Certifying Multimode Light-Matter Interaction in Lossy Resonators

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Quantum models based on few-mode master equations have been a central tool in the study of resonator quantum electrodynamics, extending the seminal single-mode Jaynes-Cummings model to include loss and multiple modes. Despite their broad application range, previous approaches within this framework have either relied on a Markov approximation or a fitting procedure. By combining ideas from pseudomode and quasinormal mode theory, we develop a certification criterion for multi-mode effects in lossy resonators. It is based on a witness observable, and neither requires a fitting procedure nor a Markov approximation. Using the resulting criterion, we demonstrate that such multi-mode effects are important for understanding previous experiments in x-ray cavity QED with Mössbauer nuclei and that they allow one to tune the nuclear ensemble properties.

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In the study of quantum matter interacting with light fields, an important theoretical [1,2] and experimental paradigm [3,4] is the coupling to a single cavity mode [5]. More recently, regimes beyond single-mode light-matter interaction have moved into the focus of attention. In particular, multimode resonator quantum electrodynamics has opened new possibilities for many-body quantum systems [6], for example, by enabling the tuning of effective interactions [7], and for quantum information processing [8,9]. Also for single atoms, qualitatively new dynamics can be realized when multiple modes participate, such as in the multimode strong coupling regime [10], where the extreme light-matter coupling bridges the free spectral range between the modes [11,12].

For many platforms, the realization of extreme coupling strengths or the required mode control remain technically challenging. However, multimode features may also appear in the opposite regime of open and absorptive resonators, e.g., if the mode widths exceed their frequency spacing [13–15]. Interestingly, strong losses do not necessarily represent an obstacle, but may instead cause fascinating phenomena, such as for lasers [16–18] and in the context of non-Hermitian physics [19–24]. Within quantum optics and particularly nanophotonics, where complex mode structures can be engineered [25] and losses are often sizable [26], non-Hermitian effects have attracted considerable interest, with potential applications ranging from quantum plasmonics [27] to cavity-controlled materials [28,29] and chemistry [30–34].

Another example is x-ray cavity QED (cQED) with ensembles of Mössbauer nuclei [35]. It uniquely combines the possibility of high-precision x-ray spectroscopy using the extreme quality factors of the involved nuclear resonances with cavities, which are restricted to the lossy regime due to the low refractive index contrast at hard x-ray energies. As a result, single-mode models fail to quantitatively describe the nuclear spectra [36,37] and standard fitting procedures for the multimode case require heuristic extensions [38]. This regime is of practical importance, since such cavity-nuclei systems facilitate the engineering of artificial few-level quantum systems, which are otherwise inaccessible at hard x-ray energies [36,39–48].

From a theory perspective, these regimes of cQED are particularly challenging, due to an interplay of strong light-matter coupling and large cavity losses. The latter implies a breakdown of standard Jaynes-Cummings (JC) models [13,49,50], which feature a few discrete cavity modes coupled to an external bath [51,52]. Similar issues with such models are encountered at ultrastrong coupling [53–56]. While many alternative approaches to describe non-Markovian open quantum systems exist [57,58], including various types of master equations only involving the matter degrees of freedom [59–62], quantum stochastic approaches [52,63,64], and direct treatments of the continuum [10,65–68], such methods remain challenging particularly for complex matter systems. As a result, much effort has been invested into finding suitable generalizations of and a rigorous theoretical basis for few-mode models.

Current approaches to this problem fall into three distinct categories. On the one hand, the recent quantization of quasinormal modes [69] has promoted techniques from semiclassical and perturbative light-matter interaction theory [15,70] to the quantum level. While this approach allows for insights into absorptive resonator physics [71–76], its applicability is limited by the use of Markov approximations [77], which break down for high loss regimes [13,51]. Similarly, *ab initio* system-bath theory [51,78] allows one to derive exact few-mode Hamiltonians [79], but generally requires a Markov approximation when transferring to a master equation. On the other hand, the concept of pseudomodes [80–83], its non-Hermitian variants [84–87], and the related chain mappings [83,88–90] from the theory of open quantum systems are designed to reduce the continuum bath to discrete modes with minimal or no Markov approximation at all. This idea can be seen as a special discrete mode basis [82,91] where the system-bath coupling [13,51,79,81] is frequency independent. However, current methods to construct such exactly Markovian descriptions rely on fitting procedures to match the bath properties [92], such as the spectral density in the case of cQED [91].

In this Letter, we combine these previous approaches to develop a certification criterion for multimode quantum effects that neither relies on a Markov approximation nor a fitting procedure.

Our method employs pseudomodes-based few-mode (pFM) models, which do not require a Markov approximation, but in general involve *a priori* unknown parameters. We construct a witness observable to certify the necessity of multiple modes and their interactions in the pFM model, and to classify different cases of multimode effects. By establishing a connection to classical quasinormal mode (cQNM) expansions [15,93] of the witness observable, the need for fitting model parameters is eliminated. Within x-ray cQED, we apply our certification criterion to unambiguously identify the previously measured collective nuclear Lamb shift [36] as a multimode effect, resolving a puzzle raised by earlier models [38,44]. We further show how the different types of multimode effects allow one to invert the sign of this shift by engineering suitable cavity environments.

Revisiting the single-mode case.—We start by discussing features of a standard single-mode pFM model of a generic cavity, see Fig. 1(a). Without atom, the frequency-dependent reflection coefficient is [94]

$$r_{\mathrm{cav}}(\omega) = 1 - 2\pi i \frac{|\kappa_R|^2}{\omega - \omega_1 + i\frac{\kappa}{2}}, \qquad (1)$$

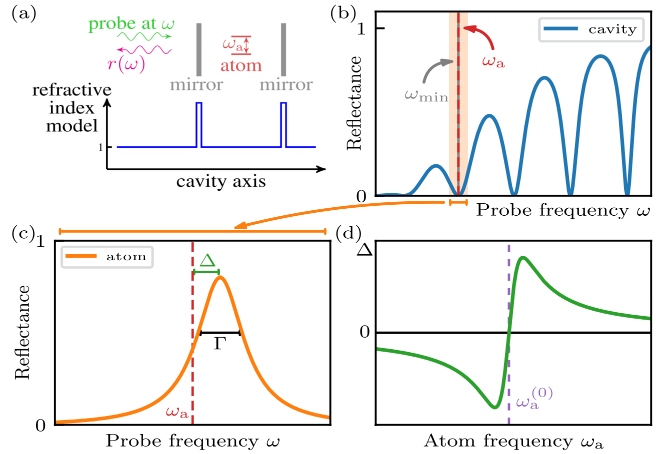

FIG. 1. Model setup and witness observable. (a) Model cavity coupled to a two-level atom at its center. (b) Reflectance for the case of spectrally broad cavity modes and a narrow atomic resonance. (c) shows a magnification of (b) around the atomic resonance. (d) Δ as a function of the atomic transition frequency.

where ω is the probing light frequency, κ_R the coupling rate in and out of the cavity mode of frequency ω_1, and κ the total cavity loss rate. We denote the relevant minimum of this empty-cavity reflectance ($|r_{\mathrm{cav}}|^2$) as ω_{min}. An example reflectance (featuring multiple modes) is shown in Fig. 1(b).

Coupling a single two-level atom with rate g to the cavity mode leads to a lossy JC model [1,3]. The linear reflection spectrum is a Lorentzian on the cavity background [79,94,103]

$$r(\omega) = r_{\mathrm{cav}}(\omega) - 2\pi i \frac{(\kappa_R^{(\mathrm{int})}(\omega))^2}{\omega - \omega_{\mathrm{a}} - \Delta(\omega) + i\Gamma(\omega)/2}, \qquad (2)$$

where the Lorentzian parameters are frequency dependent to ensure applicability at strong coupling. Specifically, $(\kappa_R^{(\mathrm{int})}(\omega))^2 = |\kappa_R g|^2/(\omega - \omega_1 + i\kappa/2)^2$ and the real quantities $\Delta(\omega)$ and $\Gamma(\omega)$ together define the function

$$\delta(\omega) \coloneqq \Delta(\omega) - i\frac{\Gamma(\omega)}{2} = \frac{gg^*}{\omega - \omega_1 + i\frac{\kappa}{2}}. \qquad (3)$$

At weak coupling, $\Delta = \Delta(\omega_{\mathrm{a}})$, $\Gamma = \Gamma(\omega_{\mathrm{a}})$ correspond to the Lamb shift and Purcell enhanced linewidth of the atom, respectively [Fig. 1(c)]. Beyond weak coupling, these quantities are closely related to bath correlation functions, e.g., $\Gamma(\omega)$ is proportional to the spectral density [91]. We further denote the atomic resonance frequency at which $\Delta = 0$ as $\omega_{\mathrm{a}}^{(0)}$.

By inspection of Eqs. (1)–(3), we find that the minimum of the empty cavity reflectance ω_{min}, the zero of the frequency shift $\Delta(\omega_{\mathrm{a}}^{(0)}) = 0$, the maximum of the line width broadening Γ, and the real part ω_1 of the pole location of $\Delta(\omega)$ in the complex frequency plane all coincide at the cavity mode frequency [$\omega_{\rm min} = \omega_{\rm a}^{(0)} = \omega_1$]. As one consequence, single-mode cavity models do not allow for nonzero shifts Δ at the minimum $\omega_{\rm min}$ of the cavity background $|r_{\rm cav}|^2$, which is at odds with the experimental observation of such a nonzero shift reported in Fig. 3 of [36]. Motivated by this, we focus the following discussion on the Lamb shift Δ as a *witness observable*, which we demonstrate to provide diagnostic information about multimode effects.

Multimode case.—Next, we generalize to the multimode case. We employ a pFM model of a recently developed form [91,94], which applies to complex and absorptive resonators described by macroscopic QED [104]. It is given by the master equation [91]

$$\dot{\rho} = -i[H;\rho] + \sum_i \frac{\kappa_i}{2}(2\hat{a}_i\rho\hat{a}_i^\dagger - \hat{a}_i^\dagger\hat{a}_i\rho - \rho\hat{a}_i^\dagger\hat{a}_i), \quad (4)$$

where $H = H_{\rm cav} + H_{\rm atom}$, with an interacting cavity mode Hamiltonian $H_{\rm cav} = \sum_{ij}\omega_{ij}\hat{a}_i^\dagger\hat{a}_j$, and the JC interaction $H_{\rm atom} = (\omega_{\rm a}/2)\hat{\sigma}^z + \sum_i[g_i^*\hat{a}_i\hat{\sigma}^+ + {\rm H.c.}]$. Equation (3) then generalizes to [79,94]

$$\delta = \underline{g}^\dagger\left[\omega_{\rm a} - \underline{\underline{\omega}}_{\rm cav} + \frac{i}{2}\underline{\underline{\kappa}}\right]^{-1}\underline{g}, \quad (5)$$

where $(\underline{g})_i = g_i$, $(\underline{\underline{\omega}}_{\rm cav})_{ij} = \omega_{ij}$, and $(\underline{\underline{\kappa}})_{ij} = \kappa_i\delta_{ij}$ are the multimode couplings, cavity mode interactions, and decay terms, respectively, in matrix-vector notation.

Importantly, despite the simple form of Eq. (4), this model does not employ a Markov approximation [91] due to its use of the pseudomodes concept [82,86,105]. In contrast, the quantized QNM approach in [69] requires an explicit Markov and related approximations [77], such that the resulting master equations are not necessarily the same as pFM models. However, obtaining the parameters of the latter currently requires fitting procedures [91,92], even in the ideal case when the cavity geometry is known without experimental uncertainties.

From few-mode models to quasinormal mode expansions.—To certify and categorize multimode effects in the pFM approach, we diagonalize Eq. (5) as

$$\delta = \sum_i \frac{\tilde{g}_i^*\tilde{g}_i}{\omega_{\rm a} - \tilde{\Omega}_i + i\frac{\tilde{\kappa}_i}{2}}, \quad (6)$$

where the diagonal basis parameters relate to the bare mode parameters in Eq. (5) by an invertible transformation matrix [14,94]. Note that here we assumed that the diagonalization exists and comprises only simple poles. A more general treatment is required, e.g., in the presence of exceptional points [20,21].

In order not to rely on a fit, we use an alternative expression from cQNM theory for the level shift in terms of the cavity's classical Green's function $\mathbf{G}$, given by $\delta = -\mu_0\omega_{\rm a}^2\mathbf{d}^*\cdot\mathbf{G}(\mathbf{r}_{\rm a},\mathbf{r}_{\rm a},\omega_{\rm a})\cdot\mathbf{d}$ [104,106–108] where $\mathbf{d}$ is the transition's dipole moment vector. $\mathbf{G}$ can then be expanded using a pole expansion [15,94,109,110], such that

$$\delta = \sum_i \frac{r_i}{\omega_{\rm a} - \tilde{\omega}_{{\rm pole},i}}. \quad (7)$$

Such expansions are known as quasinormal mode expansions in the electromagnetism literature, which have been studied extensively [15,70] and are of the same form as the diagonalized pFM expansion Eq. (6). We can therefore use cQNM expansions of the witness observable as an intermediate step to draw conclusions about the pFM model.

Certification of multimode effects.—This comparison leads to three distinct cases of multimode effects, which deviate in different ways from the single-mode formula Eq. (3). We illustrate each case by its action on the witness observable $\Delta(\omega_{\rm a})$ (see Fig. 2) and implications for the resulting pFM master equation.

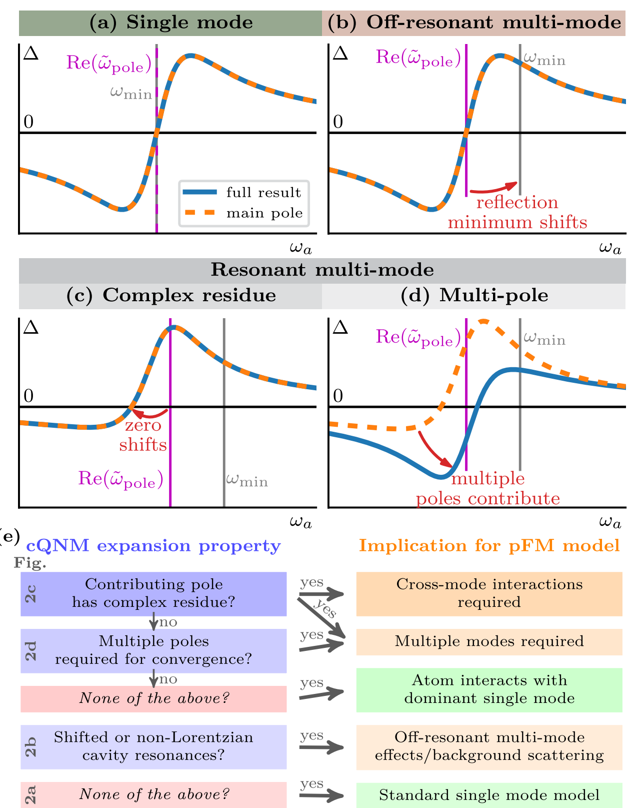

FIG. 2. Certification and classification of multimode effects, illustrated via the Lamb shift Δ as in Fig. 1(d). (a)–(d) Categories of multimode effects with their corresponding features of the witness observable Δ. (e) Decision tree for certifying multimode effects using the cQNM expansion of the witness observable.

In the fully single-mode case [Fig. 2(a)], already the main pole provides a good quantitative description, with the aforementioned frequency coincidence $\omega_a^{(0)} = \mathrm{Re}(\tilde{\omega}_{\mathrm{pole}}) = \omega_{\mathrm{min}}$.

The first condition for the single-mode case is thus that a single main pole has to be sufficient to achieve convergence of the pole expansion in a relevant frequency range. The opposite case, where multiple poles are required for good convergence, we will refer to as *multipole effects* [Fig. 2(d)].

Second, we also require real-valued residues, since the residue $|g|^2$ in the single-mode case Eq. (3) is real. *Complex-residue effects* appear if a pole has a non-negligible imaginary part of its residue [87,88,111], breaking the frequency coincidence as illustrated in Fig. 2(c). The presence of complex residues is further a direct indicator of *cross-mode interactions* [13,78,91], unless non-Hermitian interaction Hamiltonians are employed [84,85,87]. Even the case of single QNM physics may therefore require multiple modes in the pFM theory, implying that the two are distinct concepts.

Both complex-residue and multipole effects are *resonant effects*, since they directly affect the cavity-modified properties Δ and Γ of the atom, and they are *multimode effects*, since any pFM single-mode model fails to capture them. In addition, single-mode models may fail even in the absence of the atom due to *off-resonant multimode effects*, if the empty-cavity reflectance deviates from its single-mode form Eq. (1) due to spectral overlap between cavity modes [Fig. 2(b)]. On the level of the pFM model, these off-resonant effects can also be understood as a manifestation of background scattering contributions in input-output theory [79]. Similar issues are encountered in cQNM theory, where various approaches to expand the scattering matrix have been investigated recently [112–115].

Analyzing the properties of the cQNM expansion therefore allows us to certify and distinguish multimode effects in pFM models [see Fig. 2(e)]. We note that while our criterion focuses on certifying multimode effects resulting from large losses and overlapping modes, the pFM master equation fully applies at strong coupling [79,91,94]. The witness observable Δ can be interpreted as the line shift at weak coupling, but also has physical meaning beyond this regime [10].

Multimode effects in Fabry-Pérot-like cavities.—Next, we explore the relevance of the different multimode effects by analyzing a simple model reminiscent of archetype Fabry-Pérot cavities [Fig. 1(a)]. The cavity of length L is modeled via a piecewise constant one-dimensional refractive index comprising vacuum with two mirror layers of refractive index n_{mirror} and thickness $t_{\mathrm{mirror}} = L/100$.

Results are shown in Fig. 3. Panel (a) quantifies resonant multimode effects via the displacement of the zero $\omega_a^{(0)}$ of $\Delta(\omega_a)$ from the main cQNM pole position $\mathrm{Re}(\tilde{\omega}_{\mathrm{pole}})$ as a

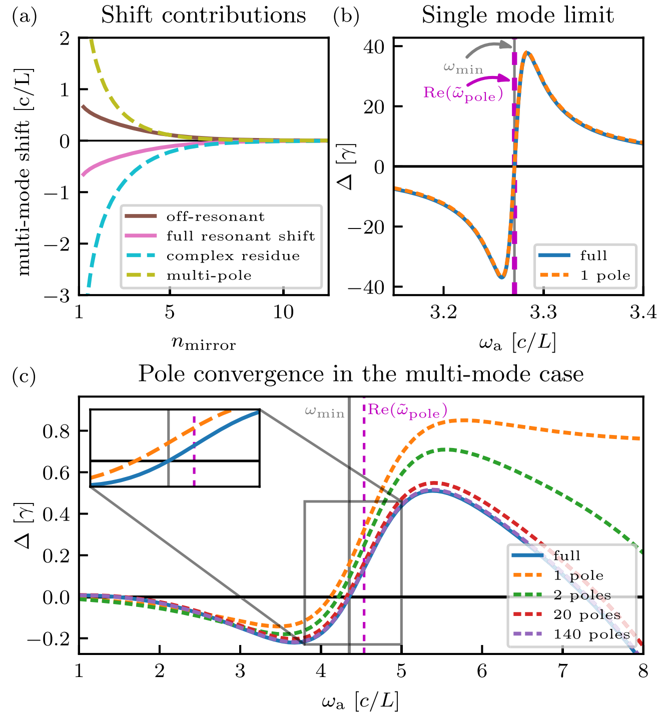

FIG. 3. Transition from the single-mode to the multimode regime in Fabry-Pérot-like cavities. Panel (a) quantifies the shift contribution of each multimode effect to the zero position of $\Delta(\omega_a)$ (see legend and text). (b) Cavity-induced energy shift Δ in the single-mode limit ($n_{\mathrm{mirror}} = 20$). (c) Corresponding results in a multimode case ($n_{\mathrm{mirror}} = 4$). The inset shows the full and single-pole results around the pole location to make their shifts visible. γ is the free space radiative linewidth.

function of n_{mirror}, and off-resonant multimode effects via the shift of $\mathrm{Re}(\tilde{\omega}_{\mathrm{pole}})$ from ω_{min}. For the single-mode case in high-quality resonators with well-isolated resonances ($n_{\mathrm{mirror}} \gg 1$), all shifts vanish. As expected, towards highly leaky resonators featuring overlapping modes, multimode effects become increasingly relevant. Panels (b),(c) show $\Delta(\omega_a)$ for a single-mode (b, $n_{\mathrm{mirror}} = 20$) and a multimode (c, $n_{\mathrm{mirror}} = 4$) scenario. In (b), one cQNM pole is sufficient to model the system, and no zero shift of Δ appears. In contrast, the multimode case in (c) requires many poles for convergence and features significantly complex residues.

Multimode level shift design.—Finally, we explore the multimode effects outlined above in the concrete platform of thin-film x-ray cQED with Mössbauer nuclei, which has recently attracted considerable theoretical [38,44,46,116–118] and experimental [36,37,39–43] attention. Similar setups with electronic processes are starting to be explored [119–121]. In these cavities, the atom is replaced by a thin layer containing an ensemble of Mössbauer nuclei. Because of the low refractive-index contrast, the cavities are probed at grazing incidence and generally feature large losses and overlapping modes [38]. In the experimentally relevant low-excitation regime, the cavity-nuclei system

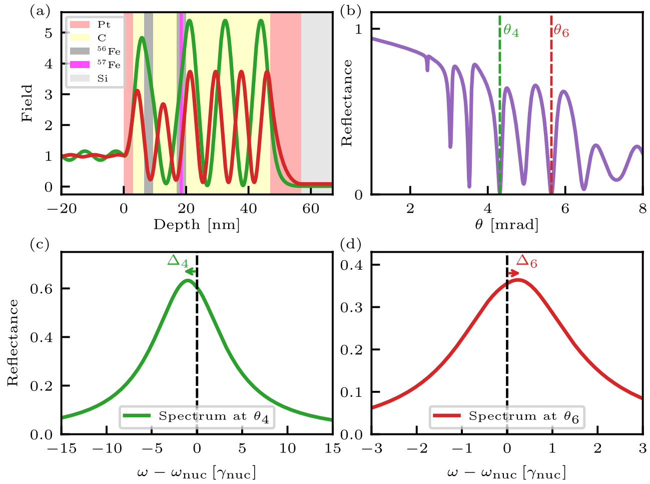

FIG. 4. Design of multimode level shifts. (a) Cavity geometry and off-resonant field distributions in the fourth (θ_4, green) and sixth (θ_6, red) cavity mode. The cavity structure is Pt(3 nm)/C(3.5 nm)/^{56}Fe(3 nm)/C(7.5 nm)/^{56}Fe(1 nm)/^{57}Fe(1 nm)/^{56}Fe(1 nm)/C(27 nm)/Pt(10 nm)/Si. (b) Off-resonant cavity reflectance as a function of incidence angle showing the overlapping mode structure. (c),(d) Nuclear spectra at θ_4, θ_6, demonstrating the reversed energy shift in the two critically coupled minima.

can be approximated as a few-level quantum system [44,46,94,122], comprising the collective ground state and excitonic single-excitation states coherently spread over the nuclei. The latter couple via transverse cavity modes at a given parallel wave vector [46]. Such cavities thus form a promising platform to implement quantum optical schemes at hard x-ray energies, which are otherwise unavailable due to the lack of suitable x-ray driving fields. For this purpose, the possibility to control the quantum optical parameters via the cavity environment is key [35,45]. From an experimental point of view, operation at a critically coupled minimum of the empty-cavity reflectance ω_{min} is highly desirable, in order to suppress the spectrally broad electronic scattering background. In this configuration, however, the single-mode case severely impedes the parameter control due to the strict coincidence of spectral features discussed earlier.

The calculations in Fig. 4 show that multimode effects provide a physical mechanism to overcome this challenge. The cavity structure shown in panel (a) features overlapping cavity resonances visible in the off-resonant cavity reflectance shown in (b). Panels (c),(d) show the resulting spectra of the nuclear resonances for driving at the two modes θ_4 and θ_6, respectively, corresponding to critically coupled minima. The cavity structure is designed such that the multimode effects lead to opposite signs of the respective collective nuclear Lamb shifts Δ. This sign flip demonstrates that tuning the mode environment through the cavity geometry allows one to deliberately alter the effective level scheme of the nuclear ensemble in a qualitative way, as it is required, e.g., to satisfy delicate detuning conditions in more complex multilevel systems [38,39,123]. A closer inspection [94] shows that the fourth mode in this cavity mainly features off-resonant multimode effects, while the sixth mode is influenced by resonant multimode effects. An analogous analysis [94] reveals that already the first experimental observation of the collective Lamb shift in a similar cavity [36] relies on an off-resonant multimode effect, illustrating the importance for the interpretation of spectroscopic observables, which has been a puzzle within previous models [38,44]. We note that while the resonance shifts are smaller than the superradiant line width, they are measurable [36,37] and understanding whether they arise due to cavity or material effects is important for the interpretation of experimental spectra, and for the design of more advanced quantum optical level schemes using tailored cavity reservoirs [47,48], where generalized witness observables may be constructed. More pronounced signatures of such multimode effects appear in multilayer systems [39], where the spectral interference strongly depends on multimode contributions [38].

The code used in this Letter can be accessed at [124] and an open-source PYTHON library for parts of these computations has been made available at [125].

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