i \frac{\hat{p}}{\hbar} \cdot \sum_{\alpha} \xi_{\alpha} \hat{\pi}_{\alpha}(x) \right], \quad (11)$$

where $\alpha$ labels multiple modes and the electromagnetic fields now depend on position $x$. At the leading order, the transformed Hamiltonian is

$$\hat{H}_U = \frac{\hat{p}^2}{2m} - \sum_{\alpha} \left( \frac{\hat{p} \cdot \xi_{\alpha}}{\Omega_{\alpha}} \right)^2 + V(x) + \sum_{\alpha} \hbar \Omega_{\alpha} \hat{b}^\dagger(x) \hat{b}_{\alpha}(x), \quad (12)$$

where $\xi_{\alpha}$ is the effective polarization vector of mode $\alpha$ [116]. This simple expression is valid when field variation is small compared to the effective length scale, i.e., $|k\xi| \ll 1$ with $|\nabla \hat{b}| \sim k \hat{b}$. We emphasize that this is different from the dipole approximation, which in general requires fields to vary little over system size. Our condition is independent of system size and is much less restrictive owing to smallness of $\xi_{\omega}$.

**Discussions.** In the limit of classical electromagnetic fields, our transformation (3) can be compared with the Kramers-Henneberger (KH) transformation, which was used to analyze atoms subject to intense laser fields [119, 120]. Besides the full quantum treatment given in our formalism, one important difference is that the KH transformation does not take into account the diamagnetic $A^2$ term other than its contribution to ponderomotive forces appearing in spatially inhomogeneous laser profiles. In our quantum setting, the asymptotic light-matter decoupling emerges only after the diamagnetic term is consistently included through the Bogoliubov transformation.

With the advent of new materials and subwavelength cavity designs, it is now possible to explore ultrastrong coupling regimes of light-matter interaction and possibilities for further
extending the interaction strength. We expect our results to be applicable in the analysis of mono- or (twisted) bilayer-2D materials embedded in high quality-factor lumped-element terahertz cavities [16], where a single mode of the electromagnetic field is isolated from higher-energy Fabry-Perot-like confined modes, as well as the electromagnetic continuum.

In summary, we presented a new formulation (4) of strongly correlated light-matter systems that is applicable to both quantum electrodynamical materials and atomic systems. Since this is a nonperturbative approach, it is valid at arbitrary coupling strengths and, in particular, allows us to consistently explore the extremely strong coupling regime for the first time. Our formalism elucidates difficulties of level truncations in the conventional frames from a general perspective, and offers a systematic way to derive the faithful tight-binding Hamiltonians (10). While the emphasis was placed on the extremely strong coupling, our formalism is versatile enough to be applied to any coupling regimes, where standard/conventional descriptions can be inadequate. It would be interesting to apply the present formulation to identify the correct tight-binding models of more complex light-matter systems. In particular, it merits further study to elucidate role of the light-induced band flattening and narrowing in genuine many-body regimes.


[34] Y. Ashida, A. Imamoglu, J. Faist, D. Jaksh, A. Cavalleri, and...
[116] See Supplemental Materials for further details on the statements and the derivations.
Supplementary Materials

Polarization dependence of the effective length scale

We here mention that the effective length scale $\xi_g$, which characterizes the light-matter interaction strength in the asymptotically decoupled (AD) frame, in general depends on a polarization of an electromagnetic mode coupled to a many-body system. To demonstrate this, we consider a two-dimensional many-body system coupled to a circularly polarized electromagnetic mode as an illustrative example:

$$\hat{A} = A \left(e\hat{a} + e^*\hat{a}^\dagger\right), \quad e = \frac{1}{\sqrt{2}} \begin{bmatrix} 1 \\ i \end{bmatrix}. \quad (S1)$$

In this case, there are no terms that are proportional to $\hat{a}\hat{a}$ or $\hat{a}^\dagger\hat{a}^\dagger$ in the $\hat{A}^2$ term; this diamagnetic term simply renormalizes the photon frequency without performing the Bogoliubov transformation. Thus, the resulting light-matter Hamiltonian in the Coulomb gauge is given by

$$\hat{H}_C = \int dx \tilde{\psi}_x^\dagger \left(-\frac{\hbar^2 \nabla^2}{2m} + V(x)\right) \tilde{\psi}_x - gx\omega_c \int dx \tilde{\psi}_x^\dagger (-i\hbar \nabla)\tilde{\psi}_x \cdot (e\hat{a} + e^*\hat{a}^\dagger) + \hbar \Omega \hat{a}^\dagger \hat{a}, \quad (S2)$$

where $g = qA\sqrt{\omega_c/m\hbar}$ and we introduce

$$x\omega_c = \sqrt{\frac{\hbar}{m\omega_c}}, \quad \Omega = \omega_c \left(1 + \frac{N \sqrt{g^2}}{\omega_c^2}\right). \quad (S3)$$

Note that the renormalized photon frequency depends on $g$ in a different way from the linearly polarized case discussed in the main text, for which $\Omega = \sqrt{\omega_c^2 + 2Ng^2}$.

To asymptotically decouple the light and matter degrees of freedom, we can use a unitary transformation

$$\hat{U} = \exp \left[-i\xi_g \int dx \tilde{\psi}_x^\dagger (-i\nabla)\tilde{\psi}_x \cdot \hat{\pi}\right], \quad \hat{\pi} = i \left(e^*\hat{a}^\dagger - e\hat{a}\right). \quad (S4)$$

where we define the effective length scale $\xi_g$ by

$$\xi_g = \frac{gx\omega_c}{\Omega} = x_{\omega_c} \frac{g/\omega_c}{1 + N \sqrt{g^2}/\omega_c^2}. \quad (S5)$$

This length scale for a circularly polarized case asymptotically vanishes in the strong-coupling limit with the scaling $\propto 1/g$, which is faster than the linearly polarized case $\propto 1/g^{1/2}$ (see Fig. S1). For the sake of completeness, we also show the full expression of the transformed Hamiltonian $\hat{H}_U = \hat{U}^\dagger \hat{H}_C \hat{U}$ in the present case:

$$\hat{H}_U = \int dx \tilde{\psi}_x^\dagger \left[-\frac{\hbar^2 \nabla^2}{2m} + V(x + \xi_g \hat{\pi})\right] \tilde{\psi}_x + \hbar \Omega \hat{a}^\dagger \hat{a} - \hbar^2 \frac{g^2}{m\Omega^2} \int dx \tilde{\psi}_x^\dagger (-i\nabla)\tilde{\psi}_x \left\{1 + \frac{\xi_g}{2\sqrt{\omega_c}} \int dx \tilde{\psi}_x^\dagger (-i\nabla)\tilde{\psi}_x\right\}^2 + \int dx dx' \frac{q^2 \tilde{\psi}_x^\dagger \tilde{\psi}_x^\dagger \tilde{\psi}_{x'}^\dagger \tilde{\psi}_{x'}^\dagger \tilde{\psi}_{x'} \tilde{\psi}_x}{4\pi \epsilon_0 |x - x'|}. \quad (S6)$$

FIG. S1. Effective length scale $\xi_g$ for a circularly polarized light (black solid curve) and a linearly polarized light (red dashed curve) against the bare light-matter coupling $g$. This length characterizes the effective interaction strength in the asymptotically decoupled frame. We set $\omega_c = \hbar = m = 1$. 
Derivation of the tight-binding models

We here provide technical details about the derivation of the tight-binding models in the AD frame and the Coulomb gauge. We consider an electron that is subject to the periodic potential \( V(x) = v[1 + \cos(2\pi x/d)] \) and coupled to an electromagnetic mode. The total Hamiltonian in the AD frame is given by [cf. Eq. (5) in the main text]

\[
\hat{H}_U = \frac{\hat{p}^2}{2m_{\text{eff}}} + v[1 + \cos(Kx)\cos(K\xi_g\hat{\pi}) - \sin(Kx)\sin(K\xi_g\hat{\pi})] + \hbar\Omega\hat{b}\hat{b}^\dagger, \tag{S7}
\]

where \( \hat{\pi} = i(\hat{b}^\dagger - \hat{b}) \) and \( K = \frac{2\pi}{d} \). To derive the effective low-energy Hamiltonian, we consider the lowest-band Bloch wavefunctions for the single-particle Hamiltonian with the effective mass \( m_{\text{eff}} \):

\[
\left[ \frac{\hat{p}^2}{2m_{\text{eff}}} + V(x) \right] \psi_k = \epsilon_{k,g}\psi_k, \tag{S8}
\]

where \( \psi_k(x) = e^{ikx}u(x) \) with \( u(x) = u(x + d) \). Here, we note the \( g \) dependence of the dispersion \( \epsilon_{k,g} \), which comes through the effective mass \( m_{\text{eff}} = m[1 + 2(g/\omega_c)^2] \). The corresponding Wannier function is

\[
w_j(x) = \int \frac{dk}{K} e^{-ikjd}\psi_k(x). \tag{S9}
\]

We now introduce a manifold of light-matter wavefunction spanned by product states consisting of these Wannier orbitals and an electromagnetic mode:

\[
|\Psi_j\rangle = \int dx w_j(x)|x\rangle \otimes |\psi_{\text{photon}}\rangle, \tag{S10}
\]

where \( |\psi_{\text{photon}}\rangle \) is an arbitrary photon state. When we consider the projection of \( \hat{H}_U \) onto this manifold, the contribution from the term proportional to \( \sin(Kx) \) in Eq. (S7) vanishes. This is because the Hamiltonian \( \hat{H}_U \) has the parity symmetry under \( x \rightarrow -x \) and \( \hat{\pi} \rightarrow -\hat{\pi} \) and the lowest states reside in the even parity sector. Since the lowest-band Wannier state \( u(x) \) respects the even parity symmetry, this fact indicates that the photon wavefunction is also symmetric against \( \hat{\pi} \rightarrow -\hat{\pi} \), leading to \( \langle \sin(K\xi_g\hat{\pi}) \rangle = 0 \). Thus, at the leading order, the projection results in the effective Hamiltonian

\[
\langle \Psi_j | \hat{H}_U | \Psi_i \rangle = \langle \Psi_j | \frac{\hat{p}^2}{2m_{\text{eff}}} + V(x) + v \cos(Kx) \left[ \cos(K\xi_g\hat{\pi}) - 1 \right] + \hbar\Omega\hat{b}\hat{b}^\dagger | \Psi_i \rangle \\
\simeq t_g (\delta_{i,j+1} + \delta_{i,j-1}) + \mu_g \delta_{i,j} + \left[ t'_g (\delta_{i,j+1} + \delta_{i,j-1}) + \mu'_g \delta_{i,j} \right] \delta_{g} + \hbar\Omega\hat{b}\hat{b}^\dagger, \tag{S11}
\]

where we introduce the renormalized tight-binding parameters depending on \( g \) as

\[
t_g = \int \frac{dk}{K} \epsilon_{k,g} e^{ikd} \in \mathbb{R}, \quad \mu_g = \int \frac{dk}{K} \epsilon_{k,g} \tag{S12}
\]

\[
t'_g = v \int dx w_{i-1}^* \cos(Kx)w_i \in \mathbb{R}, \quad \mu'_g = v \int dx w_i^* \cos(Kx)w_i, \tag{S13}
\]

and the operator describing the electromagnetically induced fluctuation by

\[
\delta_g = \cos(K\xi_g\hat{\pi}) - 1. \tag{S14}
\]

After transforming to the second quantization notation, we obtain the tight-binding Hamiltonian in the AD frame, which provides Eq. (9) in the main text

\[
\hat{H}_U^{\text{TB}} = \left( t_g + t'_g \delta_g \right) \sum_i \left( \hat{c}_i^\dagger \hat{c}_{i+1} + \text{h.c.} \right) + \left( \mu_g + \mu'_g \delta_g \right) \sum_i \hat{c}_i^\dagger \hat{c}_i + \hbar\Omega\hat{b}\hat{b}^\dagger, \tag{S15}
\]

where the annihilation operator should be understood in terms of the expansion \( \hat{\psi}_x = \sum_j w_j(x) \hat{c}_j \). We remark that, while this tight-binding description is valid when low-energy equilibrium properties are of interest, one may have to include further correction terms for analyzing nonequilibrium dynamics. For instance, the contribution from the \( \sin(Kx) \) term in Eq. (S7) can be relevant when excitations to higher bands are nonnegligible.
For the sake of comparison, we next explain the construction of the tight-binding Hamiltonian in the Coulomb gauge. We
start from the Coulomb-gauge Hamiltonian
\[ \hat{H}_C = \frac{\hat{p}^2}{2m} + V(x) - gx\Omega \hat{p}(\hat{b} + \hat{b}^\dagger) + \hbar\Omega \hat{b}^\dagger \hat{b}. \] (S16)

Similar to the above discussion, we consider the lowest-band Wannier states for the single-particle Hamiltonian with the \textit{bare} mass \( m \):
\[ \left[ \frac{\hat{p}^2}{2m} + V(x) \right] \hat{\psi}_k = \hat{\epsilon}_k \hat{\psi}_k, \quad \hat{w}_j(x) = \int \frac{dk}{K} e^{-ikjd} \hat{\psi}_k(x), \] (S17)
and consider a manifold spanned by the following light-matter states
\[ |\tilde{\Psi}_j\rangle = \int dx \hat{w}_j(x) |x\rangle \otimes |\text{photon}\rangle. \] (S18)

We note that the dispersion \( \hat{\epsilon}_k \) is independent of \( g \) as we here consider the bare mass \( m \). The projection of \( \hat{H}_C \) onto this manifold results in the tight-binding Hamiltonian
\[ \langle \tilde{\Psi}_j | \hat{H}_C | \tilde{\Psi}_i \rangle = \langle \tilde{\Psi}_j \rangle \frac{\hat{p}^2}{2m} + V(x) - gx\Omega \hat{p}(\hat{b} + \hat{b}^\dagger) + \hbar\Omega \hat{b}^\dagger \hat{b} \rangle |\tilde{\Psi}_i\rangle \]
\[ \approx \hat{\epsilon}_i (\delta_{i,j+1} + \delta_{i,j-1}) + \hat{\mu}_i \delta_{i,j} - \left( \lambda_y \delta_{i,j+1} + \lambda_y^* \delta_{i,j-1} \right) (\hat{b} + \hat{b}^\dagger) + \hbar\Omega \hat{b}^\dagger \hat{b}, \] (S19)
where the tight-binding parameters are defined by
\[ \hat{i} = \int \frac{dk}{K} \hat{\epsilon}_k e^{ikd} \in \mathbb{R}, \quad \hat{\mu} = \int \frac{dk}{K} \hat{\epsilon}_k, \quad \lambda_y = \hbar gx\Omega \int dx \hat{w}_i^* \hat{w}_i (i\partial_x) \hat{w}_i \in i\mathbb{R}. \] (S20)

We again emphasize that, in contrast to the AD-frame case above, the tight-binding parameters are defined in terms of the single-particle states with the bare mass \( m \); thus, in particular, \( \hat{i}, \hat{\mu} \) are independent of the light-matter coupling \( g \).

In the second quantization notation, the tight-binding Hamiltonian can be written as
\[ \hat{H}_C^{\text{TB}} = \sum_i \left( \hat{i} - \lambda_y \left( \hat{b} + \hat{b}^\dagger \right) \right) \hat{c}_i^\dagger \hat{c}_{i+1} + \text{h.c.} + \hat{\mu} \sum_i \hat{c}_i^\dagger \hat{c}_i + \hbar\Omega \hat{b}^\dagger \hat{b}, \] (S21)
where the annihilation operator is defined in terms of the Wannier function with the bare mass, \( \hat{\psi}_x = \sum_j \hat{w}_j(x) \hat{c}_j \). Its eigen-spectrum can analytically be given by
\[ \hat{\epsilon}_k^{\text{TB}} |_{\text{Wannier}} = 2\hat{\epsilon}_i \cos (kd) + \hat{\mu} - \frac{\lambda_y^2}{\Omega} \sin^2 (kd) + \hbar\Omega n, \] (S22)
where \( n = 0, 1, 2 \ldots \). The results plotted in Fig. 2 in the main text correspond to the \( n = 0 \) sector of this dispersion. It is evident from Eq. (S22) that the tight-binding spectrum in the Coulomb gauge is completely independent of \( g \) at \( k = 0, \pm \pi/d \), which clearly indicates difficulties of level truncations in the Coulomb gauge.

Finally, we remark that the one-dimensional tight-binding model acquires additional contributions in the case of the circularly polarized light. To see this, it is sufficient to consider the following single-particle continuum model (see Eq. (S3) for the definitions of the microscopic parameters):
\[ \hat{H}_C = \frac{\hat{p}^2}{2m} + V(x) + \frac{m\Omega_y^2}{2} \left( e\hat{a} + e^* \hat{a}^\dagger \right) + \hbar\Omega \hat{a}^\dagger \hat{a}, \] (S23)
where \( e = \frac{1}{\sqrt{2}} [1, i]^T \) is the polarization vector, and the electron is tightly localized in the transverse \( y \) direction via the potential \( m\Omega_y^2 \Omega^2 / 2 \), while it is subject to the periodic potential \( V(x) \) in the \( x \) direction. Using the unitary transformation (S4), we obtain
\[ \hat{H}_U = \frac{\hat{p}^2}{2m_{\text{eff}}} + V\left( x + i\xi_y (\hat{a}^\dagger - \hat{a}) / \sqrt{2} \right) + \frac{m\Omega_y^2}{2} \left[ y + \xi_y (\hat{a}^\dagger + \hat{a}) / \sqrt{2} \right]^2 + \hbar\Omega \hat{a}^\dagger \hat{a}, \] (S24)
where \( m_{\text{eff}} = m[1 + (g/\omega_c)^2] \). It is now clear that, even when we are interested in 1D electron dynamics in the \( x \) direction and aim to derive the tight-binding model along this direction, we must consistently take into account the contribution from the third term in the RHS of Eq. (S24) that arises from the coupling between the transverse motion and the circularly polarized light.
Photon-number cutoff dependence of the low-energy spectra

We here briefly mention the photon-number cutoff dependence of the low-energy spectra in different frames. We compare the spectra for the double-well potential $V = -\lambda x^2/2 + \mu x^4/4$ in the AD frame $H_U = \hat{U} \hat{H}_C \hat{U}^\dagger$ with $\hat{U} = \exp(-i\xi_0 \hat{p} \hat{x}/\hbar)$ and the Power-Zienau-Woolley (PZW) frame $\hat{H}_{VZW} = \hat{U}_{PZW}^\dagger \hat{H}_C \hat{U}_{PZW}$ with $\hat{U}_{PZW} = \exp(i q x \hat{A}/\hbar)$:

$$\hat{H}_U = \frac{\hat{p}^2}{2m} + V(x + \xi_0 \hat{p}) + \hbar \Omega \hat{b}^\dagger \hat{b},$$

$$\hat{H}_{VZW} = \frac{\hat{p}^2}{2m} + V(x) + mg^2 x^2 + i g \sqrt{m \hbar \omega_c} (\hat{a}^\dagger - \hat{a}) + \hbar \omega_c \hat{a}^\dagger \hat{a}. \quad (S25)$$

In Fig. 3 in the main text, we show the results at the large photon-number cutoff $n_c = 100$, and demonstrate that the PZW frame fails to capture the key features in the extremely strong coupling (ESC) regime, such as the level degeneracy and narrowing. This is caused by the slower convergence of the PZW results at larger coupling $g$ with respect to the photon-number cutoff $n_c$. To see this explicitly, we plot the low-lying energies (subtracted by the lowest eigenvalue) in different frames in Fig. S2. While the results in the AD frame efficiently converge already for low cutoff $n_c \sim 5 - 10$ at any coupling strength $g$, the convergence in the PZW frame becomes worse as $g$ is increased. In particular, in the ESC regime (roughly corresponding to $g/\omega_c \gtrsim 10$), the PZW results typically fail to converge within a tractable value of the photon-number cutoff. This difficulty stems from the rapid increase of the mean-photon number in an energy eigenstate due to large entanglement among high-lying levels present in the PZW frame.

Derivation of the multimode generalization of the unitary transformation

We provide details about the derivation of the multimode generalization of our formalism presented in the main text. We start from the light-matter Hamiltonian including multiple spatially varying electromagnetic modes in the Coulomb gauge:

$$\hat{H}_C = \frac{\hat{p}^2}{2m} + V(x) - \frac{q}{2m} (\hat{p} \cdot \hat{A}(x) + \text{h.c.}) + \frac{q^2 \hat{A}^2(x)}{2m} + \sum_{k \lambda} \hbar \omega_k (\hat{a}_{k \lambda}^\dagger \hat{a}_{k \lambda} + \text{h.c.}), \quad (S27)$$

where we consider the vector potential expanded by plane waves

$$\hat{A}(x) = \sum_{k \lambda} \epsilon_{k \lambda} \hat{A}_k \left( \hat{a}_{k \lambda} e^{i k \cdot x} + \text{h.c.} \right), \quad \mathbf{k} \cdot \mathbf{e}_{k \lambda} = 0, \quad \epsilon_{k \lambda} \cdot \epsilon_{k' \lambda'} = \delta_{\lambda' \lambda}, \quad (S28)$$

with $\lambda$ denoting polarization. To generalize the asymptotically decoupling unitary transformation $\hat{U}$ to this multimode case, we first introduce the field operators

$$\hat{X}_{k \lambda}(x) \equiv \sqrt{\frac{\hbar}{2 \omega_k}} \left( \hat{a}_{k \lambda} e^{i k \cdot x} + \hat{a}_{k \lambda}^\dagger e^{-i k \cdot x} \right), \quad \hat{P}_{k \lambda}(x) \equiv \sqrt{\frac{\hbar \omega_k}{2}} i \left( \hat{a}_{k \lambda}^\dagger e^{-i k \cdot x} - \hat{a}_{k \lambda} e^{i k \cdot x} \right), \quad (S29)$$

and define the coupling as

$$g_k = q \hat{A}_k \sqrt{\frac{\omega_k \hbar}{m \hbar}}. \quad (S30)$$

We then rewrite the (quadratic) photon part of the Hamiltonian as

$$\frac{q^2 \hat{A}^2(x)}{2m} + \sum_{k \lambda} \hbar \omega_k (\hat{a}_{k \lambda}^\dagger \hat{a}_{k \lambda} + \hat{a}_{k \lambda} \hat{a}_{k \lambda}^\dagger) = \sum_{k \lambda} \frac{\tilde{P}_{k \lambda}(x)}{2} + \frac{1}{2} \sum_{k \lambda k' \lambda'} \left( \delta_{k \lambda, k' \lambda'} \omega_k^2 + 2 g_k g_{k'} \epsilon_{k \lambda} \cdot \epsilon_{k' \lambda'} \right) \hat{X}_{k \lambda}(x) \hat{X}_{k' \lambda'}(x)$$

$$= \frac{1}{2} \sum_{\alpha} \left( \tilde{P}_{\alpha}^2(x) + \Omega_{\alpha} \hat{X}_{\alpha}(x) \right), \quad (S31)$$

where we define the diagonalized basis labeled by $\alpha$ via

$$\hat{X}_{k \lambda}(x) = \sum_{k \lambda} O_{k \lambda, \alpha} \hat{X}_{\alpha}(x) \quad (S32)$$

with $O_{k \lambda, \alpha}$ being an orthogonal matrix.
FIG. S2. Comparisons of convergence of low-energy spectra in the AD frame (blue solid curves) and the PZW frame (red dotted curves) with respect to the photon-number cutoff \( n_c \) at different coupling strengths \( g \). The AD-frame energies efficiently converge at low \( n_c \), while the results in the PZW frame require an increasingly large cutoff at stronger \( g \), and do not converge for \( g/\omega_c \gtrsim 10 \) at least in the plotted scale.

We set \( \omega_c = \sqrt{\hbar/m_\omega} = 1 \) and choose the parameters \( \lambda = 3, \mu = 3.85 \).

We next introduce the \( x \)-dependent annihilation operators via
\[
\hat{b}_\alpha(x) = \sqrt{\Omega_\alpha/2} \hat{X}_\alpha(x) + \frac{i}{\sqrt{2\Omega_\alpha}} \hat{P}_\alpha(x),
\]
and also define the vector-valued variables labeled by \( \alpha \) as
\[
\zeta_\alpha = \sum_{k\lambda} \epsilon_{k\lambda} g_k x_{\Omega_\alpha} O_{k\lambda,\alpha},
\]
where \( x_{\Omega_\alpha} = \sqrt{\hbar/m\Omega_\alpha} \). We now introduce the unitary transformation in the multimode case by
\[
\hat{U} = \exp \left[ -i \frac{\hat{p}}{\hbar} \sum_{\alpha} \xi_\alpha \tilde{\alpha}_\alpha(x) \right], \quad \tilde{\alpha}_\alpha(x) = i \left( \hat{b}_\alpha(x) - \hat{\beta}_\alpha(x) \right), \quad \xi_\alpha = \frac{\zeta_\alpha}{\Omega_\alpha}.
\]

We note that, since the electromagnetic modes now explicitly depend on the position \( x \), they do not commute with the momentum operator \( \hat{p} \) in the transformation \( \hat{U} \), and thus, the transformed Hamiltonian in general acquires additional contributions compared to the simple expression obtained in the single-mode case [cf. Eq. (S25)]. Nevertheless, significant simplification can occur when the field variation is small compared with the effective length scale:
\[
k|\xi| \ll 1 \text{ for } |\nabla \hat{b}| \sim k\hat{b}.
\]
We emphasize that this condition is independent of system size and thus much less restrictive than the standard dipole approximation. In particular, Eq. (S36) can, in principle, be attained for any \( k \) if the coupling \( g \) is taken to be sufficiently strong such that the effective length scale \( |\xi| \) is short enough to satisfy this condition. Under this condition, the derivative terms of the field operators \( b(x) \), \( \pi(x) \) can be neglected, resulting in the simple transformed Hamiltonian:

\[
\hat{H}_U = \hat{U}^\dagger \hat{H}_C \hat{U} \simeq \frac{\hat{p}^2}{2m} - \sum_\alpha \frac{(\hat{p} \cdot \zeta_\alpha)^2}{\Omega_\alpha} + V \left( x + \sum_\alpha \xi_\alpha \pi_\alpha(x) \right) + \sum_\alpha \hbar \Omega_\alpha \hat{b}_\alpha^\dagger(x) \hat{b}_\alpha(x), \tag{S37}
\]

which provides Eq. (12) in the main text.