Non-linear optical processes in cavity light-matter systems

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We study non-linear optical effects in electron systems with and without inversion symmetry in a Fabry-Perot cavity. General photon up- and down-conversion processes are modeled by the coupling of a noninteracting lattice model to two modes of the quantized light field. Effective descriptions retaining the most relevant states are devised via downfolding and a generalized Householder transformation. These models are used to relate the transition amplitudes for even order photon-conversion processes to the shift vector, a topological quantity describing the difference in polarization between the valence and conduction band in non-centrosymmetric systems. We also demonstrate that the truncated models, despite their small Hilbert space, capture correlation effects induced by the photons in the electronic subsystem.

I. INTRODUCTION

Understanding the properties of light-matter coupled systems is a longstanding challenge in condensed matter physics. The goal of controlling material properties with classical light has been pursued actively in recent years, and remarkable phenomena such as interaction tuning or qualitative changes in the band structure via Floquet engineering have been theoretically demonstrated.\cite{1–8} On the spectroscopy side, research on high-harmonic generation (HHG) has shown that non-linear optical processes inherit important fingerprints of the electronic structure and even the Berry curvature of solids.\cite{9–14}

Another stimulating prospect is the exploration and control of strongly coupled light-matter systems in cavities, where the quantized nature of the photon field plays an important role.\cite{15–19} Despite decades of studies of cavity quantum electrodynamics (CQED) problems, as exemplified by the Rabi or Dicke model,\cite{20} this field has attracted renewed interest in the condensed matter community with the discussion of purported super-radiant states and the possibility of engineering novel states of matter.\cite{21–25} With phenomena such as HHG in mind, one may ask how photon up- and down-conversion occurs in these fully quantized light-matter systems, and what these nonlinear phenomena reveal about the (topological) properties of the material.

Following up on the pioneering work of Sipe et al.,\cite{26,27} a recent Floquet study by Morimoto and Nagaosa\cite{28} showed that in systems with time-reversal symmetry (TRS), but without inversion symmetry (IS), nonlinear optical processes can be related to topological quantities. Specifically, it was demonstrated that Floquet theory for an effective two-band model provides a suitable framework to investigate the shift current and non-linear Hall conductivity. Here, we extend this approach to a cavity set-up with quantized light. Instead of Floquet sidebands, we will consider a low energy theory in a system with two dominant photon modes. This simple set-up allows us to derive expressions for photon up- and down-conversion processes which are analogous to those in Refs. 26–28.

We further show that it is possible to capture the most relevant effects of the photons on the electronic states in an effective description involving a small number of “molecular orbitals.” This description is obtained by a block Householder transformation, which enables systematic truncations of the Hilbert space. Even after a truncation to just four states, the effective model correctly describes the photon-conversion processes, and provides qualitatively correct results for the kinetic energy and charge correlation functions.

The paper is organized as follows. In Sec. II we derive our minimal model for photon conversion in solids interacting with quantized light. This model is downfolded to an effective photon model in Sec. III, and it is shown that the transition amplitudes for even order up- or down-conversion are related to the shift vector. In Sec. IV we introduce the block Householder transformation and derive few-states electron-photon models. Section V tests the few-states effective description against the full model for a one-dimensional chain coupled to two photon modes.

II. MODEL

A. Coulomb gauge Hamiltonian

We consider a matter Hamiltonian representing a non-interacting lattice model with two orbitals in each unit cell. To describe the coupling to an electromagnetic field, we employ here the Coulomb gauge\cite{29} (similar to the velocity gauge in Ref. 30). The form of our Hamiltonian is thus different from the Hamiltonians encountered in recent studies which employ either a dipole gauge, obtainable through a Power-Zienau-Wolley (PZW)\cite{22,31} gauge transformation of the Coulomb gauge Hamiltonian, or the multi-center PZW transformation which preserves translational invariance.\cite{22,31,32} We refer to Appendix A for a discussion of the mappings between these different representations.

The Coulomb gauge Hamiltonian in a second quantized
form can be written as
\[ \hat{H}_{CG} = \sum_{k,\alpha,\beta} \hat{c}_{k,\alpha}^\dagger (u_{k,\alpha} | \hat{h}_0(k - q\sum_{\mu} g_{\mu} \hat{A}_{\mu}) | u_{k,\beta}) \hat{c}_{k,\beta} \]
\[ + \sum_{\mu} \frac{\Omega_{\mu}}{2} (\hat{\Pi}_{\mu}^2 + \hat{A}_{\mu}^2), \]
(1)
with \( \mu \) denoting the different modes of the transverse electromagnetic field. The vector potential and its conjugate variable are defined as \( \hat{A}_{\mu} = \frac{1}{\sqrt{2}}(\hat{a}_{\mu} + \hat{a}_{\mu}^\dagger) \) and \( \hat{\Pi}_{\mu} = \sqrt{2} (\hat{a}_{\mu} - \hat{a}_{\mu}^\dagger) \) in terms of the photon creation (annihilation) operators \( \hat{a}_{\mu} \) (\( \hat{a}_{\mu}^\dagger \)), and satisfy the commutation relations \([\hat{A}_{\nu}, \hat{\Pi}_{\mu}] = i \delta_{\mu,\nu} \) (all other commutators being zero). The photon energy for mode \( \mu \) is \( \Omega_{\mu} \), and the photon coupling (which we assume to be real) is denoted by \( g_{\mu} \).

\( \hat{c}_{k,\alpha}^\dagger \) creates an electron in a Bloch state with momentum \( k \) and band index \( \alpha \): \( \hat{c}_{k,\alpha}^\dagger | \text{vac} \rangle = | \psi_{k,\alpha} \rangle = e^{ik\xi} | u_{k,\alpha} \rangle \),
\[ \hat{h}_0(k) | u_{\alpha} \rangle = \epsilon_\alpha(k) | u_{\alpha} \rangle, \]
with \( \epsilon_\alpha(k) \) the corresponding energy. Here, \( \hat{h}_0(k) = e^{-ik\xi} \hat{H}_0 e^{ik\xi} \) with \( \hat{H}_0 \) the Hamiltonian of the noninteracting matter system. We will use units where the charge \( q = -|e| = -1 \).

For the study of general nonlinear optical processes involving two modes (see top panel of Fig. 1), one may expand
\[ \hat{h}_0(k + g_1 \hat{A}_1 + g_2 \hat{A}_2) \]
were we assume that each term for \( n \) or \( m > 0 \) in Eq. (2) scales as \( 1/\sqrt{N} \) with lattice size \( N \). Specifically, we will consider a two-band system with a conduction band (\( \alpha = c \)) and a valence band (\( \alpha = v \)), and we will be primarily interested in photon up- and down-conversion processes where \( \Omega_2 = 2\Omega_1 < \min_k (\epsilon_{k+c} - \epsilon_{k,v}) \) (splitting between the conduction and valence band, see bottom left panel of Fig. 1), and where the couplings \( g_{\mu} \) are so small that we only need to consider states with 0 or 2 (1) photons in mode \( \mu = 1 \) (\( \mu = 2 \)). In this case, one can restrict the sum in Eq. (2) to the first order term in \( \hat{A}_2 \) and the second order term in \( \hat{A}_1 \) and write
\[ \hat{h}_0(k + g_1 \hat{A}_1 + g_2 \hat{A}_2) \approx \hat{h}_0(k) + \hat{\nu}(k) g_2 \hat{A}_2 + \frac{1}{2} \hat{\nu}'(k) g_1^2 \hat{A}_1^2 \]
(3)
with \( \hat{\nu}(k) = \partial_k \hat{h}_0(k) \) and \( \hat{\nu}'(k) = \partial_{k k} \hat{h}_0(k) \). Note that the latter two operators may have off-diagonal elements in the band basis, since generically \( \hat{H}_0 \) does not commute with \([\hat{r}, \hat{H}_0]\) and \([\hat{r}, [\hat{r}, \hat{H}_0]]\), so that there is no complete set of common eigenstates of \( \hat{h}_0(k) \) and \( \hat{\nu}(k) \) or \( \hat{h}_0(k) \) and \( \hat{\nu}'(k) \).

In addition to the weak coupling assumption, we will employ the rotating wave approximation (RWA) and neglect terms such as \( \hat{c}_{k,v}^\dagger \hat{c}_{k,c} g_2 \hat{a}_2, \hat{c}_{k,v}^\dagger \hat{c}_{k,c} g_2 \hat{a}_1 \), etc. Also the contribution from \( \hat{a}_1^\dagger \hat{a}_1 \) in the expansion of \( \hat{A}_2^2 \), which leads to a level renormalization \( O(g_2^2) \), will be neglected.

With these approximations, the Coulomb gauge Hamiltonian becomes
\[ \hat{H}_{CG} \approx \sum_k \hat{\alpha}_{k,\alpha} \epsilon_\alpha(k) + \sum_k \left[ \frac{g_2}{\sqrt{2}} \nu_{v,c}(k) \hat{c}_{k,v}^\dagger \hat{c}_{k,c} \hat{a}_2 \right. \]
\[ + \frac{g_2^2}{4} \nu_{v,c}'(k) \hat{c}_{k,v}^\dagger \hat{c}_{k,c} (\hat{a}_1^\dagger)^2 + \text{h.c.} \]
\[ \left. + \sum_{\mu} \frac{\Omega_{\mu}}{2} (\hat{\Pi}_{\mu}^2 + \hat{A}_{\mu}^2), \right) \]
(4)
with \( \nu_{\alpha,\beta}(k) = \langle u_{k,\alpha} | \hat{\nu}(k) | u_{k,\beta} \rangle \) and similarly for \( \nu_{\alpha,\beta}'(k) \). The photon coupling strengths \( g_2 \) and \( g_1 \) are assumed to scale with system size \( N \) as \( g_2 = \tilde{g}_2 \sqrt{N} \) and \( g_1^2 = \tilde{g}_1^2 / \sqrt{N} \) where \( \tilde{g}_1 \) and \( g_2 \) are fixed parameters.

The specific set-up which we have in mind is illustrated in Fig. 1 and consists of a one-dimensional material placed within a Fabry Perot cavity with two relevant photon modes, and a one-dimensional material placed at an antinode of the second mode. The box indicates a possible choice of unit cell. Bottom panel: Illustration of the undressed valence and conduction bands (black lines), and the valence band dressed with two additional photons of energy \( \Omega_1 \) (red), as well as the valence band dressed with one photon of energy \( \Omega_2 \) (blue). Here, the box encloses the states considered in the low-energy model. The bottom right part of the figure illustrates the energy levels of the many-body system (see text).

FIG. 1. Top panel: Sketch of a Fabry Perot cavity with two relevant photon modes, and a one-dimensional material placed at an antinode of the second mode. The box indicates a possible choice of unit cell. Bottom panel: Illustration of the undressed valence and conduction bands (black lines), and the valence band dressed with two additional photons of energy \( \Omega_1 \) (red), as well as the valence band dressed with one photon of energy \( \Omega_2 \) (blue). Here, the box encloses the states considered in the low-energy model. The bottom right part of the figure illustrates the energy levels of the many-body system (see text).
the chain. In this situation, the canonical commutation relations read \([A_x(y), \Pi_z(y')] = i\delta(y - y')\). A convenient mode expansion is \(A_x(y) = \sum_{\mu} \phi_\mu(y) \hat{A}_\mu, \Pi_z(y) = \sum_\mu \epsilon(y) \phi_\mu^*(y) \hat{\Pi}_\mu\) with \(\epsilon(y)\) the dielectric function in the cavity, \([\hat{A}_\mu, \hat{\Pi}_\nu] = \delta_{\mu,\nu}\), and \(\int dy \epsilon(y) \phi_\mu^*(y) \phi_\nu(y) = \delta_{\mu,\nu}\). As noted in Ref. 34, the free-field part of Eq. (1) will depend on \(\epsilon(y)\) in dielectric media. In the following, we will use \(\epsilon = 1\) for simplicity. If \(\epsilon(y) \neq 1\), one can apply a suitable scaling transformation on the variables in Eq. (1). The cavity may be finite or infinitely extended along the directions \(x\) and \(z\), as long as there is no mixing with any modes with wavevectors \(q_{x,z} \neq 0\).

### B. Low energy model

For the electron-photon coupled many-body system, it is convenient to introduce the basis states |\(\alpha\rangle |m, n\rangle\), where \(|\alpha\rangle \equiv \otimes_{\alpha=1}^N |\alpha_i\rangle\) with \(|\alpha_i\rangle \in \{|c_i\rangle = |\psi_{k_i,c}\rangle, |f_i\rangle = |\psi_{k_i,f}\rangle\}\) representing an electron with momentum \(k_i\) in either the conduction or valence band, and \(m\) \((n)\) the number of photons with frequency \(\Omega_1\) \((\Omega_2)\). The relevant states for photon up- and down-conversion can be identified by looking at the bottom panel in Fig. 1. This figure displays the band structure, including photon-dressed states, and suggests to consider a subspace (black box) comprised of conduction band states (states of the type \(|v_1, \ldots, v_{i-1}, c_i, v_{i+1}, \ldots, v_N\rangle|0, 0\rangle\) as well as the valence band dressed with 1 or 2 photons of frequency \(\Omega_2\) and \(\Omega_1\), respectively (states \(|v_1, \ldots, v_{N}\rangle|0, 1\rangle\) and \(|v_1, \ldots, v_{N}\rangle|2, 0\rangle\)). These many-body states, along with their energies, are sketched in the bottom right panel of the figure. The corresponding low-energy model is described by the Hamiltonian matrix \(H_{\text{low}}\) expressed in the basis \(|\langle v_1, v_2, \ldots, v_{N}\rangle|0, 1\rangle, |\langle v_1, \ldots, v_{N}\rangle|0, 2\rangle\), \(|\langle v_1, \ldots, v_{N}\rangle|0, 0\rangle\), which reads

\[
H_{\text{low}} = \begin{pmatrix}
\Omega_2 & 0 & \frac{g_2}{\sqrt{2}} v_{vc}(k_1) & \frac{g_2}{\sqrt{2}} v_{vc}(k_2) & \ldots & \frac{g_2}{\sqrt{2}} v_{vc}(k_N) \\
0 & 2\Omega_1 & \frac{g_2}{2\sqrt{2}} v_{vc}(k_1) & \frac{g_2}{2\sqrt{2}} v_{vc}(k_2) & \ldots & \frac{g_2}{2\sqrt{2}} v_{vc}(k_N) \\
\frac{g_2}{\sqrt{2}} v_{vc}(k_2) & \frac{g_2}{2\sqrt{2}} v_{vc}(k_2) & \epsilon_c(k_1) - \epsilon_v(k_1) & 0 & \ldots & 0 \\
\vdots & \vdots & \vdots & \ddots & \vdots & \vdots \\
\frac{g_2}{\sqrt{2}} v_{vc}(k_N) & \frac{g_2}{2\sqrt{2}} v_{vc}(k_N) & 0 & \ldots & \epsilon_c(k_N) - \epsilon_v(k_N)
\end{pmatrix}
\]

### III. PHOTON CONVERSION AND SHIFT VECTOR

Even though model (5) has no direct coupling between the different photon states, photon conversion processes are induced via the coupling to electron-hole excitations. Similar phenomena are observed in HHG experiments on semi-conductors, where electron excitation and de-excitation processes result in photon up-conversion. More specifically, the conversion from two \(\Omega_1\) photons to a single \(\Omega_2\) photon in our cavity set-up is reminiscent of second harmonic generation in semiconductors (without inversion symmetry), where electrons are excited from the valence to the conduction band by an electric field with frequency \(\Omega_1\), while the de-excitation process generates radiation with frequency \(\Omega_2 = 2\Omega_1\). Analogous photon up- and down-conversions have also been discussed in atomic physics.

Following Ref. 36, we introduce the states \(|i\rangle, |f\rangle\) representing the initial and final states of the photon conversion process, respectively, and evaluate the transition amplitude through time-dependent perturbation theory. This calculation is valid if \(|i\rangle, |f\rangle\) are similar in energy and their energy difference to other states \(|j\rangle\) is sufficiently large. Splitting the Hamiltonian in Eq. (4) into

\[
\hat{H}_1 = \Omega_1 \hat{a}_1^\dagger \hat{a}_1 + \Omega_2 \hat{a}_2^\dagger \hat{a}_2 + \sum_{k,\alpha} \epsilon_\alpha(k) \hat{n}_{k,\alpha},
\]

where we have used that \(\Omega_2 = (\hat{\Pi}_2^\dagger + \hat{\Pi}_2) = \Omega_\mu \hat{a}_\mu^\dagger \hat{a}_\mu\) up to a constant, and

\[
\hat{H}_2 = \frac{g_2}{\sqrt{2}} \sum_k v_{cv}(k) \hat{c}_{k,c}^\dagger \hat{c}_{k,v} + \frac{g_2^2}{4} \sum_k v_{cv}(k) \hat{c}_{k,v}^\dagger \hat{c}_{k,v} + \text{h.c.},
\]

we obtain in second order perturbation theory the effective photon model

\[
\hat{H}_{\text{ph}} = d_{\text{trans}}|f\rangle \langle i| + d_{\text{trans}}^\ast |i\rangle \langle f| + d_x(|f\rangle \langle f| - |i\rangle \langle i|),
\]

with

\[
d_{\text{trans}} = \sum_{j \neq i, f} \frac{|\langle f| H_2 |j\rangle \langle j| H_2 |i\rangle|}{E_i - E_j}.
\]

Equation (9) corresponds to the amplitude for second order photon conversion if we choose \(|i\rangle = |2, 0\rangle\) and
\[ |f \rangle = |0, 1 \rangle, \text{ where } |m, n \rangle = |v_1, \ldots, v_N \rangle |m, n \rangle, \text{ and } E_i = 2\Omega + E_v = \Omega_2 + E_v = E_f (E_v \text{ denotes the energy of the full valence band}). \] The state \(|j\rangle\) describes the system without photons, but the electron with momentum \(k_j\) in the conduction band, which has energy \(E_j = \epsilon_c(k_j) - \epsilon_v(k_j) + E_v\). Hence \((j| \hat{H}_2 |2, 0\rangle = \frac{g^2}{2\sqrt{2}}(v')_{cv}(k_j), \langle 0, 1 | \hat{H}_2 |j\rangle = \frac{g^2}{\sqrt{2}} v_{vc}(k_j)\), and we get
\[ d_{\text{trans}}^{(2)} = \frac{g^2 g_2}{4} \sum_k \frac{v_{vc}(k)(v')_{cv}(k)}{2\Omega_1 - (\epsilon_c(k) - \epsilon_v(k))}, \tag{10} \]
where the superscript \(^{(2)}\) indicates that this is the amplitude for the second order photon conversion process \((\Omega_2 = 2\Omega_1)\). Furthermore, \(d_{\text{trans}}^{(2)}\) is given by
\[ d_{\text{trans}}^{(2)} = \frac{1}{2} \sum_k \frac{(\frac{g_2}{\sqrt{2}} v_{vc}(k))^2 - \frac{g^2}{2\sqrt{2}} (v')_{cv}(k))^2}{2\Omega_1 - (\epsilon_c(k) - \epsilon_v(k))}. \tag{11} \]
Assuming time reversal symmetry \((v_{\alpha\alpha}(-k) = -v_{\alpha\alpha}(k) \text{ and } \epsilon_{\alpha}(-k) = \epsilon_{\alpha}(k))\) one can show that \(d_{\text{trans}}^{(2)}\) is purely imaginary. In fact, in the numerator of the sum in Eq. (10) we recognize the product\(^{38}\)
\[ v_{vc}(k)(v')_{cv}(k) = |v_{vc}|^2 \left[ \partial_k \ln v_{vc} - i(\xi_c - \xi_v) + \frac{v_{vc} - v_{vc}}{\epsilon_c - \epsilon_v} \right], \tag{12} \]
where we have used that \((v'_{cv}) = (\frac{g_2}{2\sqrt{2}} v_{vc}) = \partial_k v_{cv} - \langle \partial_k u_{k,c} | \hat{v}(k) | u_{k,c} \rangle - \langle u_{k,c} | \hat{v}(k) | \partial_k u_{k,c} \rangle\) and inserted \[|u_{k,v}\rangle|u_{k,c}\rangle + |u_{k,c}\rangle|u_{k,v}\rangle = 1\] in order to obtain the last two terms above. Furthermore, we note that
\[ \langle u_{k,c} | \partial_k | u_{k,v} \rangle = -\frac{v_{vc}(k)}{\epsilon_c(k) - \epsilon_v(k)}, \tag{14} \]
and recall the definition of the Berry connection\(^{30,37}\)
\[ \xi_{\alpha}(k) = i \langle u_{k,\alpha} | \partial_k | u_{k,\alpha} \rangle, \tag{15} \]
which is real, since \[\partial_k \langle u_{k,\alpha} | u_{k,\alpha} \rangle = \langle \partial_k u_{k,\alpha} | u_{k,\alpha} \rangle + \langle u_{k,\alpha} | \partial_k u_{k,\alpha} \rangle = 0\]. Using that \(\text{Re} [\partial_k \ln v_{vc}] = \text{Re} [\partial_k \ln \frac{|v_{vc}|}{|v_{vc}|}], \) that \(\partial_k |v_{vc}\rangle(|v_{vc}|)\) is an odd (even) function of \(k\), and that the denominator is real and even, one thus finds that the real parts in the sum cancel. The remaining imaginary terms can be expressed as
\[ d_{\text{trans}}^{(2)} = i\frac{g^2 g_2}{4} \sum_k \frac{|v_{vc}(k)|^2 R_{cv}^k}{2\Omega_1 - (\epsilon_c(k) - \epsilon_v(k))}. \tag{16} \]
In Eq. (16) we introduced the shift vector \(R_{cv}^k\), defined as\(^{38}\)
\[ R_{cv}^k = \partial_k \text{Im} (\ln v_{vc}) = (\xi_c - \xi_v). \tag{17} \]
The shift vector is a quantity which appears in the second order optical response of systems that break inversion symmetry and its integral over the Brillouin zone can be related to the polarization difference between the valence and conduction band.\(^{38}\)
These results can be readily generalized to higher order photon conversion processes, although the low energy models derived from them may be more difficult to justify. If the mode frequencies are tuned to satisfy the resonance condition \(\Omega_2 = n\Omega_1\), with \(n > 1\) some integer, the low energy model is given by
\[ \hat{H}_{\text{CG}} \approx \sum_k \hat{n}_{k,\alpha} \epsilon_{\alpha}(k) + \sum_k \left[ \frac{g^2}{\sqrt{2}} v_{vc}(k) \hat{c}_{k,v} \hat{c}_{k,c} \hat{a}_{k}^\dagger \right] + \left[ \frac{g^2}{2\sqrt{2}} \frac{\partial^2 \hat{h}(k)}{\partial k^2} \right] + \frac{\Omega_\mu^2}{2} (\hat{A}_\mu + \hat{A}_\mu^\dagger), \]
and \(\hat{H}_{\text{ph}}\) involves the coupling
\[ d_{\text{trans}}^{(n)} = \frac{g^2 g_2}{4} \sum_k \frac{v_{vc}(k)(v^{(n-1)}_{cv}(k)}{n\Omega_1 - (\epsilon_c(k) - \epsilon_v(k))}, \tag{18} \]
where we introduced \(\hat{v}^{(n-1)}(k) = \frac{\partial^{n-1} \hat{h}(k)}{\partial k^{n-1}}\). If \(\hat{h}(k)\) is time-reversal symmetric and comprised of trigonometric functions, then \(\partial_k \hat{v}(k) = -\partial_k \hat{v}(k)\), which leads to
\[ v_{vc}(v^{(n-1)}_{cv}) = \left[ \sin \left( \frac{n\pi}{2} \right) v_{vc}^2 - \cos \left( \frac{n\pi}{2} \right) v_{vc}(v')_{cv} \right]. \tag{19} \]
The calculation of \(d_{\text{trans}}^{(n)}\) and \(d_{\text{trans}}^{(n)}\) then proceeds as before. In particular, if we repeat the calculation for third order processes \((n = 3)\), assuming \(\Omega_2 = 3\Omega_1\), we find that \(d_{\text{trans}}^{(3)}\)
is real, and not related to the shift vector:

$$d^{(3)}_{\text{trans}} = -\frac{g_1^2 g_2}{4\cdot \sqrt{3!}} \sum_k \frac{|v_{\text{exc}}(k)|^2}{3\Omega_1 - (\epsilon_c(k) - \epsilon_e(k))}. \quad (20)$$

For even \(n\), the result is purely imaginary, and nonzero only in the presence of a non-vanishing shift vector, while for \(n\) odd, the result is real and independent of the shift vector. As illustrated in Fig. 2, we can define a vector \(d = (d_x, d_y, d_z)\) with \(d_x = \text{Re}[d^{(n)}_{\text{trans}}]\), \(d_y = \text{Im}[d^{(n)}_{\text{trans}}]\), \(d_z = d^{(n)}_{\text{trans}}\), which rotates (in steps of 90°) around the \(z\)-axis in a generically elliptic spiral pattern as \(n\) increases. If \(d_z\) is nonzero, as is usually the case, the expectation values of \(\sigma_x, \sigma_y\) in the downfolded model are less than one.

In Appendix B we show that the same effective photon model \(H_{\text{ph}}\) can also be obtained from the low-energy model (5) by a downfolding procedure in which the states with excited electrons are integrated out.

**IV. EFFECTIVE ELECTRON-PHOTON MODEL**

The model derived in the previous section allows us to analyze the photon-conversion process, but does not provide information on how the coupling to the photons affects the charge degrees of freedom. We will next devise a simple few-states model which captures this back action on the electrons.

The matrix of the low-energy model (5) represents a generalized star geometry, illustrated in the top left panel of Fig. 3, in which two photon states (black dots) are coupled to \(i = 1, \ldots, N\) electronic states (grey dots representing \(|v_1, \ldots, v_i, \ldots, v_N\rangle\langle 0, 0\rangle\)). In this geometry, it is not obvious how to optimally truncate the model and to derive a few-states model which captures the essential physics. For example, a naive truncation from \(N\) to two conduction band states does not yield an appropriate model, since the effect of excitations at other \(k\) points is simply ignored. Instead, we implement a procedure which is analogous to the mapping of the Anderson impurity model from a star geometry to a chain geometry. After the latter transformation, the coupling to the first site of the chain is given by the geometric mean of all possible impurity-bath couplings. The first site in the chain hence represents a “molecular bath orbital,” and even after a truncation of the chain, the model captures some relevant non-local physics.

In the following, we employ a similar mapping involving \(2 \times 2\) block matrices. In deriving this transformation, we draw on previous results for block transformations of symmetric matrices. For notational convenience, we subsume some symbols in Eq. (5) and write

$$H^\text{low} = \begin{pmatrix} \Omega_2 & 0 & V_1 & V_2 & \ldots & V_N \\ 0 & 2\Omega_1 & V_1^\dagger & V_2^\dagger & \ldots & V_N^\dagger \\ V_1^* & (V_1^\dagger)^* & \epsilon_1 & 0 & \ldots & \vdots \\ V_2^* & (V_2^\dagger)^* & 0 & \epsilon_2 & \ldots & \vdots \\ \vdots & \vdots & \vdots & \vdots & \ddots & 0 \\ V_N^* & (V_N^\dagger)^* & 0 & 0 & \ldots & \epsilon_N \end{pmatrix}. \quad (21)$$

The high degree of entanglement is evident by the off-diagonal couplings between the electronic subsystem and the photons. In analogy to the chain representation of the Anderson impurity model, we seek a generalized Householder transformation that can transform the problem into a tridiagonal block matrix form. Adapting the derivation of Ref. 41 to suit the present problem of a Hermitian starting matrix, we define the following block Householder transformation

$$U_1 = \begin{pmatrix} \Omega_2 & 0 & V_1 & V_2 & \ldots & V_N \\ 0 & 2\Omega_1 & V_1^\dagger & V_2^\dagger & \ldots & V_N^\dagger \\ V_1^* & (V_1^\dagger)^* & \epsilon_1 & 0 & \ldots & \vdots \\ V_2^* & (V_2^\dagger)^* & 0 & \epsilon_2 & \ldots & \vdots \\ \vdots & \vdots & \vdots & \vdots & \ddots & 0 \\ V_N^* & (V_N^\dagger)^* & 0 & 0 & \ldots & \epsilon_N \end{pmatrix} \begin{pmatrix} 1_{2\times 2} & 0_{2\times N} \\ 0_{N\times 2} & \frac{1}{\hbar_1} \end{pmatrix},$$

where

$$\hbar_1 = 1 - 2V_A(V_A^\dagger V_A)^{-1}V_A^\dagger \quad (22)$$

and \(V_A\) is defined in terms of a still to be determined matrix \(X\) as

$$V_A = \begin{pmatrix} A_1 + X \\ A_2 \end{pmatrix}, \quad (23)$$
In general, a diagonal non-negative $n \times n$ matrix $H^{\text{low}}$ can be transformed to a block tri-diagonal form. The unitary matrix $U$ defined by the product

$$U = U_1 \cdot U_2 \ldots U_{N/2-2} \cdot U_{N/2-1}$$

thus produces the desired mapping.

Since as in the case of the Anderson impurity model, the first sites of the ladder represent molecular orbitals, the truncation of the model at this level (four-states model) still captures the coupling of the photons to the entire lattice. We will show in Sec. V that this simple model indeed provides an accurate description of the photon conversion amplitudes, and a meaningful description of the back action of the photons on the electrons.

To measure observables in the effective four-states model, we need to perform the same basis transformation on the corresponding operator matrices prior to truncation, symbolically denoted by $O \rightarrow TU^\dagger OUT$, where $T$ represents the truncation of the transformed operator to a given number of states.

In particular, the photon conversion in our model will be described by the operator matrices

$$S_x = \begin{pmatrix} \sigma_x & 0_{2 \times N} \\ 0_{N \times 2} & 0_{N \times N} \end{pmatrix}, \quad S_y = \begin{pmatrix} \sigma_y & 0_{2 \times N} \\ 0_{N \times 2} & 0_{N \times N} \end{pmatrix},$$

where $\sigma_i$, $i = x, y$ are the Pauli matrices.

![Plot of the band structure of model (33)](image)
V. RESULTS

A. The model

As in the previous sections, we consider a spinless electron system with two bands, which is coupled to two photon modes with frequencies $\Omega_1$, $\Omega_2$ and coupling constants $g_1$ and $g_2$, and we assume $\Omega_2 = 2\Omega_1$. The electronic part is given by a one-dimensional chain with a staggered potential $Q$ and bond strength $\ell$, corresponding to the real-space Hamiltonian:\footnote{V. J. Milliken, J. Phys. C 16, 93 (1983).}

$$\hat{H}_0 = \sum_i 2t[1-(-1)^i](\hat{c}_i^{\dagger}\hat{c}_{i+1} + \text{h.c.}) + Q(-1)^i\hat{c}_i^{\dagger}\hat{c}_i. \quad (33)$$

This model has TRS but breaks IS if both $\ell$ and $Q$ are nonzero.\footnote{P. W. Anderson, Phys. Rev. 101, 147 (1956).} In a sublattice basis we can write $\hat{H}_0 = \sum_k \psi^\dagger(k)h_0(k)\psi(k)$ with

$$h_0(k) = \begin{pmatrix} -2t\cos(k) & 2i\ell\sin(k) + Q \\ -2i\ell\sin(k) + Q & 2t\cos(k) \end{pmatrix} \quad (34)$$

and $\psi^\dagger_k = (\hat{c}_k, \hat{c}_{k+\pi})$. The energy bands for the model parameters $\Omega_2 = 0.853$, $\ell = -0.2$, $\ell = 0.1$, $Q = 0.6$, as well as the corresponding matrix elements $u_{vc}$ and $v'_{vc}$ in the band basis are shown in Fig. 4. In calculating $u_{vc}$ and $v'_{vc}$, we fix the arbitrary complex phase of the Bloch functions in such a manner as to keep the first entry of the eigenstates real. We will use this set-up in the following analysis.

B. Tests of the few-states model

To investigate the accuracy of the few-states models derived by the Householder scheme we focus on the expectation values of $\hat{S}_x$ and $\hat{S}_y$. In the practical implementation, there is a freedom in choosing the ordering of the $k$ points in the matrix (21). Let us define an ordered set of points $k_i = -\pi + (i-1)(2\pi/N)$, $i = 1, \ldots, N$. The upper panel of Fig. 5 shows the convergence of the truncated models towards the exact results (light red and light blue lines) with increasing number of retained states, for the choice $k_i = \tilde{k}_i$. The results derived by the simple truncation of the matrix (cross symbols) converge very slowly, and these models require either the full set of $k$ points, or exactly half the set of $k$ points to recover the exact reference values. On the other hand, the few-states models obtained through the Householder transformation produce the correct expectation values independent of the number of states retained, and in particular already for the smallest four-states model.

The lower panels of Fig. 5 illustrate how this result changes under cyclic permutations of the $k$ points: $\pi_{cyc}(\tilde{k}_i) = \tilde{k}_{i+1}$ (with periodic boundary conditions applied). The label $n$ on the horizontal axis refers to the number of cyclic permutations, i.e., the corresponding $k$ points are defined as $k_i = \pi_{cyc}^n(\tilde{k}_i)$. What is shown on the vertical axes are the expectation values of $\hat{S}_x$ and $\hat{S}_y$ from the four-states models obtained by the Householder scheme (left panel) and by the simple truncation (right panel). Again, the light red and light blue lines indicate the exact reference values from the solution of the full model, while the dashed horizontal lines in the right panel show the average of the results over all cyclic permutations. In the simple truncation scheme, even such an average does not recover the correct result for $\hat{S}_y$.

Next, let us consider the effect of a random permutation $\pi_{rand}$ of $\tilde{k}_i$. Figure 6 shows the results analo-
The number of states retained and the ordering of the $k$ produce the exact expectation values independent of the $S$. It makes a difference if this is the unity resolved in the truncated or non-truncated Hilbert space. To be precise, for any operator expression, we perform the unitary transformation on all the electronic density operators in the expression and subsequently apply a truncation. Specifically,

$$\hat{O}_k(t) \rightarrow e^{i\hat{H}^{-1/2}\hat{U}\hat{T}\hat{U}^\dagger_{t}} \hat{O}_k \hat{T} e^{-i\hat{H}^{-1/2}t},$$

which is consistent with the implicit rule we have employed when truncating the Hamiltonian (which is only comprised of electronic density terms). Following this convention, in the Lehmann formula, we insert the unity $1 = \sum_n |n_\text{HH}\rangle \langle n_\text{HH}|$ between the operators $\hat{n}_{k,\alpha}(t)$ and $\hat{n}_{k,\alpha}(0)$, where $|n_\text{HH}\rangle$ are the eigenstates of $\hat{T}\hat{U}^\dagger \hat{U} \hat{H}$ with $\hat{H}$ the Hamiltonian operator corresponding to Eq. (5). (For an additional subtlety regarding the representation of the full model see Appendix D.)

In the top panel of Fig. 7, we display the absolute value of the charge correlation function of the full model for $\alpha = c$. The region of nonzero weight covers the semi-continuum of energy excitations obtained from the diagonalization of model (5). In the middle panel, results from the truncated models obtained by the indicated numbers of Householder iterations are shown. With just two “molecular orbitals” the four-states model is of course not able to accurately capture the full range of possible excitations, but it provides two peaks which in a reasonable way represent the continuum of excitations in the full model. At the next iteration (6-states model) the energy range of possible excitations is already well captured, while after ten iterations (22-states model) the envelope of the excitation spectrum starts to be correctly captured. In contrast, the simple truncation produces an erratic collection of peaks that converges very slowly with increasing dimension of the truncated space (not shown).

Alternatively, the accuracy of the Householder scheme can be assessed by looking at the time-dependent density-density correlations and in particular at the critical time after which the approximate correlation functions start to deviate from the reference data from the full model (grey line in the bottom panel of Fig. 7). The four-states model captures the first oscillation, the 6-states model the first three oscillations, while the 22-states model correctly reproduces all oscillations up to $t \approx 200$.

One may wonder if it is also possible to compute site-dependent correlation functions. By assuming translational invariance, we may define the charge correlation function between sites $0$ and $R$ as

$$\xi_{\alpha}(R, t) \equiv \langle \hat{n}_{R,\alpha}(t)\hat{n}_{0,\alpha}(0) \rangle - \langle \hat{n}_{R,\alpha}(0) \rangle \langle \hat{n}_{0,\alpha}(0) \rangle.$$
Focusing on the conduction band and using $\hat{c}_{k,c}^\dagger$ are multiplied by 100. The corresponding Hilbert space sizes are $(2i + 2)$. The model parameters are $\tilde{g}_1 = 0.035$, $\tilde{g}_2 = 0.005$, $\Omega_2 = 0.853$, $t = -0.2$, $t = 0.1$, $Q = 0.6$, $N = 100$, for which the band structure is shown in the inset. All shown results are multiplied by 100.

Figure 8 plots $E_c = \sum_k \epsilon_c(k) \langle \hat{n}_{k,c} \rangle$, which, just like the charge correlation functions discussed previously, is a quantity that does not scale with system size in our model. The inset panel shows the modifications in the band structure as $Q$ is changed from a low value, where the photon-dressed valence band touches the conduction band (semi-transparent band structure), to the higher values corresponding to a substantial gap (opaque band structure). The thick grey line represents the results obtained for the full model (Eq. (5)). For all values of $Q$, the accuracy of the truncated Householder model increases systematically with increasing $i$, but the deviations get larger for smaller $Q$.

Close to the value $Q = 0.38$, where the bands touch, we observe a strong enhancement of the kinetic energy. In this parameter regime, electron-hole excitations become more probable due to the decreased energy gap. We caution however that even the full model must be treated with care in this region, due to the increased likelihood of multi-electron excitations to the conduction band, which are not captured in Eq. (5). Unless the coupling strengths are decreased correspondingly, even the full model description will become inaccurate.

In general, in the presence of band crossings, new basis states must be introduced in the low-energy model, including states that have a substantial fraction of electrons in the conduction band.

Finally, we plot in Fig. 9 the kinetic energy contri-
parameters are \( g \) for different values of the photon coupling strength \( g \).

**FIG. 9.** Kinetic energy contribution of the conduction band for different values of the photon coupling strength \( g \). The parameters are \( g_1 = 0.035, \Omega_2 = 0.85, t = -0.2, i = 0.1, Q = 0.6 \) and \( N = 100 \), and the corresponding band structure is shown in the left inset. The exact result is plotted as a thick grey line, while the other curves indicate the results after \( i \) Householder steps and subsequent truncation to a \((2i + 2)\)-states model. The upper right inset shows the difference in \( E_c \) between the full and truncated model.

The distribution of the valence band as a function of the photon coupling strength \( g_2 \) for a set-up with a sufficiently large splitting between the bands that our low-energy model (5) is justified. The deviations between the full calculation (thick grey line) and the results from the truncated Householder models increase with increasing coupling strength, but the results of the truncated models systematically and rapidly converge towards the exact result with increasing number of states kept in the effective description (see also the right inset, which plots the difference to the exact result). These data confirm that the effective few-states models derived via the Householder transformations correctly capture the effect of the photon coupling on the electronic properties.

**VI. CONCLUSIONS**

We have introduced a minimal model describing photon conversion processes in cavity light-matter systems. If the lattice model representing the matter subsystem has TRS, but no IS, even order nonlinear processes are activated, with a transition amplitude that is related to the shift vector. This result generalizes previous analyses for lattice models driven by classical light to the quantum domain. It should be noted, however, that the set-up considered in our study differs in one important aspect from the Floquet study in Ref. 28. There, a low-energy model was considered which describes a Floquet sideband of the valence band overlapping with the conduction band. In the present study, we considered situations with a sufficiently large gap between the photon-dressed valence band and the conduction band, and weak photon couplings, so that it is meaningful to restrict the low-energy model to states with at most one electron in the conduction band.

While a photon-only model capturing the effect of the light-matter coupling on the photon states can be easily derived by time-dependent perturbation theory or downfolding, the long-ranged correlations induced by the photons make the derivation of an effective few-states model with electronic degrees of freedom a nontrivial task. We employed a generalized Householder transformation to introduce a coupling of the photons to extended molecular orbitals. This transformation enables systematic truncations to effective models with a small number of electronic degrees of freedom, which nevertheless capture the interplay between the photons and the electronic subsystem. In particular, we showed that the photon conversion processes are still accurately described even by a four-states model, while the back-action of the photons on the electronic subsystem can be at least qualitatively captured with a modest number of states. This is in stark contrast to a simple truncation in the original basis of our minimal model, which due to the highly entangled nature of the photon and electron subsystems, does not result in a meaningful description.

We illustrated the usefulness of the Householder approach by demonstrating the fast convergence of the effective model descriptions to the full result with increasing number of states kept, for different observables related to the electronic and photonic subsystems. This is in particular the case if there exists a sufficiently large energy gap between the nonzero photon states and those with electronic excitations. As this gap shrinks, states with multiple electrons in the conduction band become more likely, and both our original model and its effective few-states descriptions will eventually break down.

Generalizations of the Householder method to multimode systems or other light-matter systems with a high degree of entanglement are interesting prospects. It is also worthwhile to investigate more closely the structure of the effective orbitals generated by the Householder scheme, and to search for models in cavity QED, for which the application of the block Householder transformation results in an analytically solvable problem. This could produce valuable insights into the nontrivial correlations induced by strong light-matter coupling.

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Appendix A: Gauge considerations

1. General remarks

Traditionally, light-matter coupled systems in the semi-classical approximation have been described in either the length gauge or velocity gauge.\textsuperscript{47,48} Both representations have their merits, and the effect of basis truncations in both of them has been discussed in terms of the (multi-center) PZW transformation.\textsuperscript{22,31,32} Here, we demonstrate that these representations are connected by unitary transformations and hence equivalent in the non-truncated Hilbert space. The strategy is to pass via the length gauge, corresponding to a term $\hat{r} \cdot \vec{E}$ in the Hamiltonian in first quantization. Since this breaks translational invariance (something which is also apparent in the Hamiltonian of Ref. 32), one needs to consider the infinite volume limit. Blount\textsuperscript{49} showed that in this limit, the matrix element of $\hat{r}$ between Bloch states can be defined in terms of Eq. (A5) below. In a subsequent limit, the matrix element of the infinite volume limit, where it is to be interpreted as shown that in this

\[
\int dr \hat{\Psi}^\dagger(r) r \hat{\Psi}(r) = \frac{1}{(2\pi)^d} \int dkdk' \sum_{\alpha,\beta} c_{k,\alpha}^\dagger \langle \psi_{k,\alpha} | \hat{r} | \psi_{k',\beta} \rangle c_{k',\beta}^\dagger
\]

where in the Bloch basis

\[
\int dr \hat{\Psi}^\dagger(r) r \hat{\Psi}(r)
\]

\[
\int dkd\mathbf{k}' \sum_{\alpha,\beta} c_{k,\alpha}^\dagger r_{\alpha,\beta}(k,k') c_{k',\beta}^\dagger.
\]

Note that this matrix element is ill defined, except in the infinite volume limit, where it is to be interpreted as\textsuperscript{30,49}

\[
r_{\alpha,\beta}(k,k') = (2\pi)^d \delta_{\alpha,\beta} [ -i \nabla_{\mathbf{k}'} \delta(k'-k) + \xi_{k,\alpha,\beta} \delta(k'-k) ]
\]

\[
+ (2\pi)^d (1 - \delta_{\alpha,\beta}) \xi_{k,\alpha,\beta} \delta(k'-k)
\]

which suggests to define intra- and inter-band matrix elements as follows: $r_{\alpha,\beta}(k,k') = (r_{\alpha,\beta} (k,k') + (r_{\alpha,\beta}(k,k'))$, with

\[
(r_{\alpha,\beta} (k,k')) = (2\pi)^d \delta_{\alpha,\beta} [ -i \nabla_{\mathbf{k}'} \delta(k'-k) + \xi_{k,\alpha,\beta} \delta(k'-k) ]
\]

and

\[
(r_{\alpha,\beta}(k,k')) = (2\pi)^d (1 - \delta_{\alpha,\beta}) \xi_{k,\alpha,\beta} \delta(k'-k)
\]

with $\xi_{k,\alpha,\beta} = i \langle u_{\alpha,\beta} | \nabla_{\mathbf{k}} | u_{\beta,\alpha} \rangle$. Application of the Baker-Campbell-Hausdorff (BCH) formula $e^B e^C = \sum_n \frac{1}{n!} [B, ... [B, [B, A]]]$ gives

\[
\hat{\mathcal{U}} \frac{1}{(2\pi)^d} \int d\mathbf{k} c_{k,\alpha}^\dagger \langle u_{\alpha,\beta} | \hat{h}_0(\mathbf{k}) | u_{\beta,\alpha} \rangle \hat{c}_{k,\alpha} \hat{\mathcal{U}}^\dagger
\]

\[
= \frac{1}{(2\pi)^d} \int d\mathbf{k} c_{k,\alpha}^\dagger \langle u_{\alpha,\beta} | \hat{h}_0(\mathbf{k}) | u_{\beta,\alpha} \rangle \hat{c}_{k,\alpha} \hat{\mathcal{U}}^\dagger
\]

Note that the off-diagonal elements in (A6) are generated by $\mathbf{r}$. Using Eq. (A6), we may write the Hamiltonian in Eq. (A1) as

\[
\hat{H}_{CG} = \hat{\mathcal{U}} \frac{1}{(2\pi)^d} \int d\mathbf{k} c_{k,\alpha}^\dagger \langle u_{\alpha,\beta} | \hat{h}_0(\mathbf{k}) | u_{\beta,\alpha} \rangle \hat{c}_{k,\alpha} \hat{\mathcal{U}}^\dagger
\]

\[
+ \frac{\Omega_0}{2} (\hat{\mathbf{P}}^2 + \hat{\mathbf{A}}^2).
\]

2. Coulomb gauge Hamiltonian

Using the notations and definitions of the main text, the Coulomb gauge Hamiltonian of the light-matter coupled system, obtained through the minimal-coupling procedure,\textsuperscript{29} reads

\[
\hat{H}_{CG} = \frac{1}{(2\pi)^d} \int d\mathbf{k} c_{k,\alpha}^\dagger \langle u_{\alpha,\beta} | \hat{h}_0(\mathbf{k} - gg \hat{A}) | u_{\beta,\alpha} \rangle \hat{c}_{k,\beta}
\]

\[
+ \frac{\Omega_0}{2} (\hat{\mathbf{P}}^2 + \hat{\mathbf{A}}^2).
\]

For simplicity, we consider here a single photon mode with energy $\Omega_0$, $\hat{\mathbf{P}} = \frac{i}{\sqrt{2}}(\hat{a}^\dagger - \hat{a})$, $\hat{\mathbf{A}} = \frac{i}{\sqrt{2}}(\hat{a} + \hat{a}^\dagger)$, and $[\hat{A}, \hat{\mathbf{P}}] = i$, while the fermionic operators satisfy $\{ \hat{c}_{k,\alpha}, \hat{c}^\dagger_{k',\beta} \} = (2\pi)^d \delta_{\alpha,\beta} \delta(k-k')$. Further, we define $\hat{\mathbf{A}} = \hat{\mathbf{A}} \cdot \hat{\mathbf{n}}$, $\hat{\mathbf{P}} = \hat{\mathbf{p}} \cdot \hat{\mathbf{n}}$ with $\hat{\mathbf{n}}$ the polarization direction of the mode.

In this section, we discuss how to generate the above Hamiltonian from the one without field by means of a unitary transformation. The PZW transformation can be written as\textsuperscript{51}

\[
\hat{\mathcal{U}} = e^{ig \hat{\mathbf{A}} \cdot \int dr \hat{\Psi}^\dagger(r) r \hat{\Psi}(r)}
\]

3. Connection to the dipole gauge Hamiltonian

We define the dipole gauge Hamiltonian as

\[
\hat{H}_{DG} = \hat{\mathcal{U}}^\dagger \hat{H}_{CG} \hat{\mathcal{U}}.
\]

By another application of the BCH formula, one finds

\[
\hat{\mathcal{U}}^\dagger \hat{\mathbf{P}} \hat{\mathcal{U}} = \hat{\mathbf{P}} + \frac{1}{(2\pi)^d} \int dkd\mathbf{k}' \sum_{\alpha,\beta} c_{k,\alpha}^\dagger r_{\alpha,\beta}(k,k') \hat{c}_{k',\beta}^\dagger.
\]
such that

\[
\hat{H}_{DG} = \frac{1}{(2\pi)^d} \sum_{\alpha} \int d k \hat{c}_{k,\alpha}^\dagger (u_{k,\alpha} \hat{h}_0(k) |u_{k,\alpha}) \hat{c}_{k,\alpha} \\
+ \frac{\Omega_0}{2} \left( \hat{I} + q g \frac{1}{(2\pi)^d} \int d k d k' \sum_{\alpha,\beta} \hat{c}_{k,\alpha}^\dagger r_{\alpha,\beta}(k, k') \hat{c}_{k',\beta} \right)^2 \\
+ \frac{\Omega_0}{2} \hat{A}^2.
\]

Ref. 22, 31, and 32 all used the so-called multi-center PZW transformation, motivated by the fact that the standard PZW transformation in (A2) leads to a Hamiltonian breaking translational invariance. However, \(\hat{H}_{DG}\) above is unitarily equivalent to the Hamiltonian in the mentioned references, as one can show by applying the following unitary transformation

\[
\hat{U} = e^{-qg\hat{A} \frac{1}{(2\pi)^d} \int d k d k' \sum_k \hat{c}_{k,\alpha}^\dagger \nabla_{k'}(k' - k) \hat{c}_{k,\alpha}}.
\]

The action of \(\hat{U}\) on some operator \(\hat{O} = \frac{1}{(2\pi)^d} \int d k \sum_{\gamma,\delta} \hat{c}_{\gamma,\delta}^\dagger O_{\gamma,\delta}(k) \hat{c}_{\gamma,\delta}\) is

\[
\hat{U}^\dagger \hat{O} \hat{U} = \frac{1}{(2\pi)^d} \int d k \sum_{\gamma,\delta} \hat{c}_{\gamma,\delta}^\dagger \nabla_{k} (k - qg\hat{A}) \hat{c}_{\gamma,\delta}.
\]

The Hamiltonian in Eq. (A10) thus transforms as

\[
\hat{U}^\dagger \hat{H}_{DG} \hat{U} = \frac{1}{(2\pi)^d} \int d k \sum_{\alpha} \hat{c}_{k,\alpha}^\dagger \epsilon_{\alpha}(k - qg\hat{A}) \hat{c}_{k,\alpha} \\
+ \frac{\Omega_0}{2} \hat{U}^\dagger \left( \hat{I} + q g \frac{1}{(2\pi)^d} \int d k d k' \sum_{\alpha,\beta} \hat{c}_{k,\alpha}^\dagger r_{\alpha,\beta}(k, k') \hat{c}_{k',\beta} \right)^2 \hat{U} \\
+ \frac{\Omega_0}{2} \hat{A}^2.
\]

Using

\[
\hat{U}^\dagger \hat{H} \hat{U} = \hat{H}_{DG} + i q g \frac{1}{(2\pi)^d} \int d k d k' \sum_{\alpha,\beta} \hat{c}_{k,\alpha}^\dagger \nabla_{k'}(k' - k) \hat{c}_{k',\beta}
\]

and

\[
\hat{U}^\dagger \left( q g \frac{1}{(2\pi)^d} \int d k d k' \sum_{\alpha,\beta} \hat{c}_{k,\alpha}^\dagger r_{\alpha,\beta}(k, k') \hat{c}_{k',\beta} \right) \hat{U} = -i q g \frac{1}{(2\pi)^d} \int d k d k' \sum_{\alpha,\beta} \hat{c}_{k,\alpha}^\dagger \nabla_{k'}(k' - k) \hat{c}_{k',\beta} \\
+ q g \frac{1}{(2\pi)^d} \int d k \sum_{\alpha,\beta} \hat{c}_{k,\alpha}^\dagger \epsilon_{k - qg\hat{A},\alpha,\beta} \hat{c}_{k,\beta}
\]

we get

\[
\hat{U}^\dagger \hat{H}_{DG} \hat{U} = \frac{1}{(2\pi)^d} \int d k \sum_{\alpha} \hat{c}_{k,\alpha}^\dagger \epsilon_{\alpha}(k - qg\hat{A}) \hat{c}_{k,\alpha} \\
+ \frac{\Omega_0}{2} \left( \hat{I} + q g \frac{1}{(2\pi)^d} \int d k \sum_{\alpha,\beta} \hat{c}_{k,\alpha}^\dagger \epsilon_{k - qg\hat{A},\alpha,\beta} \hat{c}_{k',\beta} \right)^2 \\
+ \frac{\Omega_0}{2} \hat{A}^2.
\]

which corresponds to an infinite volume version of the dipolar Hamiltonian in momentum space [Eq. (54)] presented in Ref. 22.

One could in principle also compute the results of the main text in one of these alternative gauges. However, the observable related to photon conversion (Eq. (32)) can be expected to become a mixture of photon and electron operators if one applies the transformations above. Hence, the analysis relating photon conversion to the shift vector is most conveniently done in the Coulomb gauge.

### Appendix B: Downfolding

In the downfolding approach\(^5^2\), we split a generic Hamiltonian into low-energy and high-energy subspaces,

\[
H = \begin{pmatrix}
H_{ll} & H_{lh} \\
H_{hl} & H_{hh}
\end{pmatrix}.
\]

In the present context, \(H_{ll}\) corresponds to the top left \(2 \times 2\) block of Eq. (5) or Eq. (21). To obtain an effective model for the low-energy space we can iteratively solve the eigenvalue equation \(H_{\text{eff}}(\epsilon) |\psi_{\text{eff}}\rangle = \epsilon |\psi_{\text{eff}}\rangle\) with

\[
H_{\text{eff}}(\epsilon) = H_{ll} + H_{lh} \frac{1}{\epsilon - H_{hh}} H_{hl}.
\]

To a first approximation one may use the eigenenergy of the unperturbed low-energy space, \(\epsilon = \Omega_2 = 2\Omega_1\). This yields

\[
H_{\text{eff}}(\Omega_2) = \begin{pmatrix}
\Omega_2 + d_z & d_{\text{down}}^* \\
d_{\text{down}} & 2\Omega_1 - d_z
\end{pmatrix}
\]

with

\[
d_{\text{down}} = \sum_j \frac{V_j V_j^{*}}{\Omega_2 - (\epsilon_c(k_j) - \epsilon_c(k_j))},
\]

\[
d_z = \frac{1}{2} \sum_j \frac{|V_j|^2 - |V_j^{*}|^2}{2\Omega_1 - (\epsilon_c(k_j) - \epsilon_v(k_j))}.
\]

This \(H_{\text{eff}}\) is identical to the effective Hamiltonian obtained from the second order perturbation calculation.

### Appendix C: Permutation of \(k\) points

The numerical evidence presented in the main text suggests that the four-states models derived via the Householder scheme do not depend on the ordering of the \(k\) points in the original Hamiltonian matrix. Here, we will provide a proof that for given \(k_1\) and \(k_2\) (states selected in the first Householder step), the result does not change upon permutation of the remaining \(k\) points.
We have
\[
A_2 \to \begin{pmatrix}
V_{\pi(3)} & V'_{\pi(3)} \\
V_{\pi(4)} & V'_{\pi(4)} \\
\vdots \\
V_{\pi(N)} & V'_{\pi(N)}
\end{pmatrix} := U A_2,
\]
where \( \pi(m) \) denotes a permutation of the index \( m \).
Due to the fact that permutation matrices are orthogonal (and hence unitary since they have real entries), we have \( V_A^T V_A' = V_A V_A' \) and Eq. (22) becomes \( h'_1 = 1 - 2V_A^T (V_A V_A')^{-1} V_A' \). Equation (26) now takes the form
\[
h'_1 H_{\text{low}}^{k_3, \{k_4 \}} h'_1 = \left( \frac{1_{2 \times 2} \times 0_{2 \times (N-2)}}{0_{(N-2) \times 2}} \right) U,
\]
leading us to conclude that \( H_{\text{low}}^{k_3, \{k_4 \}} \) is unaffected by the permutations of \( k_3 \) through \( k_N \).

Appendix D: Details on the charge correlation functions

In this appendix, we provide further details on how the form of Eq. (38) was obtained. We will consider the string of operators
\[
e^{i \hat{H}_t} \hat{c}_{k_1, c} \hat{c}_{k_m, c} e^{-i \hat{H}_t} \hat{c}_{k_j, c} \hat{c}_{k_j, c},
\]
which will be evaluated in our truncated space. Let \( \mathcal{B} \) denote the Hilbert space comprised of all the states entering into Eq. (5). Denoting an arbitrary state in \( \mathcal{B} \) as \( |B\rangle \), we have that
\[
\hat{c}_{k_1, c} \hat{c}_{k_j, c} |B\rangle = \langle v_{k_1} \ldots \{c_{k_1} v_{k_i} \} \ldots v_{k_N} \rangle |v_{k_1} \ldots c_{k_j} \ldots v_{k_N} |B\rangle
\]
if \( i \neq j \), and effectively that \( \langle B'| \hat{c}_{k_1, c} \hat{c}_{k_j, c} |B\rangle = \langle B'| \hat{v}_{k_1} \ldots \hat{c}_{k_j} |B\rangle \delta_{i,j} \) if \( |B'| \) is another state in \( \mathcal{B} \). To obtain frequency dependent quantities, we will employ the Lehmann representation and insert unity in the basis \( \mathcal{B} \):
\[
\sum_n \langle B'| e^{i \hat{H}_t} \hat{c}_{k_1, c} \hat{c}_{k_m, c} e^{-i \hat{H}_t} |n\rangle \langle n| \hat{c}_{k_1} \hat{c}_{k_j} |B\rangle.
\]
Given that \( |n\rangle \in \mathcal{B} \) together with the fact that \( e^{i \hat{H}_t} \) with \( \hat{H} \) in Eq. (5) induces transitions within \( \mathcal{B} \), the following simplification will arise
\[
\sum_{l,m,i,j} \sum_n \langle B'| e^{i \hat{H}_t} \hat{c}_{k_1, c} \hat{c}_{k_m, c} e^{-i \hat{H}_t} |n\rangle \langle n| \hat{c}_{k_1} \hat{c}_{k_j} |B\rangle
\]
\[
= \sum_{l,i} \sum_n \langle B'| e^{i \hat{H}_t} \hat{n}_{k_1} e^{-i \hat{H}_t} |n\rangle \langle n| \hat{n}_{k_1} |B\rangle
\]
\[
= \sum_n \langle B'| e^{i \hat{H}_t} \hat{N} e^{-i \hat{H}_t} |n\rangle \langle n| \hat{N} |B\rangle.
\]
It is important to stress that inserting unity in the way done above involves an implicit truncation to \( \mathcal{B} \).

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Note that $V_A$ is not a square matrix. $V_A(V_A^†V_A)^{-1}$ is the so-called Moore-Penrose inverse of $V_A^†$.

If $A_1$ is invertible, then $(A_1^{-1})^† = (A_1^†)^{-1}$ and from $A_1^†X = X^†A_1$ we have that $X^† = A_1^†X A_1^{-1}$. Therefore $Z^† = (A_1^{-1})^† A_1^†X A_1^{-1} = Z$.

The second identity is a consequence of the fact that $Z^2 = (Z^†)^2$.

$\Xi_c(t) = \sum_n e^{i(E_0 - E_n)t} \langle 0 | \hat{N}_c | n \rangle^2 - | \langle 0 | \hat{N}_c | 0 \rangle |^2$.

The quantity we Fourier transform is $\Xi_c(t) = \sum_n e^{i(E_0 - E_n)t} | \langle 0 | \hat{N}_c | n \rangle |^2 - | \langle 0 | \hat{N}_c | 0 \rangle |^2$.

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