

Cavity Spectroscopy for Strongly Correlated Polaritonic Systems

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Embedding materials in optical cavities has emerged as an intriguing perspective for controlling quantum materials, but a key challenge lies in measuring properties of the embedded matter. Here, we propose a framework for probing strongly correlated cavity-embedded materials through direct measurements of cavity photons. We derive general relations between photon and matter observables inside the cavity, and show how these can be measured via the emitted photons. As an example, we demonstrate how the entanglement phase transition of an embedded H_2 molecule can be accessed by measuring the cavity photon occupation, and showcase how dynamical spin correlation functions can be accessed by measuring dynamical photon correlation functions. Our framework provides an all-optical method to measure static and dynamic properties of cavity-embedded materials.

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Introduction—Controlling materials with light is a key objective of modern condensed matter research [1–5]. In addition to transient material modifications induced by ultrafast laser pulses, which have led to phenomena like light-induced superconductivity [6,7] and quantum anomalous Hall effects [8], embedding matter in optical cavities has emerged as a promising approach for controlling ground state properties by coupling to quantized vacuum fluctuations of the photon field [9–13]. Such embedding can lead to the formation of hybrid light-matter states with drastically different properties compared to free space [13], and can be controlled by designing the electromagnetic environment. Starting with the demonstration of the Purcell effect [14], the growing research area of cavity material engineering now encompasses enhanced superconductivity [15,16], the optical control of magnetic states [17–19], the breakdown and enhancement of topological protection

[20,21], and the modification of a metal-to-insulator transitions [22].

A challenge in this emerging field is the measurement of the embedded matter. In general, the embedded systems have collective modes in the regime 0.25–2.5 THz but dimensions on the order of 1 μm , placing them far below the diffraction limit of standard THz spectroscopy. Recent work has tried to bridge this gap via time-resolved on-chip THz transmission measurements [23], or by spatially confining the probe field via THz resonators [24]. However, in such setups the observed material modifications might be attributed to spurious effects other than the interaction of matter with cavity vacuum fluctuations, e.g., heating due to the cavity mirrors or the dressing of probe particles [22,25,26]. To disentangle spurious effects and the formation of strongly hybridized light-matter states, it is hence desirable to develop a more direct probe of cavity embedded materials.

A promising direction is to use the photons emitted by the cavity to extract information about the hybrid light-matter state. During its formation, the hybrid state develops finite light-matter entanglement [27,28], such that information about matter is imprinted on the cavity photons. Recently, this connection was used to relate topological properties of matter to cavity observables, in noninteracting systems and for weak light-matter couplings [29]. However, as we will demonstrate below, the relationship between light and matter observables is more general, and extends to strongly correlated systems.

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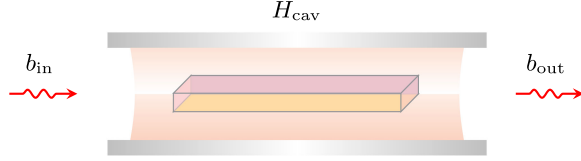


FIG. 1. Theoretical setup. An incoming photonic wave-packet $b_{\text{in}}(t)$ (can also be the vacuum) interacts with the system H_{cav} consisting of a material embedded in a cavity, and is scattered into a new state $b_{\text{out}}(t)$. The input-output formalism provides a relation between $b_{\text{in}}(t)$ and $b_{\text{out}}(t)$ in terms of cavity photon correlation functions, which are in turn related to matter correlations.

In this Letter, we propose a scheme to probe cavity embedded correlated materials by observing cavity photons through optical measurements (Fig. 1). Demonstrating how photonic cavity observables become experimentally accessible outside the cavity through a quantum optical input-output formalism [30–32], we illustrate their utility in accessing information about the embedded matter. Specifically, we show how the entanglement phase transition of an embedded H_2 molecule can be detected by probing the cavity occupation. In explaining this phenomenon, we develop a versatile framework that links photonic and matter correlations functions in strongly correlated systems, where the effective low-energy theory can be obtained via a cavity Schrieffer-Wolff transformation. This framework is not restricted to static observables, but extends to dynamical correlation functions as well. We exemplify this by showing how the dynamical dimer-dimer correlation function of a small spin cluster is probed by the intensity autocorrelation function of the cavity, providing explicit analytical expressions in the limit of weak light-matter coupling. Our framework offers a new approach for probing both static and dynamic properties of cavity-embedded correlated materials.

Light-matter Hamiltonian—To establish the measurability of cavity correlation functions, we derive a general relation between photon correlation functions inside and outside the cavity using the quantum optical input-output formalism [30–33]. To that end, assume that our setup consists of a cavity-matter system described by the general Hamiltonian (see Fig. 1)

$$H_{\text{cav}} = H_{\text{m}}(\mathbf{a}^\dagger, \mathbf{a}) + \sum_n \Omega_n a_n^\dagger a_n, \quad (1)$$

with $H_{\text{m}}(\mathbf{a}^\dagger, \mathbf{a})$ denoting the light-coupled matter system and $a_n^{(\dagger)}$ the ladder operators of the n th cavity mode with frequency Ω_n . The system is coupled to a photonic continuum $H_B = \int_\nu \omega_\nu b_\nu^\dagger b_\nu$ outside the cavity via the cavity-bath coupling

$$H_c = -i \sum_n \int_\nu \gamma_n(\nu) (a_n^\dagger b_\nu + b_\nu^\dagger a_n), \quad (2)$$

with ladder operators $[b_\nu^\dagger, b_{\nu'}] = \delta(\nu - \nu')$. The coupling of cavity and bath with $\gamma_n(\nu)$ models the leakage of cavity mirrors and allows photons to escape to the electromagnetic continuum. Using that the Hamiltonian is quadratic in the bath operators, they can be integrated out, and their influence on the dynamics of the cavity solely enters via incoming $b_{\text{in}}(t)_H$ and outgoing $b_{\text{out}}(t)_H$ wave packet operators (in the Heisenberg picture)

$$b_{\text{in}}(t)_H = \frac{1}{\sqrt{2\pi}} \int_\nu e^{-i\nu(t-t_0)} b_\nu(t_0)_H, \quad (3)$$

$$b_{\text{out}}(t)_H = \frac{1}{\sqrt{2\pi}} \int_\nu e^{-i\nu(t-t_1)} b_\nu(t_1)_H. \quad (4)$$

The reference times satisfy $t_0 < t < t_1$, such that $b_{\text{in}}^\dagger(t)_H|0\rangle$ describes an incoming wave packet and $b_{\text{out}}^\dagger(t)_H|0\rangle$ an outgoing one (Fig. 1). The distribution of $b_{\text{in}}(t)_H$ specifies the initial conditions of the impinging photons, while $b_{\text{out}}(t)_H$ characterizes the scattered state, measured after interaction with the system.

Input-output theory provides a relation between incoming and outgoing wave packet operators. For the theory to be well-behaved, the lower bound of the bath integral has to be extended to $-\infty$, viz. $\int_\nu = \int_0^\infty d\nu \approx \int_{-\infty}^\infty d\nu$. This assumes a cavity with high-Q factor ($\gamma \ll \Omega_n$) and employs the approximation that the dynamical modifications of the system due to the bath coupling are negligible compared to its internal dynamics [33].

Employing the commonly used Markov approximation $\gamma_m(\nu) = (2\pi)^{-1/2} \sqrt{\gamma_m}$ [30–33], the input-output relation reads

$$b_{\text{out}}(t)_H = b_{\text{in}}(t)_H + \sum_n \sqrt{\gamma_n} a_n(t)_H. \quad (5)$$

It directly relates the experimentally observable output operators to cavity observables. For a coherent state input $b_{\text{in}}(t)|\alpha\rangle = \alpha(t)|\alpha\rangle$, the connected expectation values $\langle\langle AB \rangle\rangle = \langle(A - \langle A \rangle)(B - \langle B \rangle)\rangle$ of the output and cavity fields are directly proportional, and for a single-mode cavity

$$\begin{aligned} & \langle\langle b_{\text{out}}(t_1)^\dagger \dots b_{\text{out}}^\dagger(t_m) b_{\text{out}}(t_{m+1}) \dots b_{\text{out}}(t_n) \rangle\rangle \\ &= \gamma^{n/2} \langle\langle a^\dagger(t_1) \dots a^\dagger(t_m) a(t_{m+1}) \dots a(t_n) \rangle\rangle. \end{aligned} \quad (6)$$

Here we have assumed (anti-)time ordering of the (annihilation) creation operators. The relation between $\langle\langle A^n \rangle\rangle$ and $\langle A^n \rangle$ is invertible and depends on $\langle A^m \rangle$, $m \leq n$ [40], such that Eq. (6) provides a prescription for measuring cavity correlation functions by probing photons outside the

cavity, e.g., achievable through homodyne and heterodyne detection [29,41,42]. The decay rate γ is a fundamental parameter of a given cavity setup. It can be determined through transmission measurements of the empty cavity as the full width at half maximum [43].

Considering a leading order expansion in γ , and a weak off-resonant input field with $\gamma|\alpha|^2/(\Delta^2 + \gamma^2/4)^{-1} \lesssim (t/U)^2$ and Δ denoting the detuning (see Ref. [33] for a detailed discussion of the estimate), the expectation values on the right-hand side of Eq. (6) can be evaluated without coupling to the bath. Under these conditions, the drive does not induce a long-lived population in the cavity, but instead probes the system with minimal disturbance. We henceforth work in this limit of weak driving [33]. The output correlation functions directly probe the cavity correlations, which in turn carry information about matter degrees of freedom.

Entanglement transition of H_2 —To illustrate this scheme on a simple system, we consider an H_2 molecule embedded in a cavity. Assuming a single effective cavity mode with linear polarization parallel to the optical dipole moment, the system is described by a single-band Hubbard model coupled to light via the Peirls substitution [33,44,45]. The Hamiltonian is given by

$$H_m = -\sum_{\langle ij \rangle \sigma} t_{ij} e^{-i\theta_{ij}} c_{i\sigma}^\dagger c_{j\sigma} - \mathbf{B} \cdot \mathbf{S} + U \sum_i n_{i\uparrow} n_{i\downarrow}, \quad (7)$$

with total spin $\mathbf{S}$, the Peirls phase in dipole approximation given by $\theta_{ij} = (e/\hbar)(\mathbf{R}_j - \mathbf{R}_i) \cdot \hat{\mathbf{A}} = g_{ij}/\sqrt{N}(a^\dagger + a)$ and the cavity ladder operators satisfying $[a^\dagger, a] = 1$. For the H_2 molecule the indices are restricted to $i, j \in \{1, 2\}$, and Eq. (7) describes electrons hopping between two ionic cores with an on-site repulsion U . The Zeeman coupled magnetic field $\mathbf{B} = B\mathbf{e}_z$ tunes the ground state between an entangled spin singlet ($S = 0, B < B^*$) and a polarized spin triplet ($S = 1, B > B^*$), with electrons localized at their respective sites. We solve the system using exact diagonalization, focusing on the regime of weak off-resonant driving [33].

This entanglement phase transition of the embedded molecule can be detected by observing the cavity photons, as shown in Fig. 2. Figure 2(a) presents the entanglement phase diagram, where the color map denotes the cavity occupation $\langle a^\dagger a \rangle$. A sharp discontinuity at $B^*(U)$ (black line), marks the transition from finite to zero entanglement entropy between the lattice sites. This behavior is further emphasized in Figs. 2(b) and 2(c) (red), which show the lattice site entanglement $\mathcal{S}_{\text{lattice}}$ and cavity occupation across the phase transition for fixed U , respectively. Thus, the cavity occupation $\langle a^\dagger a \rangle$ serves as an order parameter for the entanglement phase transition.

To gain deeper insights, we analyze the entanglement entropy $\mathcal{S}_{\text{lm}}$ between light and matter across the phase transition [Fig. 2(b) (blue)]. It exhibits the same

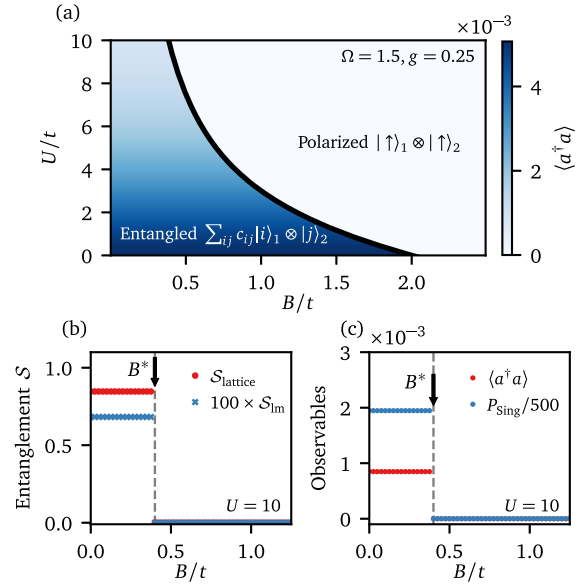


FIG. 2. Entanglement transition of the H_2 molecule. (a) Entanglement phase diagram as a function of magnetic field B and on site repulsion U . For a given U there exists a critical $B^*(U)$ (black line) where the ground state transitions from an entangled spin singlet ($B < B^*$) into a polarized triplet state ($B > B^*$). The color map indicates the cavity occupation $n = \langle a^\dagger a \rangle$ that acts as an order parameter for the transition. (b) Entanglement entropy as a function of magnetic field B across the phase transition (gray dashed; arrow). Both dimer-dimer entanglement (red) and light matter entanglement (blue) vanish in the polarized phase. (c) Photonic occupation $\langle a^\dagger a \rangle$ and singlet projection operator $P_{\text{Sing}} = \langle \mathbf{S}_1 \cdot \mathbf{S}_2 - 1/4 \rangle$ across the phase transition, illustrating that both provide valid order parameters.

discontinuous behavior as $\langle a^\dagger a \rangle$ and vanishes in the polarized phase, indicating that in this regime the ground state is a product state between light and matter $|\text{Matter}\rangle \otimes |\text{Light}\rangle$. This occurs because the polarized states span one dimensional Hilbert spaces with distinct magnetic quantum numbers $m = 0, \pm 1$, which are not connected by hopping processes and therefore cannot be dressed by the Peierls phase. As a result, the polarized state does not couple to light, explaining the discontinuous behavior of $\langle a^\dagger a \rangle$ and confirming its role as an order parameter.

Including spin-orbit interactions, which manifest as spin-flip hopping processes, or multiple bands would complicate this simple argument. However, since both effects are negligible for the H_2 dimer, they do not invalidate the observed phenomenology. Further details, including an estimate of the magnetic field required to induce the polarization transition, are provided in Supplemental Material [33].

General formalism—These arguments offer a problem-specific explanation of how, and more importantly why, the cavity can be used to probe the entanglement structure of the embedded molecule, but for a broad class of systems,

the relationship between light and matter observables can even be made explicit through the Schrieffer-Wolff (SW) transformation [46,47]. The SW transformation is a unitary block diagonalization procedure,

$$\tilde{H} = e^S H e^{-S} = H + [S, H] + \frac{1}{2!} [S, [S, H]] + \dots, \quad (8)$$

that eliminates the coupling between energetically separated low- and high-energy sectors of the Hilbert space using the antiunitary SW operator $S = -S^\dagger$. These sectors are represented by projectors $\mathcal{P}$ (low energy) and $\mathcal{Q}$ (high energy) with $\mathcal{P} + \mathcal{Q} = 1$ and $\mathcal{P}\mathcal{Q} = 0$. The new basis is defined by $\mathcal{P}\tilde{H}\mathcal{Q} = 0 = \mathcal{Q}\tilde{H}\mathcal{P}$, viz. the condition that there is no coupling between the sectors, defining an effective low-energy theory.

As a basis transformation, it does not only modify the Hamiltonian, but also leads to a corresponding dressing $A_{\text{eff}} = e^S A e^{-S}$ of observables. Because S depends on the matter degrees of freedom, this introduces an explicit dependence of the effective photonic observables on matter correlation functions through the correction introduced by S . To illustrate this further, we use the paradigmatic Hubbard model in Eq. (7) as a guiding example.

The SW operator S can in general not be determined in closed form, and is instead computed as a perturbation expansion, in this case in $t/U \ll 1$. The coupling between low- and high-energy sectors, spanned by states with doubly occupied sites, is eliminated order by order. By expanding the Hamiltonian equation (7) in the photon number basis, we obtain the leading order SW operator [33]

$$S^{nn'} = - \left(\mathcal{P} H_t^{nn'} \mathcal{Q} \frac{1}{U + \Omega(n' - n)} + \mathcal{Q} H_t^{nn'} \mathcal{P} \frac{1}{-U + \Omega(n' - n)} \right) + \mathcal{O}(t/U)^3, \quad (9)$$

where $H_t^{nn'} = \langle n | H_t | n' \rangle$ is the hopping Hamiltonian evaluated between cavity number states. Using Eq. (9), the effective low energy theory of Eq. (7) takes the form of a photon dressed Heisenberg model [19,33,48],

$$H_{\text{m,eff}} = \sum_{\langle ij \rangle} J_{ij}(a^\dagger, a) \left(\mathbf{S}_i \cdot \mathbf{S}_j - \frac{1}{4} \right) + \mathcal{O}(t/U)^3, \\ J_{ij}^{nn'} = 2|t_{ij}|^2 \sum_k \phi_{ij}^{nk} \phi_{ji}^{kn'} \left(\frac{1}{U + \Omega(k - n)} + [n \leftrightarrow n'] \right), \quad (10)$$

with the matrix element $\phi_{ij}^{nm} = \langle n | e^{-i\theta_{ij}} | m \rangle$ of the Peirls phase. Note that this expansion is only valid for $U \neq n\Omega$, $n \in \mathbb{Z}$, but already becomes unreliable close to these resonances due to diverging energy denominators [33].

We are in particular interested in how cavity observables are dressed in the low-energy theory. To explore this, we consider the expectation value of the cavity occupation $n = a^\dagger a$. We find [33]

$$\langle n_{\text{eff}} \rangle = \langle n \rangle + \sum_{\langle ij \rangle} \mathcal{N}_{ij} \langle \mathbf{S}_i \cdot \mathbf{S}_j - 1/4 \rangle + \mathcal{O}(t/U)^4, \quad (11)$$

where the photon dependent prefactor is defined as $\mathcal{N}_{ij} = -(U/2) \sum_k \phi_{ij}^{0k} \phi_{ji}^{k0} k (U + \Omega k)^{-2} \in \mathbb{R}$. Note that the bare contribution gives $\langle n \rangle \in \mathcal{O}(t/U)^4$, since the SW ground state wave function has $|\psi\rangle = |\psi_m\rangle \otimes |n=0\rangle + \mathcal{O}(t/U)^2$, a universal feature for Peirls coupled tight-binding models [33]. Equation (11) provides a direct connection between the cavity occupations and matter correlations, which is straightforward to generalize to other photon observables; see Supplemental Material [33].

For the Hubbard dimer discussed in the previous section, we have $\langle n_{\text{eff}} \rangle \sim \langle \mathbf{S}_1 \cdot \mathbf{S}_2 - 1/4 \rangle$, which corresponds to the singlet projection operator. It gives a finite value for singlet states and zero for triplet states [Fig. 2(c) (red)], offering an alternative and generalizable explanation of why the cavity occupation can be used to diagnose the entanglement phase transition discussed earlier.

Measuring dynamical correlations—The relationship between light and matter extends beyond static observables to dynamic correlation functions, which are routinely used to probe properties of matter [40]. Specifically, as we will show below, dynamical photon correlation functions $C(t) = \langle \mathcal{O}_1(t) \mathcal{O}_2 \rangle$ are directly linked to dynamical matter correlators. In the case of autocorrelation functions, where $\mathcal{O}_1 = \mathcal{O}_2^\dagger$, they also provide information about the selection rule resolved excitation spectrum. This becomes evident in their frequency representation,

$$C(\omega) = \int_0^\infty dt e^{i(\omega + i\eta)t} C(t) = \left\langle \mathcal{O}_1 \frac{i}{\omega + i\eta - \Delta H} \mathcal{O}_2 \right\rangle,$$

where $\Delta H = H - E_{\text{GS}}$ is the Hamiltonian relative to the ground state energy. For autocorrelation functions, $C(\omega)$ probes the resolvent operator in state $\mathcal{O}_1 | \text{GS} \rangle$, effectively capturing excitations generated when $\mathcal{O}_1$ acts on the ground state.

To illustrate the connection to matter and to provide a specific example for the measured correlation functions, we once again turn to the cavity embedded Hubbard model Eq. (7), focusing on the intensity-intensity correlation function $N_2(t) = \langle n(t)n \rangle$. Downfolding to the low-energy theory followed by a perturbative expansion for weak light-matter interactions, we find that the dominant contribution probes a light-matter interaction modulated dimer-dimer correlation function of the matter [33]

$$\begin{aligned}
 \langle n(t)n \rangle_{\text{eff}} \sim & \left\langle \left[\sum_{\langle i,j \rangle} \frac{g_{ij}^2}{N} J_{ij} (\mathbf{S}_i \cdot \mathbf{S}_j - 1/4) \right] (t) \right. \\
 & \times \left. \left[\sum_{\langle k,l \rangle} \frac{g_{kl}^2}{N} J_{kl} (\mathbf{S}_k \cdot \mathbf{S}_l - 1/4) \right] \right\rangle \\
 & + N_{2,\text{doub}}(t) + \mathcal{O}[g^6, (t/U)^4], \quad (12)
 \end{aligned}$$

with $g_{ij} \sim \mathbf{A} \cdot (\mathbf{R}_i - \mathbf{R}_j)$ [see Eq. (7)]. The low-energy dynamics, governed by the dimer-dimer correlation function in Eq. (12), is superimposed by the fast high-energy oscillations $N_{2,\text{doub}}(t)$ due to doublon-polariton excitations, which occur at a higher energy scales and are less prominent in shaping the overall behavior [49]. Consequently, we focus on the low-energy dynamics moving forward.

For uniform light-matter interactions, where $g_{ij} \equiv g$ as in 1D systems, the dimer operators in Eq. (12) commute with the Hamiltonian leading to trivial low-energy dynamics. For inhomogeneous light matter interactions, on the other hand, where $g_{ij} \neq g$, Eq. (12) probes nontrivial, dynamical dimer-dimer correlation functions. Additionally, it captures information about excitations generated by the dimer operator acting on the ground state. Since these operators commute both with $\mathbf{S}^2$ and S_z , the excitations correspond to states with $\Delta S = \Delta m = 0$.

Inhomogeneous light-matter couplings can arise from nontrivial geometries of the model. A minimal example is the 2×2 site Hubbard plaquette, where the lattice sites form a square and an electromagnetic field is applied along one of the bonds. This model geometry is depicted in the inset of Fig. 3, which shows $N_2(\omega)$ calculated from a full electronic model (black), a low-energy spin model (blue), and the perturbative in light-matter coupling formula [Eq. (12) (red dashed)]. All approaches show excellent agreement in the low-energy sector. In addition to the zero frequency peak, corresponding to no excitations, we observe a low-energy magnetic excitation at ΔE_{mag} , the only low-energy excitation of the model satisfying the desired selection rules. The excellent agreement between the full low-energy spin and perturbative approach, establishes $N_2(\omega)$ as a probe for the dimer-dimer correlation functions specified in Eq. (12). For additional details and the analysis of alternative photonic observables like $g^{(1)}$, as well as the effect of spin-orbit interactions, see Supplemental Material [33].

To read out matter correlations with a cavity, a possible experimental protocol is the following: first, the desired photonic quantity is measured for an empty cavity under an external drive. Second, the measurement is repeated for a cavity with an embedded material. The leading order contribution to the difference signal of these measurements is the correlation functions analyzed above [33]. As our scheme is based on a perturbative expansion in t/U

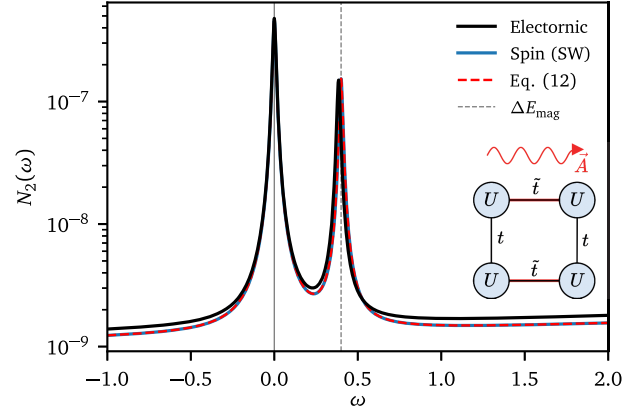


FIG. 3. Dynamical cavity correlation function $N_2(\omega)$ for a 2×2 site Hubbard plaquette at $U = 20$, $\Omega = 2.35$, $g = 0.15$, and $\eta = 0.01$ with an electromagnetic field $\mathbf{A} = A\mathbf{e}_x$, resulting in photon dressing of hoppings in x direction only. The exact electronic result (black) is compared with the approximate low energy spin theory (blue) [Eq. (10)], showing good agreement at low energies. The plot illustrates the zero photon peak at $\omega = 0$ and the selection rule allowed ($\Delta S, \Delta m = 0$), singlet excitation at $\omega = \Delta E_{\text{mag}}$ (gray dashed). The perturbative linear response expansion Eq. (12) (red dashed) aligns closely with the full spin result, illustrating how the photonic $N_2(\omega)$ probes the geometry modulated dimer-dimer matter correlation function.

and γ , it is expected to be effective for strongly correlated compounds in moderate to high finesse cavities ($Q \sim 10^3$ – 10^6), subject to weak off-resonant drives. We estimate it holds for drive intensities below $I \sim 1 \text{ W m}^{-2}$ placing it at the low end of contemporary experimental capabilities [33].

Conclusions—We developed a framework to probe correlated, cavity-embedded materials by measuring correlation functions of the cavity photons, which we connected to matter correlation functions through a general scheme relying only on the down-folding to an effective low-energy theory. This optical method is therefore widely applicable, and enables the exploration of both static and dynamic properties of cavity-embedded materials, overcoming the diffraction limit of conventional spectroscopic techniques. Although here the cavity was used mainly as a probe of matter, an exciting possibility is to combine this framework with the study of systems where the cavity also induces a phase transition of the embedded material [15,18,20,22,50]. This allows to probe the material via correlations of the same cavity photons responsible for the modified ground state. As our framework can be extended to non-classical input states, another interesting application is the study of topological phases of matter like fractional quantum Hall states or quantum spin liquids, which are known to not be accessible with classical optical probes. Such nonclassical cavity spectroscopy opens a new pathway towards probing highly entangled cavity-embedded materials via optical measurements.

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