Letter

Optimal quantum spectroscopy using single-photon pulses

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We provide the ultimate precision attainable in spectroscopy of a quantum emitter using single-photon pulses. We find that the maximum precision of linewidth estimation is independent of the details of the emitter's bare Hamiltonian, though this is not true for the detunings. We also identify optimal pulse shapes attaining these precisions.

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Introduction. Spectroscopy seeks to estimate parameters that model a matter system by using light as a probe. Recent studies used quantum states of light such as single photons [1–3], squeezed [4,5], or entangled states of light [6,7], to achieve enhanced precision in spectroscopy beyond what is possible with classical light [8,9]. However, the maximum possible precision attainable in spectroscopy using quantum states of light remains unidentified.

In this Letter, we provide the ultimate precisions in estimating the transition energies (detunings) and linewidths of a quantum emitter starting in its ground state and probed by single-photon pulses [1–3,10–12], as illustrated in Fig. 1. For conceptual clarity, we assume that all photons emitted from the matter system are detected without loss. This could be physically realized using quantum emitters embedded in waveguides [13–15].

Our results are summarized in Table I. They are obtained using the tools of quantum estimation theory [16], which quantifies the ultimate precision with which a parameter can be estimated using quantum probes. An upper bound on this precision, the variance of an unbiased estimator $\hat{\theta}$ of a parameter θ is set by the Cramér-Rao bound [16]

$$\mathrm{Var}(\hat{\theta}) \geqslant \frac{1}{\mathcal{Q}(\rho_\theta, \theta)}, \tag{1}$$

where $\mathcal{Q}(\rho_\theta, \theta)$ is the quantum Fisher information (QFI). It depends on the quantum state of the probe ρ_θ and its derivative with respect to θ.

In spectroscopy, the QFI serves as a measure of the information about θ contained in the quantum state of the light after it has interacted with the matter system [17–20]. We thus maximize its QFI over all possible input states of light.

Model. We model the quantum matter system (M) as a point emitter, localized at $z = 0$ and possessing a ground state $|g\rangle$ defined to have zero energy. Since we are interested in the single-photon driving of the emitter which is assumed to be in the ground state at the start of the experiment, it will at most accommodate a single excitation through the course of the interaction. Thus, the total state space of the emitter can be effectively described by an $(N+1)$-dimensional Hilbert space, where N is the dimension of the singly excited subspace (SES) of the emitter (see Fig. 1). For example, in a coupled dimer system built from two two-level systems, $|g\rangle$ represents the state with both TLSs in the ground state, and the SES is spanned by $N = 2$ states.

The light pulse is a propagating quantized radiation field characterized by a set of frequency-dependent bosonic operators $a(\omega)$ obeying the commutation relation $[a(\omega), a^\dagger(\omega')] = \delta(\omega - \omega')$. The free-field Hamiltonian is given by $H_P = \int d\omega\, \omega a^\dagger(\omega) a(\omega)$ (assuming natural units characterised by $\hbar = 1$). We also assume that the incoming quantum optical pulse is sufficently narrow in spectral bandwidth $B \ll \omega_c$ around the carrier frequency ω_c, a reasonable assumption for optical frequencies.

The electromagnetic field then couples to the emitter through the dipole interaction Hamiltonian $H_{MP} = -\vec{\mathbf{d}} \cdot \vec{\mathbf{E}}$, where $\vec{\mathbf{d}}$ is the emitter dipole moment, and

$$\vec{\mathbf{E}} = i\hat{\mathbf{e}} \int_0^\infty d\omega \sqrt{\frac{\omega}{4\pi\epsilon_0 cA}}\, a(\omega) e^{-i\omega t} + \mathrm{H.c.} \tag{2}$$

is the linearly polarized electric field operator in the paraxial regime [21], with A being the transverse quantization area and $\hat{\mathbf{e}}$ the unit polarization vector. Assuming also that the incoming light pulse couples only very weakly to the matter system M (so timescales for light-matter dynamics are well separated from matter-only dynamics), we can employ the rotating wave and Markovian white-noise approximations [22–24] to

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TABLE I. Upper bounds to the QFI $[\mathcal{Q}_{\max}(\cdot)]$ and the optimal incident pulse states ($|1_{\text{opt}}\rangle$) for estimating the linewidth Γ and detunings Δ_j from the scattered pulse at long time ($t \gg 1/\Gamma$) for a $N+1$ level emitter (second column) and a two-level emitter (third column).

	$N+1$ level emitter	TLS
$\mathcal{Q}_{\max}(\Gamma)$	$\left(\frac{2}{\Gamma}\right)^2$	$\left(\frac{2}{\Gamma}\right)^2$
$\|1_{\text{opt}}\rangle$	$\frac{1}{\sqrt{2}}[\|1_{\Omega^{\Gamma}_{\max}}\rangle + e^{i\varphi}\|1_{\Omega^{\Gamma}_{\min}}\rangle]$	$\frac{1}{\sqrt{2}}[\|1_{\Delta+\frac{\Gamma}{2}}\rangle + e^{i\varphi}\|1_{\Delta-\frac{\Gamma}{2}}\rangle]$
$\mathcal{Q}_{\max}(\Delta_j)$	$(\mu^{\Delta_j}_{\max})^2 = (\max \mathcal{X}^{\Delta_j}_{\text{M}}(\omega))^2$	$\left(\frac{4}{\Gamma}\right)^2$
$\|1_{\text{opt}}\rangle$	$\frac{1}{\sqrt{2}}[\|1_{\Omega^{\Delta_j}_{\max}}\rangle + e^{i\varphi}\|1_{\infty}\rangle]$	$\frac{1}{\sqrt{2}}[\|1_{\Delta}\rangle + e^{i\varphi}\|1_{\infty}\rangle]$

express the total interaction-picture light-matter Hamiltonian as (see *Appendix A: Microscopic derivation of the light-matter Hamiltonian* for a microscopic derivation)

$$H(t) = H_{\text{M}} - i\sqrt{\Gamma}(\Sigma^{\dagger}a(t) - \Sigma a^{\dagger}(t)), \tag{3}$$

where H_{M} is the time-independent Hamiltonian of the emitter M, and Γ is the Weisskopf-Wigner rate of spontaneous emission characterizing the strength of the light-matter coupling, inversely proportional to the emitter's radiative lifetime, which, in turn, determines the natural linewidth of the emitted light ($\Gamma/2\pi$) to be estimated later. The time-dependent field operators $a(t) = \int_{-\infty}^{\infty} d\omega\, a(\omega)e^{-i(\omega-\omega_c)t}/\sqrt{2\pi}$ are the Fourier transform of the field operators $a(\omega)$, dubbed the "quantum white-noise operators," which, in the Markovian limit, satisfy the commutation relation $[a(t), a^{\dagger}(t')] = \delta(t-t')$.

The transition operator that couples to the field is taken to be $\Sigma = |g\rangle\langle\gamma|$, where $|\gamma\rangle$ is a normalized vector in the SES that typically depends on the dipole moment operator of the emitter [2,25]. This form of the transition operator that connects only the ground state to the SES states and excludes all higher excited states is employed under the constraint of the traveling light pulse carrying a single-excitation to the ground state of the emitter at the starting time $t = t_0$ before the pulse interacts with M (see *Appendix A: Microscopic derivation of the light-matter Hamiltonian*).

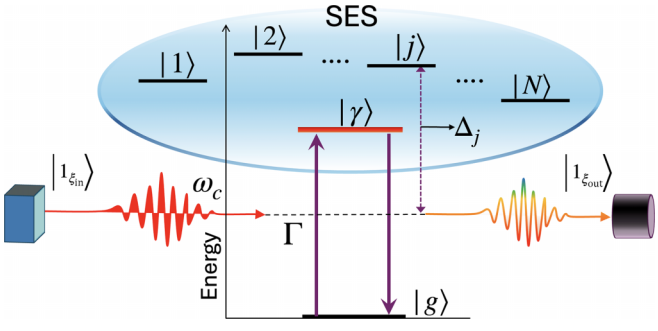

FIG. 1. The single-photon $|1_{\xi_{\text{in}}}\rangle$ excites the emitter from the ground state $|g\rangle$ to the single-excitation subspace (SES) of the emitter which is illustrated by the light blue shaded oval. Its elements $\{|j\rangle\}$ $(j = 1, 2, \ldots, N)$ (with energies ω_j relative to $|g\rangle$ which is defined to be zero) are illustrated with black solid lines in the SES. The detuning of the jth singly excited state is $\Delta_j = \omega_j - \omega_c$, where the thin horizontal dashed line along the path of the pulse indicates the carrier frequency ω_c. Γ denotes the coupling strength with the pulse and $|\gamma\rangle$ (thick red line) is a normalized superposition of the states $\{|j\rangle\}$ in the SES. The coupling encodes information of the emitter in the state of the scattered pulse $|1_{\xi_{\text{out}}}\rangle$.

The light-matter Hamiltonian $H(t)$ preserves the total number of excitations which is defined by the operator $\mathbf{N}_{\text{MP}} = \mathbf{I}_{\text{M}} + \int_0^{\infty} d\omega a^{\dagger}(\omega)a(\omega)$, where $\mathbf{I}_{\text{M}}$ is the identity operator in the SES. At $t = t_0$, the joint light-emitter state is $|\Psi(t_0)\rangle = |g\rangle|1_{\xi_{\text{in}}}\rangle$ where the incident single-photon state of the field is given by

$$|1_{\xi_{\text{in}}}\rangle = \int_{-\infty}^{\infty} d\tau\, \xi_{\text{in}}(\tau)a^{\dagger}(\tau)|0_{\text{P}}\rangle, \tag{4}$$

where $a^{\dagger}(\tau)|0_{\text{P}}\rangle$ is an element of the temporal mode basis of the single photon and $\xi_{\text{in}}(\tau)$ is the incident wavepacket of the pulse. This single-photon excites the emitter from $|g\rangle$ to the SES which subsequently spontaneously decays into $|g\rangle$ by emitting a photon, assuming M couples to no other environments. Thus, the final light-emitter state at times $t \gg 1/\Gamma$ takes the form $|\Psi(t \gg 1/\Gamma)\rangle = |g\rangle|1_{\xi_{\text{out}}}\rangle$, where $|1_{\xi_{\text{out}}}\rangle$ is the scattered single-photon state. Solving the Schrödinger equation corresponding to light-matter coupling in Eq. (3), we obtain [25–27]

$$\xi_{\text{out}}(\tau) = \xi_{\text{in}}(\tau) - \int_{-\infty}^{\infty} dt'\, \xi_{\text{in}}(t')f_{\text{M}}(\tau - t'), \tag{5}$$

$$\text{with} \quad f_{\text{M}}(t) = \Gamma\Theta(t)\langle\gamma|\exp\left(-iH_{\text{M}} - \Gamma\frac{|\gamma\rangle\langle\gamma|}{2}\right)t\,|\gamma\rangle,$$

where $\Theta(\cdot)$ is the Heaviside theta function. In the frequency domain

$$\tilde{\xi}_{\text{out}}(\omega) = \tilde{\xi}_{\text{in}}(\omega)\left(\frac{1 - i\chi_{\text{M}}(\omega)}{1 + i\chi_{\text{M}}(\omega)}\right), \tag{6}$$

where $\tilde{\xi}_{\text{in/out}}(\omega) = \int_{-\infty}^{\infty} d\tau \xi_{\text{in/out}}(\tau)e^{i\omega\tau}/\sqrt{2\pi}$ and

$$\chi_{\text{M}}(\omega) = \Gamma\langle\gamma|[\![(\omega\mathbf{I}_{\text{M}} - H_{\text{M}})]\!]^{-1}|\gamma\rangle/2, \tag{7}$$

where $[\![.]\!]^{-1}$ denotes the operator inverse.

Mathematically, $\chi_{\text{M}}(\omega)$ characterizes the matter-induced response that modulates the pulse's spectral phase since Eq. (6) can be rewritten as $\tilde{\xi}_{\text{out}} = \tilde{\xi}_{\text{in}}\exp[-2i\tan^{-1}(\chi_{\text{M}})]$.

Since the final state of the scattered pulse is a pure state $|1_{\xi_{\text{out}}}\rangle$, we can treat the total evolution of the pulse as a unitary encoding process with an effective unitary operator U_{B} such that $|1_{\xi_{\text{out}}}\rangle = U_{\text{B}}|1_{\xi_{\text{in}}}\rangle$. Using Eq. (6),

$$U_{\text{B}} = \int_{-\infty}^{\infty} d\omega\left(\frac{1 - i\chi_{\text{M}}(\omega)}{1 + i\chi_{\text{M}}(\omega)}\right)|1_{\omega}\rangle\langle 1_{\omega}|, \tag{8}$$

where $|1_{\omega}\rangle = a^{\dagger}(\omega)|0_{\text{P}}\rangle$. This operator is diagonal in the frequency basis, and its unitarity is ensured by the

diagonal elements which represent a frequency-dependent phase. Therefore, we can define a dimensionless effective Hamiltonian that generates the unitary $U_{\rm B} = \exp(-iH_{\rm B})$, expressed explicitly as

$$H_{\rm B} = \int_{-\infty}^{\infty} d\omega\, 2\tan^{-1}[\chi_{\rm M}(\omega)]\, |1_\omega\rangle\langle 1_\omega| . \tag{9}$$

This effective Hamiltonian formulation of the unitary parameter encoding onto the single-photon pulse due to the light-matter interaction forms the basis of the evaluation of quantum precision bounds, and is one of our central results.

Equations (6) and (8) show that even when $\chi_{\rm M}(\omega)$ is unbounded, the phase it imparts in the spectral wavepacket of the scattered photon remains bounded. Physically, singularities in $\chi_{\rm M}(\omega)$ then correspond to a π spectral phase imparted by the effective Hamiltonian, as shown by Eq. (9) [28].

Spectroscopy. For unitary parameter encoding generated by the Hamiltonian operator $H_{\rm B}$, the following tight upper bound to the QFI of estimating a parameter θ encoded in the eigenvalues of $H_{\rm B}$ can be obtained [29–31]:

$$\mathcal{Q}(|1_{\xi_{\rm out}}\rangle, \theta) \leqslant \left(\mu^{\theta}_{\rm max} - \mu^{\theta}_{\rm min}\right)^2 \equiv \mathcal{Q}_{\rm max}(\theta). \tag{10}$$

Here $\mu^{\theta}_{\rm min/max}$ represent the minimum or maximum eigenvalues of the operator $\partial_\theta H_{\rm B}$. These correspond to the global minima and maxima of the function

$$\mathcal{X}^{\theta}_{\rm M}(\omega) = \frac{2\partial_\theta \chi_{\rm M}(\omega)}{1+\chi_{\rm M}(\omega)^2}.$$

The upper bound in Eq. (10) is saturated when the incident pulse is an equally weighted superposition of the eigenstates of $\partial_\theta H_{\rm B}$ corresponding to $\mu^{\theta}_{\rm max}$ and $\mu^{\theta}_{\rm min}$. In our case, this state is given by

$$|1_{\rm opt}\rangle = \frac{1}{\sqrt{2}}\left(|1_{\Omega^{\theta}_{\rm max}}\rangle + e^{i\varphi}\,|1_{\Omega^{\theta}_{\rm min}}\rangle\right), \tag{11}$$

wherein the frequencies are given by the solutions of $\mathcal{X}^{\theta}_{\rm M}(\Omega^{\theta}_{\rm max/min}) = \mu_{\rm max/min}$, where $e^{i\varphi}$ is an arbitrary phase. For a degenerate $\partial_\theta H_{\rm B}$ with nonunique states $|1_{\Omega^{\theta}_{\rm max/min}}\rangle$, any such superposition will saturate the bound. In the continuous frequency space, these states are represented by a combination of Dirac delta functions with peaks located at the frequencies $\Omega^{\theta}_{\rm max/min}$.

We now obtain the ultimate precision of estimating the linewidth Γ of the single-photon transition, and the detunings Δ_j in the SES of the emitter.

Γ *estimation*. Using Eq. (7) and that $H_{\rm M}$ is independent of Γ, $\partial_\Gamma \chi_{\rm M}(\omega) = \chi_{\rm M}(\omega)/\Gamma$. Noting that $-1 \leqslant \frac{2\chi_{\rm M}(\omega)}{1+\chi_{\rm M}(\omega)^2} \leqslant 1$, where the equalities hold when $\chi_{\rm M}(\omega) = \pm 1$, the maximum and the minimum values of $\mathcal{X}^{\Gamma}_{\rm M}(\omega)$ are $\pm 1/\Gamma$. This leads to

$$\mathcal{Q}(1_{|\xi_{\rm out}\rangle}, \Gamma) \leqslant (2/\Gamma)^2. \tag{12}$$

Curiously, this upper bound has no explicit dependence on the emitter Hamiltonian $H_{\rm M}$ or the SES vector $|\gamma\rangle$, allowing us to conclude that quantum mechanical bound on precision of lifetime estimation only depends on the level structure of $H_{\rm M}$ insofar as its effect on the Weisskopf-Wigner rate Γ corresponding to the emitter.

The optimal pulse that saturates this bound is an equally weighted superposition of two frequency-mode states with

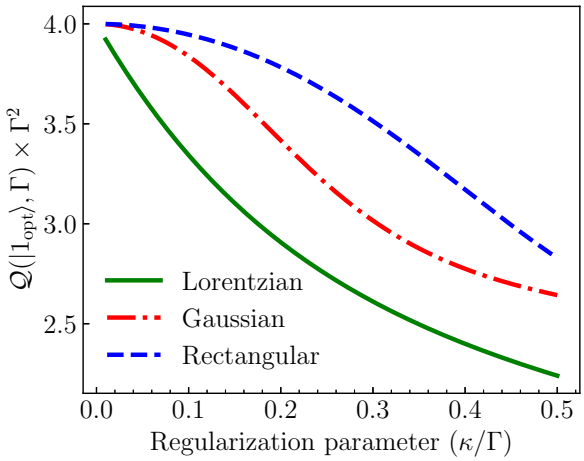

FIG. 2. The dimensionless QFI of estimating Γ for a TLS using regularized optimal incident pulses as a function of the regularization parameter κ/Γ. The regularized delta functions are defined as the Lorentzian $\delta_\kappa(\omega) = \kappa/[\pi(\kappa^2+\omega^2)]$, the Gaussian $\delta_\kappa(\omega) = e^{-\frac{\omega^2}{2\kappa^2}}/(\kappa\sqrt{2\pi})$, and, the rectangular $\delta_\kappa(\omega) = \Theta(\omega)\Theta(\kappa-\omega)/\kappa$.

frequencies $\Omega^{\Gamma}_{\rm max/min}$, which satisfy $\chi_{\rm M}(\omega) = \pm 1$. These equations have real solutions in ω for a general emitter system, ensuring the bound is tight (see *Appendix B: Proof of existence of solutions to* $\chi_M(\omega) = \pm 1$ for details).

For the specific case of a two-level system (TLS), these solutions are given by $\Omega^{\Gamma}_{\rm max/min} = \Delta \pm \Gamma/2$, where $\Delta = \omega_0 - \omega_c$ is the detuning and ω_0 is the transition frequency of the TLS. According to Eq. (11) the optimal incident pulse for a TLS is

$$|1_{\rm opt}\rangle = \frac{1}{\sqrt{2}}\left[\,|1_{\Delta+\frac{\Gamma}{2}}\rangle + e^{i\varphi}\,|1_{\Delta-\frac{\Gamma}{2}}\rangle\,\right]. \tag{13}$$

This optimal incident pulse is expressed in the frequency domain by a combination of two delta functions $|\tilde{\xi}_{\rm opt}(\omega)|^2 = [\delta(\omega-\Delta+\Gamma/2)+\delta(\omega-\Delta-\Gamma/2)]/2$. Engineering such a pulse shape would be experimentally challenging due to the singular nature of the spectral profile. However, the ultimate precision can be approached with a regularized spectral profile of the pulse. In Fig. 2 we plot the QFI for the optimal incident pulse constructed by three distinct regularized representations of the Dirac delta $\delta_\kappa(\omega)$ where $\delta(\omega) = \lim_{\kappa\to 0}\delta_\kappa(\omega)$ as a function of the dimensionless regularization parameter κ/Γ. For all these representations, the QFI monotonically approaches the upper bound in the limit $\kappa \ll \Gamma$.

Detuning estimation. Spectroscopy also involves estimating the tranistion energies of the emitter Hamiltonian $H_{\rm M}$. We now present ultimate precisions for estimating these detunigs $\Delta_j = \langle j|\, H_{\rm M}\, |j\rangle = \omega_j - \omega_c$ (ω_j is the energy of the jth state), defined as the diagonal elements of $H_{\rm M}$ in the SES basis $\{|j\rangle\}$, see Fig. 1.

Differentiating $\chi_{\rm M}(\omega)$ from Eq. (7) with respect to Δ_j we get $\partial_{\Delta_j}\chi_{\rm M}(\omega) = \Gamma|\,\langle j|\,[\![\omega \mathbf{I}_{\rm M} - H_{\rm M}]\!]^{-1}\,|\gamma\rangle\,|^2/2$. This indicates that the function $\mathcal{X}^{\Delta_j}_{\rm M}(\omega)$ is nonnegative and it approaches the minima zero for $\omega \ll \epsilon_{\rm min}$ or $\omega \gg \epsilon_{\rm max}$, where $\epsilon_{\rm min\,/\,max}$ are the minimum and maximum eigenvalues of $H_{\rm M}$. The global maxima of $\mathcal{X}^{\Delta_j}_{\rm M}(\omega)$, denoted as $\mu^{\Delta_j}_{\rm max}$, depends on the emitter parameters and can be calculated for any

emitter system. This leads to the following precision bound $\mathcal{Q}(|1_{\xi_{\text{out}}}\rangle, \Delta_j) \leqslant (\mu_{\max}^{\Delta_j})^2$.

For a TLS, this upper bound simplifies to $\mathcal{Q}(|1_{\xi_{\text{out}}}\rangle, \Delta) \leqslant (4/\Gamma)^2$. The corresponding optimal pulse is given by $|1_{\text{opt}}\rangle = \frac{1}{\sqrt{2}}[|1_\Delta\rangle + e^{i\varphi}|1_\infty\rangle]$, where $|1_\Delta\rangle$ is a frequency-mode state resonant with the detuning Δ and $|1_\infty\rangle$ is a frequency-mode state far from resonance.

Conclusion. We identified the ultimate precision of estimating two key spectroscopic parameters as summarized in Table I. The corresponding optimal incident pulses are shown to be combinations of delta functions in the frequency domain whose locations are specific to the emitter system and the estimation task.

Our theoretical limit of $4/\Gamma^2$ for Γ estimation surpasses the maximum precision of about $2.5/\Gamma^2$ achieved for a TLS with both Gaussian and rectangular input pulses, and the maximum of $2/\Gamma^2$ achieved for decaying and rising exponential pulses [17], Fig. 2]. Indeed, the pulse which is most efficiently absorbed (rising exponential) is not the most informative spectroscopically [11,17,32,33].

A recent study with pulsed coherent light probing a TLS revealed that the QFI per photon respects the same bound [34] of $4/\Gamma^2$ for estimating Γ. This suggests that our bounds are valid beyond single-photon pulses. Future research may investigate the validity of our bounds for other quantum probe states and more general emitter states, as well as for imperfect couplings of the traveling light state to the matter system.

While ideal emitter-pulse coupling is assumed here, practical spectroscopy is limited by detector imperfections, such as dark counts, and nonradiative channels. These factors are expected to reduce the maximum precision from $4/\Gamma^2$. For instance, when a TLS is coupled to phonons, the QFI for estimating Γ is approximately the product of the bare TLS QFI and the Franck-Condon factor for the zero-phonon line [35], Eq. (13)]. As the coupling strength increases, the latter decay, causing the maximum QFI to drop from $4/\Gamma^2$. However, the optimal pulse shape remains the same as in Table I because the incident photon only influences the bare TLS QFI which is maximized in this study.

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Data availability. No data were created or analyzed in this study.

Appendix A: Microscopic derivation of the light-matter hamiltonian. In this Appendix, we derive the total light-matter Hamiltonian in Eq. (3) from the dipole interaction Hamiltonian $H_{\text{MP}} = -\vec{\mathbf{d}} \cdot \vec{\mathbf{E}}$. The dipole operator of our emitter system takes the following form:

$$\vec{\mathbf{d}} = \sum_{k=1}^{N} \vec{d}_{gk} \, |k\rangle \langle g| + \text{H.c.}, \tag{A1}$$

where $\vec{d}_{gk} = \langle g| \vec{\mathbf{d}} \, |k\rangle$ are the relevant matrix elements of the dipole operator and $\{|k\rangle\}$ is a basis of the SES. Note that this dipole operator neglects the matrix elements within the SES. This is physically valid because the energy separation within the singly excited states is always lower than the energy separations between the ground state and the excited states with which the optical field is resonant. However, the transitions within the excited states can happen due to the dipole-dipole coupling within the molecule which are accounted for by the bare emitter Hamiltonian $\bar{H}_{\text{M}}$. Since we assume the ground state has zero energy, $\bar{H}_{\text{M}}$ has support only in the SES. Thus, the total light-matter Hamiltonian can be written as $H = \bar{H}_{\text{M}} + H_{\text{P}} + H_{\text{MP}}$.

Transforming to the interaction picture with respect to the static Hamiltonian $H_0 = \omega_c \sum_{k=1}^{N} |k\rangle \langle k| + H_{\text{P}}$ and rotating away the rapidly oscillating terms, the total light-matter Hamiltonian takes the following form:

$$H(t) = H_{\text{M}} - i\Bigg[\sum_{k=1}^{N} D_k \, |g\rangle \langle k| \otimes \int_0^\infty d\omega \, \mathcal{E}(\omega) a(\omega) e^{-i(\omega-\omega_c)t} - \text{H.c.}\Bigg], \tag{A2}$$

where $D_k = \vec{d}_{gk} \cdot \hat{\mathbf{e}}$, and the amplitude of the electric field $\mathcal{E}(\omega) = \sqrt{\frac{\omega}{4\pi\epsilon_0 cA}}$. $H_{\text{M}} = \bar{H}_{\text{M}} - \omega_c \sum_{k=1}^{N} |k\rangle \langle k|$ is the detuned emitter Hamiltonian which is referred to as the emitter Hamiltonian in the main text for brevity.

Now, we impose the white-noise approximation which is often referred to as the Markovian approximation in quantum optics, which assumes that the incident pulse is sufficiently narrow-band around the carrier frequency ω_c. Under this approximation, the amplitude of the electric-field operator becomes only dependent on ω_c and the limits to the integral over the frequency modes can be set to $-\infty$ to $+\infty$. Thus, we can write

$$H(t) = H_{\text{M}} - i\sqrt{\Gamma}[\Sigma^\dagger a(t) - \Sigma a^\dagger(t)], \tag{A3}$$

where the coupling strength

$$\Gamma = \left(\frac{\omega_c}{4\pi\epsilon_0 cA}\right) \times \sum_{k=1}^{N} |D_k|^2,$$

and $\Sigma = |\gamma\rangle \langle g|$ where $|\gamma\rangle$ is a normalized state in the SES expressed as $|\gamma\rangle \propto \sum_{k=1}^{N} D_k \, |k\rangle$ where the normalization factor is absorbed in Γ.

Appendix B: Proof of existence of solutions to $\chi_M(\omega) = \pm 1$. Writing the emitter Hamiltonian in spectral decomposition $H_{\text{M}} = \sum_{k=1}^{N} \epsilon_k \, |\epsilon_k\rangle \langle \epsilon_k|$, where N is the dimension of the singly excited subspace of the emitter, we can get the following expression: $\chi_{\text{M}}(\omega) = \frac{\Gamma}{2} \sum_{k=1}^{N} \frac{|\langle \gamma|\epsilon_k\rangle|^2}{\omega - \epsilon_k}$. Note that $\chi_{\text{M}}(\omega)$ has poles at the frequencies $\omega = \epsilon_k$. Thus, $\chi_{\text{M}}(\omega) \to +\infty$ when $\omega \to \epsilon_k^+$ and $\chi_{\text{M}}(\omega) \to -\infty$ when $\omega \to \epsilon_k^-$ for all k. Therefore, $\chi_{\text{M}}(\omega)$ is continuous in the interval $(\epsilon_k, \epsilon_{k+1})$ and has a range of $(-\infty, +\infty)$. Thus, applying the intermediate value theorem we can state $\chi_{\text{M}}(\omega)$ has at least 1 real solution to $\chi_{\text{M}}(\omega) = c$ for any real number c. This proves that the equations $\chi_{\text{M}}(\omega) = \pm 1$ have real solutions in ω.

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