Quantization of Quasinormal Modes for Open Cavities and Plasmonic Cavity Quantum Electrodynamics

Sebastian Franke,[1,*] Stephen Hughes,[2] Mohsen Kamandar Dezfouli,[2] Philip Trøst Kristensen,[3] Kurt Busch,[3,4] Andreas Knorr,[1] and Marten Richter[1]

[1]*Technische Universität Berlin, Institut für Theoretische Physik, Nichtlineare Optik und Quantenelektronik, Hardenbergstraße 36, 10623 Berlin, Germany*

[2]*Department of Physics, Engineering Physics and Astronomy, Queen's University, Kingston, Ontario K7L 3N6, Canada*

[3]*Humboldt-Universität zu Berlin, AG Theoretische Optik und Photonik, 12489 Berlin, Germany*

[4]*Max-Born-Institut, 12489 Berlin, Germany*

(Received 4 October 2018; published 29 May 2019)

We introduce a second quantization scheme based on quasinormal modes, which are the dissipative modes of leaky optical cavities and plasmonic resonators with complex eigenfrequencies. The theory enables the construction of multiplasmon or multiphoton Fock states for arbitrary three-dimensional dissipative resonators and gives a solid understanding to the limits of phenomenological dissipative Jaynes-Cummings models. In the general case, we show how different quasinormal modes interfere through an off-diagonal mode coupling and demonstrate how these results affect cavity-modified spontaneous emission. To illustrate the practical application of the theory, we show examples using a gold nanorod dimer and a hybrid dielectric-metal cavity structure.

DOI: 10.1103/PhysRevLett.122.213901

Open cavity systems such as micropillars [1–3], photonic crystal cavities [4,5], metal resonators [6–9], and hybrid metal-dielectric cavities [10,11] are of interest for fundamental quantum optics and emerging technologies, including two-photon lasing [12], spasing [13,14], and quantum information processing [15]. In such systems, one goal of quantum optics is to describe the electric field as an operator associated with the creation and annihilation of photons or plasmons. One important example is the "modes of the Universe" approach [16–20]; another is the Jaynes-Cummings (JC) model [21,22], which describes multiphoton interactions between a quantized cavity mode and quantum emitters such as molecules, two-level atoms, or quantum dots [23–25]. A major restriction of the JC model is that the quantization procedure starts with the quantized modes of a closed cavity made from a lossless material. Such modes have real eigenfrequencies and can be normalized in a straightforward way [22]. For cavities with very low radiative loss, as usually quantified by a high quality factor Q, dissipation is often modeled through the second-order system-reservoir theory [22] or quantum stochastical differential equation methods [26,27]. For low Q cavities, however, or in the case of metallic resonators, such approaches are ambiguous, and the concept of a closed cavity is clearly problematic.

While much progress has been made by treating the dissipation through the electromagnetic environment surrounding high Q cavities as a bath, very little has been done in terms of quantized dissipative modes, especially in plasmonics. One heuristic approach has been to assume a phenomenological dissipative JC model, which for metals assumes parameters normally used for cavity modes and where the total plasmon loss is treated phenomenologically [28]. Other notable approaches include the use of a projection operator applied to dielectric coupled-cavity systems [29] or treating the electromagnetic environment by expansions in terms of pseudomodes [30,31] or quasinormal modes (QNMs) [32]. The QNMs offer tremendous insight and efficiency for describing electromagnetic scattering and semiclassical light-matter interaction [33–38]. Typically, only a few QNMs and often just one QNM is needed, and it has recently been recognized that a quantum theory based on QNMs would represent a "major milestone in quantum optics" [39]. While some progress in this direction has been made for one-dimensional dielectric structures [40,41], these particular approaches do not lead to the construction of Fock states, which forms the natural basis for studying multiplasmon or multiphoton dynamics.

In this Letter, we present a rigorous quantization scheme for leaky optical cavities and plasmonic resonators using QNMs, which—in contrast to typical approaches to lossy materials—enables the construction of Fock states, as illustrated in Fig. 1. We solve the main challenges related to the non-Hermitian nature of the problem by introducing a symmetrization scheme for creation and annihilation operators associated with the resonator QNMs. Using the symmetrized operators, which satisfy canonical commutation relations, we derive the associated QNM-JC model for solving problems in multiplasmon or multiphoton quantum optics. To illustrate the practical application of the theory,

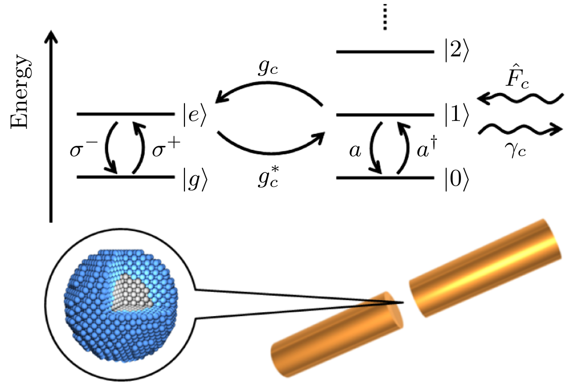

FIG. 1. Schematic of the QNM-JC model for a two-level emitter, e.g., in the form of a colloidal quantum dot coupled to a single QNM of a plasmonic dimer of gold nanorods. The electronic states $|e\rangle$ and $|g\rangle$ are coupled to the plasmonic Fock states $|n\rangle$ via the coupling constant g_c. Quantum fluctuations associated with the electromagnetic dissipation enter naturally through the operator $\hat{F}_c$.

in the case of a single QNM, we first use an example of a gold nanorod resonator, as illustrated in Fig. 1. Subsequently, we present the two-QNM-JC model and exemplify using a hybrid metal-dielectric cavity to highlight how modes quantum-mechanically interfere, leading to a dramatic breakdown of phenomenological dissipative JC models.

Theory.—We consider the interaction between a two-level emitter and the total electric field in the presence of a dispersive and absorptive, spatially inhomogeneous medium. The total Hamiltonian $H_{\rm total} = H_a + H_B + H_I$, based on the seminal approaches from Refs. [42–46], is

$$H_{\rm total} = \hbar\omega_a\sigma^+\sigma^- + \hbar\int d\mathbf{r}\int_0^\infty d\omega\omega\mathbf{b}^\dagger(\mathbf{r},\omega)\cdot\mathbf{b}(\mathbf{r},\omega) - \left(\sigma^+\int_0^\infty d\omega\mathbf{d}_a\cdot\hat{\mathbf{E}}(\mathbf{r}_a,\omega) + \text{H.a.}\right), \tag{1}$$

where $\hbar$ is the reduced Planck constant, ω_a and $\mathbf{d}_a$ are the resonance frequency and dipole moment of the emitter, respectively, $\sigma^\pm$ denote raising and lowering operators, and we use a dipole-field interaction in the rotating wave approximation; the annihilation and creation operators $\mathbf{b}(\mathbf{r},\omega)$ and $\mathbf{b}^\dagger(\mathbf{r},\omega)$ act on joint excitations of the surrounding lossy media and electromagnetic degrees of freedom and satisfy canonical commutation relations [44]. The electric field operator fulfills the equation $\nabla\times\nabla\times\hat{\mathbf{E}}(\mathbf{r},\omega) - k_0^2\epsilon(\mathbf{r},\omega)\hat{\mathbf{E}}(\mathbf{r},\omega) = i\omega\mu_0\hat{\mathbf{j}}_{\rm noise}(\mathbf{r},\omega)$, where $k_0 = \omega/c$, $c = 1/\sqrt{\mu_0\epsilon_0}$ is the speed of light, $\epsilon(\mathbf{r},\omega) = \epsilon_R(\mathbf{r},\omega) + i\epsilon_I(\mathbf{r},\omega)$ is a complex permittivity, describing passive media $[\epsilon_I(\mathbf{r},\omega) > 0]$ and fulfilling the Kramers-Kronig relations, and $\hat{\mathbf{j}}_{\rm noise}(\mathbf{r},\omega) = \omega\sqrt{(\hbar\epsilon_0/\pi)\epsilon_I(\mathbf{r},\omega)}\mathbf{b}(\mathbf{r},\omega)$, with ϵ_0 denoting the permittivity of free space [44,47]. A formal solution is $\hat{\mathbf{E}}(\mathbf{r},\omega) = i/(\omega\epsilon_0)\int d\mathbf{r}'\mathcal{G}(\mathbf{r},\mathbf{r}',\omega)\hat{\mathbf{j}}_{\rm noise}(\mathbf{r}',\omega)$, where $\mathcal{G}(\mathbf{r},\mathbf{r}',\omega)$ is the electric field Green's function, fulfilling $\nabla\times\nabla\times\mathcal{G}(\mathbf{r},\mathbf{r}',\omega) - k_0^2\epsilon(\mathbf{r},\omega)\mathcal{G}(\mathbf{r},\mathbf{r}',\omega) = k_0^2\delta(\mathbf{r}-\mathbf{r}')$ and a suitable radiation condition. At this point, the quantization scheme from Ref. [44] provides already an intuitive picture for treating the system, but the frequency and spatial indices of $\mathbf{b}^{(\dagger)}(\mathbf{r},\omega)$ prevent numerical evaluations of the density matrix for applications beyond single photons (weak excitation).

To derive creation and annihilation operators for photonic or plasmonic resonances, we begin with the vector-valued QNM eigenfunctions $\tilde{\mathbf{f}}_\mu(\mathbf{r})$, defined from

$$\nabla\times\nabla\times\tilde{\mathbf{f}}_\mu(\mathbf{r}) - \frac{\tilde{\omega}_\mu^2}{c^2}\epsilon(\mathbf{r},\tilde{\omega}_\mu)\tilde{\mathbf{f}}_\mu(\mathbf{r}) = 0, \tag{2}$$

with a suitable boundary condition, e.g., the Silver-Müller radiation condition [48]. The QNM eigenfrequencies $\tilde{\omega}_\mu = \omega_\mu - i\gamma_\mu$ are complex, with $\gamma_\mu > 0$ describing loss. For positions close to an electromagnetic resonator, the $\mathcal{G}(\mathbf{r},\mathbf{r}',\omega)$ can often be very accurately approximated by an expansion of only a few dominant QNMs of the form [33–36,49] $\mathcal{G}_{\rm QNM}(\mathbf{r},\mathbf{r}',\omega) = \sum_\mu A_\mu(\omega)\tilde{\mathbf{f}}_\mu(\mathbf{r})\tilde{\mathbf{f}}_\mu(\mathbf{r}')$, where the QNMs are normalized [33,34,36,48]; for calculations in this work, we use the form $A_\mu(\omega) = \omega/[2(\tilde{\omega}_\mu - \omega)]$. Since the QNMs $\tilde{\mathbf{f}}_\mu(\mathbf{r})$ diverge in the far field, we replace $\tilde{\mathbf{f}}_\mu(\mathbf{r})$ in $\mathcal{G}_{\rm QNM}(\mathbf{r},\mathbf{r}',\omega)$ for positions outside the resonator geometry with a regularization based on a Dyson equation approach [50], $\tilde{\mathbf{F}}_\mu(\mathbf{r},\omega) = \int_V d\mathbf{r}'\mathcal{G}_B(\mathbf{r},\mathbf{r}',\omega)\Delta\epsilon(\mathbf{r}',\omega)\tilde{\mathbf{f}}_\mu(\mathbf{r}')$, where $\mathcal{G}_B$ is the homogeneous background medium Green's function, with constant permittivity ϵ_B, $\Delta\epsilon(\mathbf{r}',\omega) = \epsilon(\mathbf{r}',\omega) - \epsilon_B$, and V is the resonator geometry volume. The electric field operator is $\hat{\mathbf{E}}(\mathbf{r}) = \int_0^\infty d\omega\hat{\mathbf{E}}(\mathbf{r},\omega) + \text{H.a.}$ and can be expanded in basis functions of the Green's function as in Refs. [51–54]. Inspired by this elegant approach [51], we use instead an expansion using few dominating (regularized) QNMs as in $\mathcal{G}_{\rm QNM}(\mathbf{r},\mathbf{r}',\omega)$, so that the source field expression of the electric field operator can be rewritten $\hat{\mathbf{E}}(\mathbf{r}) = i\sqrt{\hbar/(2\epsilon_0)}\sum_\mu\sqrt{\omega_\mu}\tilde{\mathbf{f}}_\mu(\mathbf{r})\tilde{\alpha}_\mu + \text{H.a.}$, for positions inside V, which allows us to define QNM operators $\tilde{\alpha}_\mu = \sqrt{2/(\pi\omega_\mu)}\int_0^\infty d\omega A_\mu(\omega)\int d\mathbf{r}\sqrt{\epsilon_I(\mathbf{r},\omega)}\tilde{\mathbf{f}}_\mu(\mathbf{r})\cdot\mathbf{b}(\mathbf{r},\omega)$; for positions outside V in the spatial integral, $\tilde{\mathbf{f}}_\mu(\mathbf{r})$ is replaced by the regularized QNM $\tilde{\mathbf{F}}_\mu(\mathbf{r},\omega)$. The QNM operators are formed by the integral over the oscillator operators $\mathbf{b}(\mathbf{r},\omega)$, that collectively form the QNM resonance. For most practical examples, the use of few (one or two) QNMs has been shown to provide very accurate results in the weak-coupling regime [11,55]; this is typically the case in cavity-QED systems using resonators. Using the canonical commutation relations for $\mathbf{b}(\mathbf{r},\omega)$, the equal-time commutation relations for the QNM raising and lowering operators become $[\tilde{\alpha}_\mu, \tilde{\alpha}_\eta] = [\tilde{\alpha}_\mu^\dagger, \tilde{\alpha}_\eta^\dagger] = 0$ and $[\tilde{\alpha}_\mu, \tilde{\alpha}_\eta^\dagger] \equiv S_{\mu\eta}$, respectively, in which

$$S_{\mu\eta} = \int_0^\infty d\omega \frac{2A_\mu(\omega)A_\eta^*(\omega)}{\pi\sqrt{\omega_\mu\omega_\eta}}[S_{\mu\eta}^{\mathrm{nrad}}(\omega) + S_{\mu\eta}^{\mathrm{rad}}(\omega)], \quad (3)$$

where $S_{\mu\eta}^{\mathrm{nrad}}(\omega) = \int_V d\mathbf{r}\epsilon_I(\mathbf{r},\omega)\tilde{\mathbf{f}}_\mu(\mathbf{r})\cdot\tilde{\mathbf{f}}_\eta^*(\mathbf{r})$ reflects absorption due to the resonator material and $S_{\mu\eta}^{\mathrm{rad}}(\omega) = c^2/(2i\omega^2)\int_{S_V} dA_{\mathbf{s}}\{[\hat{\mathbf{n}}_{\mathbf{s}} \times (\nabla_{\mathbf{s}} \times \tilde{\mathbf{F}}_\mu(\mathbf{s},\omega))]\cdot\tilde{\mathbf{F}}_\eta^*(\mathbf{s},\omega) - [\hat{\mathbf{n}}_{\mathbf{s}} \times (\nabla_{\mathbf{s}} \times \tilde{\mathbf{F}}_\eta^*(\mathbf{s},\omega))]\cdot\tilde{\mathbf{F}}_\mu(\mathbf{s},\omega)\}$ describes radiation leaving the system through the surface S_V with the normal vector $\hat{\mathbf{n}}_{\mathbf{s}}$ pointing into V [56]. The matrix $(\mathbf{S})_{\mu\eta}$ is a Hermitian semipositive definite overlap matrix between different QNMs μ and η and is not a Kronecker delta as would be the case for closed, dielectric cavities. It is strictly positive definite if the modes are linearly independent, which we assume here. Since noncanonical commutation relations prevent the construction of Fock states, we introduce new operators via a symmetrizing *orthonormalization* transformation [62]:

$$a_\mu = \sum_\nu (\mathbf{S}^{-1/2})_{\mu\nu}\tilde{\alpha}_\nu, \qquad a_\mu^\dagger = \sum_\nu (\mathbf{S}^{-1/2})_{\nu\mu}\tilde{\alpha}_\nu^\dagger, \quad (4)$$

yielding $[a_\mu, a_\eta^\dagger] = \delta_{\mu\eta}$. The operators a_μ and $a_\mu^\dagger$ are therefore proper annihilation and creation operators, respectively, for obtaining Fock states $|\mathbf{n}\rangle \equiv |n_1, n_2, \ldots\rangle$ from the vacuum state $|0\rangle$. The electric field expressed by a_μ, $a_\mu^\dagger$ is then

$$\hat{\mathbf{E}}(\mathbf{r}) = i\sqrt{\frac{\hbar}{2\epsilon_0}}\sum_\mu \sqrt{\omega_\mu}\,\tilde{\mathbf{f}}_\mu^s(\mathbf{r})a_\mu + \mathrm{H.a.}, \quad (5)$$

in the desired basis and with *symmetrized* QNM functions $\tilde{\mathbf{f}}_\mu^s(\mathbf{r}) = \sum_\nu (\mathbf{S}^{1/2})_{\nu\mu}\sqrt{\omega_\nu/\omega_\mu}\tilde{\mathbf{f}}_\nu(\mathbf{r})$. For a single QNM, $\mu = \nu = c$, we get $\hat{\mathbf{E}}(\mathbf{r}) = i\sqrt{\hbar\omega_c/(2\epsilon_0)}\tilde{\mathbf{f}}_c^s(\mathbf{r})a + \mathrm{H.a.}$, with $\tilde{\mathbf{f}}_c^s(\mathbf{r}) = \sqrt{S_c}\tilde{\mathbf{f}}_c(\mathbf{r})$, $S_c = S_{cc}$, and $a \equiv a_c$. The dynamics of the operators a_μ are governed by the Heisenberg equations of motion [56],

$$\frac{d}{dt}a_\mu = -\frac{i}{\hbar}[a_\mu, H_{\mathrm{sys}}] - \sum_\eta \chi_{\mu\eta}^{(-)}a_\eta + \hat{F}_\mu, \quad (6)$$

where $H_{\mathrm{sys}} = H_{\mathrm{em}} + H_a + H_I$ is the effective system Hamiltonian in the symmetrized basis and $H_{\mathrm{em}} = \hbar\sum_{\mu\eta}\chi_{\mu\eta}^{(+)}a_\mu^\dagger a_\eta$ is the electromagnetic part with nondiagonal terms $\eta \neq \mu$, that includes coupling between different symmetrized QNMs μ and η [40]; $H_I = -i\hbar\sum_\mu g_\mu\sigma^+ a_\mu + \mathrm{H.a.}$ is derived from H_I in Eq. (1) by inserting Eq. (5); this yields an emitter-QNM dipole-field interaction in the symmetrized basis $g_\mu = \sum_\eta (\mathbf{S}^{1/2})_{\eta\mu}\tilde{g}_\eta$, with $\tilde{g}_\mu = \sqrt{\omega_\mu/(2\epsilon_0\hbar)}\mathbf{d}_a\cdot\tilde{\mathbf{f}}_\mu(\mathbf{r}_a)$. In contrast to a phenomenological approach, complex QNM eigenfrequencies and complex eigenfunctions are used to obtain g_μ. The coupling between different QNMs is induced by the symmetrizing transformation, Eq. (4), and the coupling constant $\chi_{\mu\eta}^{(+)}$ is given via $\chi_{\mu\eta}^{(+)} = \frac{1}{2}(\chi_{\mu\eta} + \chi_{\eta\mu}^*)$, where $\chi_{\mu\eta} = \sum_\nu (\mathbf{S}^{-1/2})_{\mu\nu}\tilde{\omega}_\nu(\mathbf{S}^{1/2})_{\nu\eta}$. The two additional terms in Eq. (6) account for dissipation of energy through $\chi_{\mu\eta}^{(-)} = (i/2)(\chi_{\mu\eta} - \chi_{\eta\mu}^*)$ and coupling to a noise term $\hat{F}_\mu = \sum_\nu (\mathbf{S}^{-1/2})_{\mu\nu}\int_0^\infty d\omega C_\nu(\omega)\int d\mathbf{r}\sqrt{\omega\epsilon_I(\mathbf{r},\omega)}\tilde{\mathbf{f}}_\nu(\mathbf{r})\cdot\mathbf{b}(\mathbf{r},\omega)$, with $C_\nu(\omega) = i\sqrt{\omega/(2\pi\omega_\nu)}$. The presence of the noise term preserves the commutation relation $[a_\mu, a_\eta^\dagger] = \delta_{\mu\eta}$ temporally by exactly counteracting the damping due to dissipation. Equation (6) has the form of a Langevin equation with noise operators $\hat{F}_\mu$ [26,27]. In the following, we use it to set up the QNM-JC model and derive the associated quantum master equations for the illustrative cases of one and two QNMs; we refer to Ref. [56] for the general case.

(I) *One-QNM-JC model.*—We consider the material system from Fig. 1 consisting of an emitter at the position $\mathbf{r}_a$, directly in the center of a plasmonic dimer of nanorods supporting a single QNM with index c, in which case H_{em} takes the simplified form $H_{\mathrm{em}} = \hbar\omega_c a^\dagger a$, and the interaction Hamiltonian becomes $H_I = -i\hbar(g_c\sigma^+ a - \mathrm{H.a.})$, with $g_c = \sqrt{S_c}\tilde{g}_c$ and $\chi_{\mu\eta}^{(-)} = -\mathrm{Im}(\tilde{\omega}_c) = \gamma_c$. All system operators evolve according to a quantum Langevin equation similar to Eq. (6). Employing stochastic Ito-Stratonovich calculus [27], we can bring these equations into a Lindblad master equation form. To this end, we treat the quantum noise operator $\hat{F}_c$ as an input field, which represents white noise of a reservoir with temperature $T = 0$ K [56]. After some algebra, we find the one-QNM master equation

$$\partial_t\rho = -\frac{i}{\hbar}[H_{\mathrm{sys}}, \rho] + \gamma_c(2a\rho a^\dagger - a^\dagger a\rho - \rho a^\dagger a). \quad (7)$$

For a more detailed interpretation of the normalization S_c, it is instructive to consider the cavity-modified spontaneous emission rate in the bad cavity limit. We adiabatically eliminate the electromagnetic degrees of freedom from Eq. (7), to obtain a master equation for the emitter density matrix alone, consisting of the dissipator term $\mathcal{L}[\sigma^-]\rho = \Gamma(2\sigma^-\rho\sigma^+ - \sigma^+\sigma^-\rho - \rho\sigma^+\sigma^-)$, with spontaneous emission rate $\Gamma = \gamma_c|g_c|^2/(\Delta_{ca}^2 + \gamma_c^2)$ and detuning $\Delta_{ca} = \omega_c - \omega_a$ [56]. In cases where the single QNM expansion is a good approximation to the Green's function throughout the entire resonator volume, we expect to recover the semiclassical result $\Gamma = (2/\hbar\epsilon_0)\mathbf{d}_a\cdot\mathrm{Im}[\mathcal{G}(\mathbf{r}_a, \mathbf{r}_a, \omega)]\cdot\mathbf{d}_a$, i.e., the local density of states modeled via the photonic Green's function in a QNM approximation [37,55]. In the present case of the plasmonic

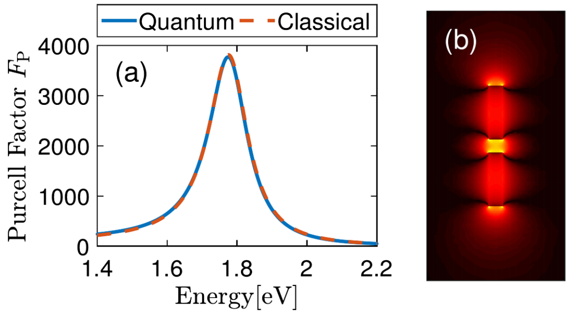

FIG. 2. (a) Purcell factor F_P as a function of the energy for the plasmonic dimer in Fig. 1. Solid and dashed curves show the results of the QNM-JC model and a semiclassical approach, using a single QNM Green's function approximation, respectively. (b) Normalized spatial profile of the QNM of interest with $\tilde{\omega}_c(\mathrm{eV}) = 1.7786 - 0.0677i$, corresponding to $Q = \omega_c/(2\gamma_c) \approx 13$.

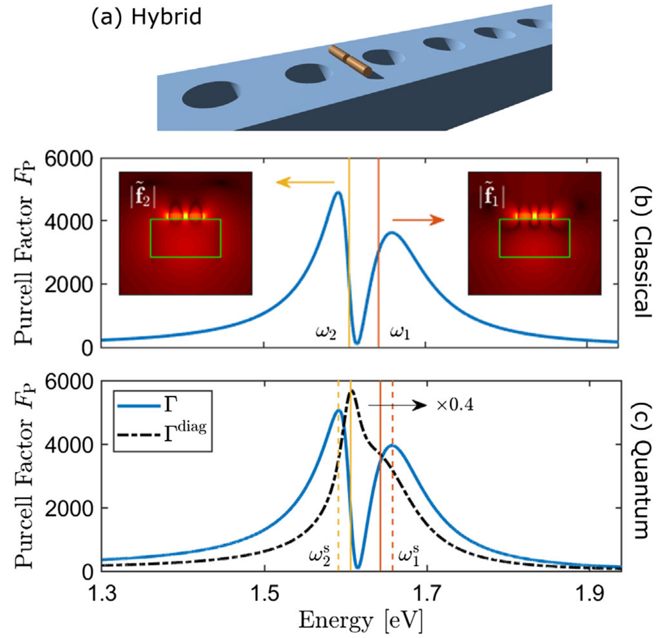

FIG. 3. (a) Gold dimer on top of a photonic crystal cavity, supporting two overlapping QNMs with frequencies $\tilde{\omega}_2(\mathrm{eV}) = 1.6063 - 0.0145i$ and $\tilde{\omega}_1(\mathrm{eV}) = 1.6428–0.0548i$ (mode 1 originates from the dimer). (b) QNM profiles and semiclassical Purcell factor as a function of the energy. (c) QNM-JC Purcell factor with diagonal contributions Γ^{diag} (black dashed line, scaled) and the full emission rate $\Gamma = \Gamma^{\mathrm{diag}} + \Gamma^{\mathrm{ndiag}}$ (solid blue line). Vertical solid and dashed lines show the shifted and original eigenfrequencies, respectively.

dimer, we find a very good agreement, as seen in Fig. 2, showing the Purcell factor $F_P = \Gamma/\Gamma^0$, where Γ^0 is the spontaneous emission rate in a homogeneous medium. Although the agreement in Fig. 2 is already striking (especially given the completely different nature of the calculations [56]), we remark that the restriction to a few dominant QNMs in the QNM-JC model, when applied to spontaneous emission, is generally different, and typically less accurate, than the use of the same approximation to the Green's function in a semiclassical approach. Whereas the latter relies only on the expansion at a single point, the QNM-JC model is based on integrals of the QNMs throughout the resonator material to obtain S_c. For the plasmonic dimer, we find $S_c^{\mathrm{nrad}} = 0.58$ and $S_c^{\mathrm{rad}} = 0.40$. In addition, S_c^{nrad} and S_c^{rad} yield the nonradiative and radiative beta factor, respectively, via $\beta^{\mathrm{nrad}} = S_c^{\mathrm{nrad}}/S_c$ and $\beta^{\mathrm{rad}} = S_c^{\mathrm{rad}}/S_c$. See [56] for details of the QNM calculation, $\tilde{\mathbf{f}}_\mu(\mathbf{r}_a)$, and material parameters.

(II) *Two-QNM-JC model.*—We next discuss a case where cross terms $\chi_{\mu\eta}$ of two QNMs μ, $\eta = 1, 2$ cause interference effects, clearly not available in phenomenological quantization approaches. Starting again from the quantum Langevin equation in Eq. (6), we derive a Lindblad master equation analogue to the one-mode case, using the additional assumptions [27] that the two input fields associated with $\hat{F}_\mu$ ($\mu = 1, 2$) are independent from each other and that the real parts of the eigenfrequencies ω_1 and ω_2 are not degenerate [63]. Again, following the approach of Ref. [27], we now obtain the two-QNM master equation

$$\partial_t \rho = -\frac{i}{\hbar}[H_{\mathrm{sys}}, \rho] + \mathcal{L}[\mathbf{a}]\rho, \tag{8}$$

where ω_μ are no longer eigenvalues of the electromagnetic part of the Hamiltonian, since an intermode coupling appears. Instead, a pair of shifted eigenfrequencies ω_μ^s is formed [see Fig. 3(c)]. We stress that the Lindblad dissipator $\mathcal{L}[\mathbf{a}]\rho = \sum_{\mu,\eta}\chi_{\mu\eta}^{(-)}(2a_\eta\rho a_\mu^\dagger - a_\mu^\dagger a_\eta\rho - \rho a_\mu^\dagger a_\eta)$ contains also processes with interacting QNMs $\mu \neq \eta$.

Although the above off-diagonal coupling may seem unusual, it is known that a significant mode interference, such as a "Fano-type" resonance, can occur because of the different phase terms of overlapping QNMs [11,64]. In the QNM-JC model, this interference is captured by the off-diagonal terms, as illustrated in Fig. 3, where we study the electromagnetic response of the metal dimer from (I) on top of a high-Q photonic crystal cavity [see Fig. 3(a)]. Figure 3(b) shows the two QNMs of interest and the semiclassical result of the Purcell factor as calculated using a two-QNM approximation [11,56]; Fig. 3(c) shows the corresponding results of the QNM-JC model in this pronounced QNM coupling regime [65]. The system parameters indicate the bad cavity limit, where the QNM-JC master equation consists of a Lindblad dissipator for spontaneous emission of the form $\mathcal{L}[\sigma^-]\rho = \Gamma(2\sigma^-\rho\sigma^+ - \sigma^+\sigma^-\rho - \rho\sigma^+\sigma^-)$, in which $\Gamma = \Gamma^{\mathrm{diag}} + \Gamma^{\mathrm{ndiag}}$ with a diagonal contribution $\Gamma^{\mathrm{diag}} = \sum_\mu S_{\mu\mu}|\tilde{g}_\mu|^2\gamma_\mu/(\Delta_{\mu a}^2 + \gamma_\mu^2)$ and a nondiagonal contribution $\Gamma^{\mathrm{ndiag}} = \sum_{\mu,\eta\neq\mu}\tilde{g}_\mu S_{\mu\eta}\tilde{g}_\eta^* K_{\mu\eta}$, which is here expressed in terms of the coupling matrix $K_{\mu\eta} = [i(\omega_\mu - \omega_\eta) + \gamma_\mu + \gamma_\eta]/[2(\Delta_{\mu a} - i\gamma_\mu)(\Delta_{\eta a} + i\gamma_\eta)]$ [56]. Comparing the results in Fig. 3, one sees that the two-QNM-JC model recovers the result of the semiclassical calculation, including the pronounced Fano-type interference effect. In a phenomenological dissipative two-mode JC model, the Lindblad dissipator is simply $\mathcal{L}[\sigma^-]\rho = \sum_i \Gamma^i(2\sigma^-\rho\sigma^+ - \sigma^+\sigma^-\rho - \rho\sigma^+\sigma^-)$ and $\Gamma^i = |g_i|^2\gamma_i/(\Delta_{ia}^2 + \gamma_i^2)$ is the diagonal cavity-modified rate for each of the two modes. Clearly, such a model cannot produce the aforementioned interference effect, as illustrated by the black dashed curve in Fig. 3(c).

By construction, the symmetrized raising and lowering operators fulfill all requirements for use in the construction of a Fock space. Combined with the fact that the one- and two-QNM-JC master equations recover the semiclassical results in the single excitation subspace, where a direct comparison to reference calculations is possible, we consider the approach rigorous enough that one can apply the QNM-JC model also to problems in multiplasmon or multiphoton Fock spaces. Although we have connected to the bad cavity limit, the QNM master equation now allows one to explore multiphoton dynamics, which will be the subject of future work.

In conclusion, by use of a symmetrization procedure, we have introduced creation and annihilation operators, allowing the construction of QNM Fock states and the derivation of a physically meaningful and intuitive QNM-JC model for use in dissipative cavity-QED valid for arbitrary dissipative structures. We have shown example applications of the theory for a plasmonic dimer to verify that the QNM-JC model recovers the semiclassical result in Purcell factor calculations. Finally, we discussed the highly nontrivial case of a two-QNM-JC model, where interference effects cannot be neglected. In this case, the model recovers the semiclassical result only because of off-diagonal coupling terms, which are not present in phenomenological dissipative JC models. Contrary to phenomenological approaches, all parameters entering the model are rigorously defined and can be calculated by use of the relevant QNMs. The model thus provides a solid foundation for the use of the JC model and a rigorous extension of the model to several modes and possibly dissipative materials, in which the quantized "cavity modes" of optical cavities or plasmonic resonators appear as linear combinations of the associated QNMs.

We acknowledge funding from the Deutsche Forschungsgemeinschaft through SFB 951 HIOS (B12/B10, Project No. 182087777) and project BR1528/8-2 (Project No. 177864488) for support and from the Natural Sciences and Engineering Research Council of Canada. S. F. acknowledges support from the School of Nanophotonics (SFB 787, Project No. 43659573). This project has also received funding from the European Unions Horizon 2020 research and innovation program under Grant Agreement No. 734690 (SONAR).

*sebastian.franke@tu-berlin.de

[1] S. Reitzenstein, C. Hofmann, A. Gorbunov, M. Strauß, S. H. Kwon, C. Schneider, A. Löffler, S. Höfling, M. Kamp, and A. Forchel, Appl. Phys. Lett. **90**, 251109 (2007).
[2] D. Bajoni, P. Senellart, E. Wertz, I. Sagnes, A. Miard, A. Lemaître, and J. Bloch, Phys. Rev. Lett. **100**, 047401 (2008).
[3] J. P. Reithmaier, G. Sek, A. Löffler, C. Hofmann, S. Kuhn, S. Reitzenstein, L. V. Keldysh, V. D. Kulakovskii, T. L. Reinecke, and A. Forchel, Nature (London) **432**, 197 (2004).
[4] T. Yoshie, A. Scherer, J. Hendrickson, G. Khitrova, H. M. Gibbs, G. Rupper, C. Ell, O. B. Shchekin, and D. G. Deppe, Nature (London) **432**, 200 (2004).
[5] V. S. C. Manga Rao and S. Hughes, Phys. Rev. Lett. **99**, 193901 (2007).
[6] O. Kulakovich, N. Strekal, A. Yaroshevich, S. Maskevich, S. Gaponenko, I. Nabiev, U. Woggon, and M. Artemyev, Nano Lett. **2**, 1449 (2002).
[7] G. M. Akselrod, M. C. Weidman, Y. Li, C. Argyropoulos, W. A. Tisdale, and M. H. Mikkelsen, ACS Photonics **3**, 1741 (2016).
[8] C. David, M. Richter, A. Knorr, I. M. Weidinger, and P. Hildebrandt, J. Chem. Phys. **132**, 024712 (2010).
[9] C. Strelow, T. S. Theuerholz, C. Schmidtke, M. Richter, J.-P. Merkl, H. Kloust, Z. Ye, H. Weller, T. F. Heinz, A. Knorr, and H. Lange, Nano Lett. **16**, 4811 (2016).
[10] M. Barth, S. Schietinger, S. Fischer, J. Becker, N. Nüsse, T. Aichele, B. Löchel, C. Sönnichsen, and O. Benson, Nano Lett. **10**, 891 (2010).
[11] M. Kamandar Dezfouli, R. Gordon, and S. Hughes, Phys. Rev. A **95**, 013846 (2017).
[12] M. O. Scully, K. Wodkiewicz, M. S. Zubairy, J. Bergou, N. Lu, and J. M. ter Vehn, Phys. Rev. Lett. **60**, 1832 (1988).
[13] D. J. Bergman and M. I. Stockman, Phys. Rev. Lett. **90**, 027402 (2003).
[14] G. Kewes, K. Herrmann, R. Rodríguez-Oliveros, A. Kuhlicke, O. Benson, and K. Busch, Phys. Rev. Lett. **118**, 237402 (2017).
[15] A. Imamoğlu, D. D. Awschalom, G. Burkard, D. P. DiVincenzo, D. Loss, M. Sherwin, and A. Small, Phys. Rev. Lett. **83**, 4204 (1999).
[16] L. Knöll, W. Vogel, and D. G. Welsch, Phys. Rev. A **36**, 3803 (1987).
[17] J. Gea-Banacloche, N. Lu, L. M. Pedrotti, S. Prasad, M. O. Scully, and K. Wódkiewicz, Phys. Rev. A **41**, 369 (1990).
[18] R. J. Glauber and M. Lewenstein, Phys. Rev. A **43**, 467 (1991).
[19] B. J. Dalton, E. S. Guerra, and P. L. Knight, Phys. Rev. A **54**, 2292 (1996).
[20] G.-I. Kweon and N. Lawandy, Opt. Commun. **118**, 388 (1995).
[21] E. T. Jaynes and F. W. Cummings, Proc. IEEE **51**, 89 (1963).
[22] G. S. Agarwal, *Quantum Optics* (Cambridge University Press, Cambridge, England, 2013).
[23] A. Zrenner, E. Beham, S. Stufler, F. Findeis, M. Bichler, and G. Abstreiter, Nature (London) **418**, 612 (2002).
[24] M. Gegg and M. Richter, New J. Phys. **18**, 043037 (2016).
[25] P. Lodahl, S. Mahmoodian, and S. Stobbe, Rev. Mod. Phys. **87**, 347 (2015).
[26] C. W. Gardiner and P. Zoller, *Quantum Noise* (Springer, New York, 2004).
[27] C. W. Gardiner and M. J. Collett, Phys. Rev. A **31**, 3761 (1985).

[28] E. Waks and D. Sridharan, Phys. Rev. A **82**, 043845 (2010).
[29] M. M. Dignam and M. K. Dezfouli, Phys. Rev. A **85**, 013809 (2012).
[30] S. Hughes, M. Richter, and A. Knorr, Opt. Lett. **43**, 1834 (2018).
[31] M. Hensen, T. Heilpern, S. K. Gray, and W. Pfeiffer, ACS Photonics **5**, 240 (2018).
[32] R.-C. Ge and S. Hughes, Phys. Rev. B **92**, 205420 (2015).
[33] K. M. Lee, P. T. Leung, and K. M. Pang, J. Opt. Soc. Am. B **16**, 1409 (1999).
[34] E. A. Muljarov, W. Langbein, and R. Zimmermann, Europhys. Lett. **92**, 50010 (2010).
[35] P. T. Kristensen, C. Van Vlack, and S. Hughes, Opt. Lett. **37**, 1649 (2012).
[36] C. Sauvan, J. P. Hugonin, I. S. Maksymov, and P. Lalanne, Phys. Rev. Lett. **110**, 237401 (2013).
[37] P. T. Kristensen and S. Hughes, ACS Photonics **1**, 2 (2014).
[38] P. Lalanne, W. Yan, K. Vynck, C. Sauvan, and J.-P. Hugonin, Laser Photonics Rev. **12**, 1700113 (2018).
[39] A. I. Fernández-Domínguez, and S. I. Bozhevolnyi, and N. A. Mortensen, ACS Photonics **5**, 3447 (2018).
[40] K. C. Ho, P. T. Leung, A. Maassen van den Brink, and K. Young, Phys. Rev. E **58**, 2965 (1998).
[41] S. Severini, A. Settimi, C. Sibilia, M. Bertolotti, A. Napoli, and A. Messina, Phys. Rev. E **70**, 056614 (2004).
[42] B. Huttner and S. M. Barnett, Phys. Rev. A **46**, 4306 (1992).
[43] A. Tip, Phys. Rev. A **56**, 5022 (1997).
[44] H. T. Dung, L. Knöll, and D.-G. Welsch, Phys. Rev. A **57**, 3931 (1998).
[45] L. G. Suttorp and A. J. van Wonderen, Europhys. Lett. **67**, 766 (2004).
[46] W. Vogel and D.-G. Welsch, *Quantum Optics* (Wiley, New York, 2006).
[47] T. Gruner and D.-G. Welsch, Phys. Rev. A **53**, 1818 (1996).
[48] P. T. Kristensen, R.-C. Ge, and S. Hughes, Phys. Rev. A **92**, 053810 (2015).
[49] M. B. Doost, W. Langbein, and E. A. Muljarov, Phys. Rev. A **87**, 043827 (2013).
[50] R.-C. Ge, P. T. Kristensen, J. F. Young, and S. Hughes, New J. Phys. **16**, 113048 (2014).
[51] T. Hümmer, F. J. García-Vidal, L. Martín-Moreno, and D. Zueco, Phys. Rev. B **87**, 115419 (2013).
[52] H. Varguet, B. Rousseaux, D. Dzsotjan, H. R. Jauslin, S. Guérin, and G. C. des Francs, Opt. Lett. **41**, 4480 (2016).
[53] D. Dzsotjan, B. Rousseaux, H. R. Jauslin, G. C. des Francs, C. Couteau, and S. Guérin, Phys. Rev. A **94**, 023818 (2016).
[54] B. Rousseaux, D. Dzsotjan, G. Colas des Francs, H. R. Jauslin, C. Couteau, and S. Guérin, Phys. Rev. B **93**, 045422 (2016).
[55] R.-C. Ge and S. Hughes, Opt. Lett. **39**, 4235 (2014).
[56] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.122.213901 for detailed calculations, including the derivation of the commutation relations, the Heisenberg equation of motion of the quasinormal mode operators, the derivation of the Lindblad master equation, and the bad cavity limit for the multi-QNM case as well as a discussion about details of the numerical calculations, which includes Refs. [57–61].
[57] C. W. Gardiner, *Stochastic Methods: A Handbook for the Natural and Social Sciences* (Springer, New York, 2009).
[58] R. A. Horn and C. R. Johnson, *Matrix Analysis* (Cambridge University Press, Cambridge, England, 2012).
[59] J. I. Cirac, Phys. Rev. A **46**, 4354 (1992).
[60] COMSOL Multiphysics: http://www.comsol.com.
[61] Q. Bai, M. Perrin, C. Sauvan, J.-P. Hugonin, and P. Lalanne, Opt. Express **21**, 27371 (2013).
[62] P. Löwdin, J. Chem. Phys. **18**, 365 (1950).
[63] M. Lax, Phys. Rev. **145**, 110 (1966).
[64] J. R. de Lasson, P. T. Kristensen, J. Mørk, and N. Gregersen, Opt. Lett. **40**, 5790 (2015).
[65] The calculated S factors are $S_{11} = 0.7485 + 0.7356$, $S_{22} = 0.8006 + 0.4409$, $S_{12} = (-0.0043 - 0.5574i) + (0.0579 - 0.4137i)$, and $S_{21} = S_{12}^*$, where the first and second parts denote $S_{\mu\eta}^{\mathrm{nrad}}$ and $S_{\mu\eta}^{\mathrm{rad}}$, respectively.