Optimal two-photon excitation of bound states in non-Markovian waveguide QED

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(Received 19 March 2021; revised 25 May 2021; accepted 7 June 2021; published 7 July 2021)

Bound states arise in waveguide QED systems with a strong frequency-dependence of the coupling between emitters and photonic modes. While exciting such bound-states with single-photon wave-packets is not possible, photon-photon interactions mediated by the emitters can be used to excite them with two-photon states. In this Letter, we use scattering theory to provide upper limits on this excitation probability for a general non-Markovian waveguide QED system and show that this limit can be reached by a two-photon wave packet with vanishing uncertainty in the total photon energy. Furthermore, we also analyze multi-emitter waveguide QED systems with multiple bound states and provide a systematic construction of two-photon wave packets that can excite a given superposition of these bound states. As specific examples, we study bound-state trapping in waveguide QED systems with single and multiple emitters and a time-delayed feedback.

DOI: 10.1103/PhysRevA.104.013705

I. INTRODUCTION

Waveguide quantum electrodynamics (wQED) [1–5] studies the interaction of quantum emitters with one-dimensional bosonic waveguide fields. While traditional analysis of wQED systems assumes a Markovian (frequency-independent) coupling of emitters and the waveguide mode [6–10], there has been recent theoretical interest in exploring physics of non-Markovian wQED systems [11–24]. Furthermore, there have been proposals as well as experimental implementations of non-Markovian systems with circuit QED [25–28] and cold atoms [29]. Several recent works have attempted to understand the dynamics of wQED systems with time-delays comparable to or larger than the lifetime of the emitters. Such non-Markovian wQED systems support a rich variety of physical phenomena including existence of bound states in continuum [17,30–32], superradiance and subradiance in the presence of time delays [18–21] as well as generation of highly entangled photonic states [23,24,33,34]. Furthermore, there is a possibility of using these physical phenomena for quantum technology applications such as quantum memory [17] and quantum computation with cluster states [34].

Of particular interest in non-Markovian wQED is the existence of single-excitation polaritonic bound states, which are normalizable eigenstates of the wQED Hamiltonian. Such bound states have been extensively studied in systems where the waveguide mode has a band gap, with the bound-state energy lying in this band gap [16,35,36]. However, a number of non-Markovian wQED systems can support bound states at frequencies that can propagate in the waveguide, i.e., they support a bound state in the continuum [17,30–32]. While these bound states cannot be excited with single waveguide photons, the emitter-mediated photon-photon interactions can allow two (or more) waveguide photons to excite them [17,30,37,38]. From a technological standpoint, this opens up the possibility of storing quantum information being carried by two-photon wave packets into the bound states. Consequently, several authors have performed analytical and numerical studies to design the two-photon wave packet that can optimally excite the bound state in a variety of non-Markovian systems [17,30]. However, it remains unclear what the limits on bound-state trapping probabilities are, and if there is a systematic design procedure for the optimal incident two-photon wave packet that reaches this limit.

In this paper, we use quantum-scattering theory to rigorously answer this question for a general non-Markovian wQED system. Our approach relies on an analytical calculation of the two-photon scattering matrix element capturing the bound-state trapping process. Using this scattering matrix, we provide an upper limit on the bound-state trapping probability. Furthermore, we show that this limit is asymptotically tight, i.e., it is reached by a two-photon wave packet with vanishing uncertainty in the total photon energy. Finally, as storage protocols for quantum information encoded in the incoming two-photon wave packets, we consider multi-emitter wQED

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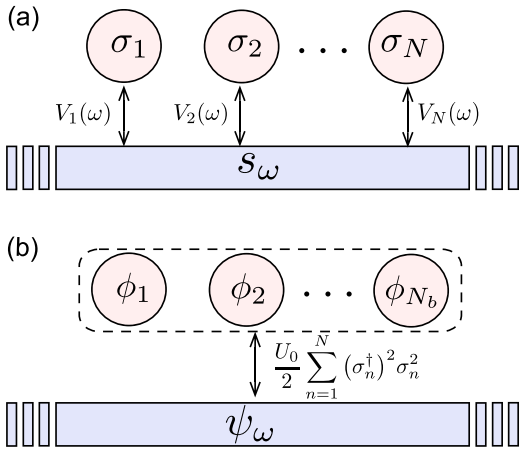

FIG. 1. (a) Schematic of the non-Markovian waveguide QED system with N emitters. The frequency-dependent coupling constant $V_n(\omega)$ capture the non-Markovian nature of the emitter-waveguide interactions. (b) An equivalent picture of the waveguide QED system when expressed in terms of the scattering state modes and the bound-state modes which are coupled to each other due to the two-particle repulsion at the qubit modes.

systems that can support more than one bound states and systematically outline the design of two-photon wave packets to excite superpositions of these bound states. As specific examples, we study bound-state trapping in waveguide QED systems with single and multiple emitters and a time-delayed feedback.

This paper is organized as follows: the two-photon scattering matrix is analytically calculated in Sec. II, and the theoretical and numerical analysis of the bound-state trapping process is presented in Sec. III.

II. SCATTERING THEORY

The wQED system under consideration is shown in Fig. 1(a)—N emitters modeled as anharmonic oscillators at frequencies $\omega_1, \omega_2, \ldots, \omega_N$ with annihilation operators $\sigma_1, \sigma_2, \ldots, \sigma_N$ couple with coupling constant $V_1(\omega), V_2(\omega), \ldots, V_N(\omega)$ to a waveguide mode with annihilation operator s_ω. The frequency dependence of the coupling constants, in either magnitude or in phase or both, gives rise to non-Markovian effects in the dynamics of the emitters. For instance, a Lorentzian frequency dependence of the coupling constant corresponds to non-Markovianity induced due to a single mode cavity coupling to the emitters [39], while a frequency dependent phase [$V_n(\omega) \sim e^{i\omega x_n}$] corresponds to non-Markovianity due to retardation effects [40,41].

The dynamics of this system can be described by a Hamiltonian expressible as $H = H_0 + V$ where H_0 is a quadratic form that describes the interaction of the emitters with the waveguide:

$$H_0 = \int_{-\infty}^{\infty} \omega s_\omega^\dagger s_\omega d\omega + \sum_{n=1}^{N} \omega_n \sigma_n^\dagger \sigma_n + \int_{-\infty}^{\infty} \sum_{n=1}^{N} [V_n(\omega) s_\omega \sigma_n^\dagger + V_n^*(\omega)\sigma_n s_\omega^\dagger] \frac{d\omega}{\sqrt{2\pi}}, \quad (1)$$

and V captures the anharmonicity of the emitters:

$$V = \sum_{n=1}^{N} \frac{U_0}{2} (\sigma_n^\dagger)^2 \sigma_n^2. \quad (2)$$

It can be noted that two-level emitters are obtained in the limit of infinite anharmonicity ($U_0 \to \infty$).

The quadratic Hamiltonian H_0 can be diagonalized into the sum of a continuum of *scattering states* with annihilation operators ψ_ω at frequencies $\omega \in \mathbb{R}$ and discrete *bound states* with annihilation operators $\phi_1, \phi_2, \ldots, \phi_{N_b}$ at frequencies $\omega_1, \omega_2, \ldots, \omega_{N_b}$ [Fig. 1(b)]:

$$H_0 = \sum_{\alpha=1}^{N_b} \omega_\alpha \phi_\alpha^\dagger \phi_\alpha + \int_{-\infty}^{\infty} \omega \psi_\omega^\dagger \psi_\omega d\omega. \quad (3)$$

These modes, by definition, satisfy the commutation relations $[\psi_\omega, \psi_\nu^\dagger] = \delta(\omega - \nu)$, $[\phi_\alpha, \phi_\beta^\dagger] = \delta_{\alpha,\beta}$, $[\psi_\omega, \phi_\alpha^\dagger] = 0$. Physically, the bound states mode in waveguide QED systems would correspond to quantum states whose overlap with the waveguide mode vanishes at distances away from the emitter. The scattering state modes, on the other hand, are plane waves propagating towards and away from the emitters when examined at large distances from the emitters.

The scattering state modes and the bound-state modes, while decoupled in the Hamiltonian H_0, are coupled due to the anharmonicity of the emitters [Eq. (2)]. Furthermore, the annihilation operators σ_n for the emitters can be expressed in terms of the bound-state operator and scattering state operators:

$$\sigma_n = \sum_{\alpha=1}^{N_b} \varepsilon_n^\alpha \phi_\alpha + \int_{-\infty}^{\infty} \xi_n(\omega) \psi_\omega d\omega, \quad (4)$$

where ε_n^α captures the overlap of the α^{th} bound-state mode with the nth emitter and $\xi_n(\omega)$ captures the overlap of the scattering state mode at frequency ω with the nth emitter.

Consider now the process of exciting the emitters with an incident two-photon state and trapping one photon in a bound state. The probability amplitude associated with this process is captured by the scattering matrix element $S_\alpha(\omega; \nu_1, \nu_2)$, which is the probability amplitude of trapping a photon in the α^{th} bound-state and scattering the second photon in a scattering state at frequency ω on excitation with two photons at frequency ν_1 and ν_2:

$$S_\alpha(\omega; \nu_1, \nu_2) = \lim_{\substack{t_i \to -\infty \\ t_f \to \infty}} \langle \mathrm{G} | \, \phi_\alpha \psi_\omega U_I(t_f, t_i) \psi_{\nu_1}^\dagger \psi_{\nu_2}^\dagger \, | \mathrm{G} \rangle \,, \quad (5)$$

where $U_I(\cdot, \cdot)$ is the interaction picture propagator for the Hamiltonian H with respect to H_0 and $|\mathrm{G}\rangle$ is the ground state of the wQED system. An exact analytical expression relating this scattering matrix to ε_n^α and $\xi_n(\omega)$ can be derived by following a procedure similar to Ref. [13]. Since $U_I(t_f, t_i) = e^{iH_0 t_f} e^{-iH(t_f - t_i)} e^{-iH_0 t_i}$, $S_\alpha(\omega; \nu_1, \nu_2)$ can be expressed in terms of Heisenberg picture operators with respect

to the Hamiltonian H_0:

$$\langle \mathrm{G}|\,\phi_\alpha\psi_\omega U_I(t_f,t_i)\psi^\dagger_{\nu_1}\psi^\dagger_{\nu_2}\,|\mathrm{G}\rangle = \langle \mathrm{G}|\,\mathcal{T}\left[\phi_\alpha(t_f)\psi_\omega(t_f)\exp\left(-\frac{iU_0}{2}\int_{t_i}^{t_f}\sum_{n=1}^{N}\sigma_n^{\dagger 2}(\tau)\sigma_n^2(\tau)d\tau\right)\psi^\dagger_{\nu_1}(t_i)\psi^\dagger_{\nu_2}(t_i)\right]|\mathrm{G}\rangle\,, \tag{6}$$

where $\mathcal{T}$ is the time-ordering operator. A Dyson series expansion of the exponential then yields

$$S_\alpha(\omega;\nu_1,\nu_2) = \sum_{k=1}^{\infty}\frac{1}{k!}\left(-\frac{iU_0}{2}\right)^k\mathcal{G}^k_\alpha(\omega;\nu_1,\nu_2), \tag{7}$$

where

$$\begin{aligned}\mathcal{G}^k_\alpha(\omega;\nu_1,\nu_2) &= \lim_{\substack{t_i\to-\infty\\ t_f\to\infty}} e^{i\theta(t_f,t_i)}\int_{\tau_1,\ldots,\tau_k=t_i}^{t_f}\langle \mathrm{G}|\,\mathcal{T}\left[\phi_\alpha(t_f)\psi_\omega(t_f)\psi^\dagger_{\nu_1}(t_i)\psi^\dagger_{\nu_2}(t_i)\prod_{n=1}^{k}\sum_{m=1}^{N}\sigma_m^{\dagger 2}(\tau_n)\sigma_m^2(\tau_n)\right]|\mathrm{G}\rangle\, d\tau_1\cdots d\tau_k,\\ &= \lim_{\substack{t_i\to-\infty\\ t_f\to\infty}} e^{i\theta(t_f,t_i)}k!\int_{\tau_1>\tau_2\cdots>\tau_k=t_i}^{t_f}\sum_{m_1,m_2,\ldots,m_k=1}^{N}\langle \mathrm{G}|\,\phi_\alpha(t_f)\psi_\omega(t_f)\left[\prod_{n=1}^{k}\sigma_{m_n}^{\dagger 2}(\tau_n)\sigma_{m_n}^2(\tau_n)\right]\psi^\dagger_{\nu_1}(t_i)\psi^\dagger_{\nu_2}(t_i)\,|\mathrm{G}\rangle\, d\tau_1\cdots d\tau_k,\end{aligned} \tag{8}$$

with $\theta(t_f,t_i) = (\omega_\alpha+\omega)t_f - (\nu_1+\nu_2)t_i$. We point out that in order to use the Dyson series expansion to nonperturbatively compute the scattering matrix, it is necessary to clarify its regime of convergence. Unlike most problems dealt with in scattering theory, we note that the problem that we consider is simpler in the sense that the operator appearing in the exponential in Eq. (6) is a bounded operator (when restricted to the two-excitation subspace, which is the setting considered in this paper). Consequently, the Dyson expansion of the propagator converges for *arbitrarily large* U_0 at finite t_i and t_f. Furthermore, the analytical result derived in this section is confirmed with numerical simulations in Sec. III. However, our analysis does indeed fall short of a rigorous proof of the existence and convergence of a scattering theory for this problem, and this is an issue we seek to resolve more generally for such models in future work.

To evaluate $\mathcal{G}^k_\alpha(\omega;\nu_1,\nu_2)$, we note that since the operators ϕ_α and ψ_ω diagonalize the Hamiltonian H_0, $\phi_\alpha(t) = \phi_\alpha e^{-i\omega_\alpha t}$ and $\psi_\omega(t) = \psi_\omega e^{-i\omega t}$. Furthermore, it follows from Eq. (4) that

$$\sigma_n(t) = \sum_{\alpha=1}^{N_b}\varepsilon_n^\alpha\phi_\alpha e^{-i\omega_\alpha t} + \int_{-\infty}^{\infty}\xi_n(\omega)e^{-i\omega t}\psi_\omega d\omega. \tag{9}$$

The evaluation of the expectation in Eq. (8) can now easily be done by using the commutators for ϕ_α, ψ_ω:

$$\begin{aligned}&\langle \mathrm{G}|\,\phi_\alpha(t_f)\psi_\omega(t_f)\left[\prod_{n=1}^{k}\sigma_{m_n}^{\dagger 2}(\tau_n)\sigma_{m_n}^2(\tau_n)\right]\psi^\dagger_{\nu_1}(t_i)\psi^\dagger_{\nu_2}(t_i)\,|\mathrm{G}\rangle\\ &\quad= e^{-i\theta(t_f,t_i)}2^{k+1}\varepsilon^{\alpha *}_{m_1}\xi^*_{m_1}(\omega)\xi_{m_k}(\nu_1)\xi_{m_k}(\nu_2)\\ &\qquad\times e^{-i(\omega+\omega_\alpha)\tau_1}e^{i(\nu_1+\nu_2)\tau_k}\left[\prod_{n=1}^{k-1}G^2_{m_n,m_{n+1}}(\tau_n-\tau_{n+1})\right],\end{aligned} \tag{10}$$

where

$$\begin{aligned}G_{m,n}(t) &= [\sigma_m(t),\sigma_n^\dagger(0)]\\ &= \sum_{n=1}^{N_b}\varepsilon_m^\alpha\varepsilon_n^{\alpha *}e^{-i\omega_\alpha t} + \int_{-\infty}^{\infty}\xi_m(\omega)\xi_n^*(\omega)e^{-i\omega t}d\omega.\end{aligned} \tag{11}$$

Using this result along with (8), we obtain

$$\begin{aligned}\mathcal{G}^k_\alpha(\omega;\nu_1,\nu_2) &= 2^{k+2}\pi\delta(\omega+\omega_\alpha-\nu_1-\nu_2)\sum_{m,n=1}^{N}\varepsilon_m^{\alpha^*}\xi_m^*(\omega)\\ &\quad\times[\mathbf{T}^{k-1}(\omega+\omega_\alpha+i0^+)]_{m,n}\xi_n(\nu_1)\xi_n(\nu_2),\end{aligned} \tag{12}$$

where $\mathbf{T}(\Omega)$ is a $N\times N$ matrix whose elements are given by

$$[\mathbf{T}(\Omega)]_{m,n} = \int_0^\infty G^2_{m,n}(t)e^{i\Omega t}dt. \tag{13}$$

Finally, substituting this expression for $\mathcal{G}^k_\alpha(\omega;\nu_1,\nu_2)$ into the series expansion in Eq. (7) and taking the limit $U_0\to\infty$, we obtain

$$S_\alpha(\omega;\nu_1,\nu_2) = \Gamma_\alpha(\omega;\nu_1,\nu_2)\delta(\omega+\omega_\alpha-\nu_1-\nu_2), \tag{14a}$$

where in the limit of $U_0\to\infty$,

$$\begin{aligned}&\Gamma_\alpha(\omega;\nu_1,\nu_2)\\ &\quad= -4\pi\sum_{m,n=1}^{N}\left(\varepsilon_m^{\alpha *}\xi_m^*(\omega)[\mathbf{T}^{-1}(\omega+\omega_\alpha+i0^+)]_{m,n}\right.\\ &\qquad\left.\times\,\xi_n(\nu_1)\xi_n(\nu_2)\right).\end{aligned} \tag{14b}$$

Here $\mathbf{T}^{-1}(\Omega)$ is the matrix inverse of $\mathbf{T}(\Omega)$ defined in Eq. (13). The δ function singularity in Eq. (14a) constrains the output photon frequency ω given input photon frequencies ν_1 and ν_2 as required by energy conservation. Furthermore, the matrix $\mathbf{T}(\Omega)$ captures the two-excitation dynamics of the multi-emitter wQED system. Finally, Eq. (14b) relate the scattering amplitude $\Gamma_\alpha(\omega;\nu_1,\nu_2)$ to this matrix and the overlap of the bound states and scattering states with the emitters.

III. BOUND-STATE EXCITATION

A. Optimal trapping of a single bound state

We now consider exciting the system with a two-photon state described by a wave function $\psi_{\mathrm{in}}(\nu_1,\nu_2)$ when expressed

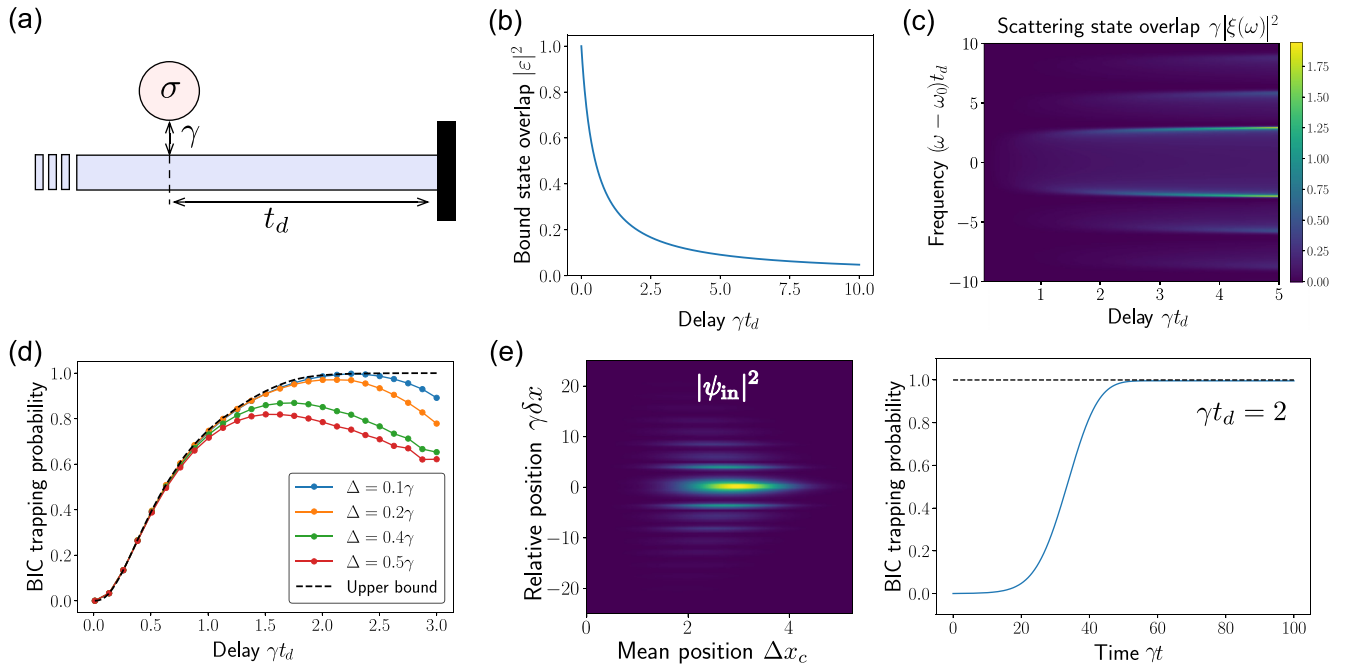

FIG. 2. Optimal excitation of bound states in time-delayed feedback system. (a) Schematic of a time-delayed feedback system with a single emitter connected to a waveguide mode terminated by a mirror. (b) Overlap of bound state with the emitter as a function of time delay t_d. (c) Overlap of the scattering state at frequency ω with the emitter for different time delays t_d. (d) Upper bound on the two-photon excitation probability (dashed black line) as a function of the delay as well as the probability obtained on using two-photon wave packets for different uncertainties in the two-photon energies. (e) Finite-difference time-domain simulations of the time-delayed feedback system with the incident two-photon state showed as a function of photon positions. It can be seen that the bound state is excited with nearly unity trapping probability with this incident two-photon state. The incident two-photon state is constructed from Eq. (19) with central two-photon energy $\Omega_0 = 2\omega_0 + 0.95\gamma$ and energy uncertainty $\Delta = 0.1\gamma$.

in terms of the scattering state modes:

$$|\psi_{\text{in}}\rangle = \frac{1}{\sqrt{2}} \int_{\nu_1,\nu_2=-\infty}^{\infty} \psi_{\text{in}}(\nu_1,\nu_2)\psi^\dagger_{\nu_1}\psi^\dagger_{\nu_2}|\text{vac}\rangle\, d\nu_1 d\nu_2. \quad (15)$$

Using the scattering matrix element in Eq. (14a), we can obtain the bound state trapping probability:

$$P_\alpha[\psi_{\text{in}}] = \frac{1}{2}\int_{-\infty}^{\infty} d\Omega \left| \int_{-\infty}^{\infty} \Gamma_\alpha(\Omega-\omega_\alpha;\nu,\Omega-\nu)\psi_{\text{in}}(\nu,\Omega-\nu)d\nu \right|^2 \quad (16)$$

We can now upper bound the trapping probability—from the Cauchy-Schwarz inequality, it follows that

$$\begin{aligned} P_\alpha[\psi_{\text{in}}] &\leqslant \frac{1}{2}\int_{-\infty}^{\infty} d\Omega \left(\int_{-\infty}^{\infty} |\Gamma_\alpha(\Omega-\omega_\alpha;\nu,\Omega-\nu)|^2 d\nu \right. \\ &\quad \left. \times \int_{-\infty}^{\infty} |\psi_{\text{in}}(\nu,\Omega-\nu)|^2 d\nu \right) \\ &\leqslant P_\alpha^{\text{ub}}, \end{aligned} \quad (17)$$

where

$$P_\alpha^{\text{ub}} = \max_{\Omega\in\mathbb{R}} \left(\frac{1}{2}\int_{-\infty}^{\infty} d\nu |\Gamma_\alpha(\Omega-\omega_\alpha;\nu,\Omega-\nu)|^2 \right). \quad (18)$$

Furthermore, it follows from Eq. (16) that an energy entangled two-photon wave packet can get arbitrarily close to this bound provided that the uncertainty in the total photon energy is sufficiently small. More specifically, consider a family of photon wave packets $\psi_{\alpha,\Delta}(\nu_1,\nu_2)$ defined by

$$\psi_{\alpha,\Delta}(\nu_1,\nu_2) = N_{\alpha,\Delta} f_{\Delta,\Omega_0}(\nu_1+\nu_2)\Gamma^*_\alpha(\Omega_0-\omega_\alpha;\nu_1,\nu_2), \quad (19)$$

where $f_{\Delta,\Omega_0}(\nu) = (\pi\Delta^2)^{-1/4}\exp[-(\nu-\Omega_0)^2/2\Delta^2]$ determines the distribution of two-photon energy, Ω_0 is the central two-photon energy chosen as the frequency that maximizes the right-hand side of Eq. (18) and $N_{\alpha,\Delta}$ is chosen to normalize the wave packet. It then follows from Eq. (16) that as $\Delta \to 0$, $P_\alpha[\psi_{\alpha,\Delta}] \to P_\alpha^{\text{ub}}$. Physically, Δ is the uncertainty in the total photon energy of the wave packet, and consequently governs the spatial spread in the center of mass of the two photons, i.e., a lower Δ would imply a larger spread of the two-photon wave packet in space. The two-photon energy Ω_0 that is needed to optimally excite the bound state, obtained by maximizing the function in Eq. (18), is typically different from the resonant frequency of the emitter. This can be attributed to the nonzero lamb-shift that the emitter frequency in the presence of a frequency dependent coupling to the waveguide mode.

As a concrete example, we consider a wQED system with time-delayed feedback as shown in Fig. 2(a). This system is equivalent to a non-Markovian waveguide QED system with one emitter and $V(\omega) = 2i\sqrt{\gamma}\sin(\omega t_d)$. If the qubit transition frequency ω_0 satisfies $\omega_0 t_d = n\pi$ for some integer n, then this system supports one bound-state mode. Furthermore, the overlap of the qubit mode with the bound state (ε) and the

scattering state $\xi(\omega)$ can be computed by diagonalizing the quadratic part of the system Hamiltonian (refer to Appendix for details):

$$\varepsilon = \frac{1}{\sqrt{1+2\gamma t_d}} \text{ and } \xi(\omega) = \frac{2i\sqrt{\gamma}\sin(\omega t_d)}{\omega - \omega_0 + 2\gamma \sin(\omega t_d) e^{-i\omega t_d}}. \tag{20}$$

In the short delay regime ($\gamma t_d \ll 1$) the bound state is completely localized to the emitter [Fig. 2(b)]. Furthermore, in this regime the system is Markovian with vanishing coupling between the emitter and waveguide [$V(\omega) \approx V(\omega_0) = 0$] and consequently the bound-state trapping probability vanishes [Fig. 2(d)]. In the long-delay regime ($\gamma t_d \gg 1$), the overlap of the emitter with the scattering states becomes significant [Fig. 2(c)], while its overlap with the bound state vanishes. For a fixed energy uncertainty Δ, we find that the trapping probability decreases for large delays [Fig. 2(d)]—this can be attributed to the vanishing overlap of the bound state with the emitter mode which reduces the effective nonlinear interaction between the bound state and the scattering states introduced by the anharmonicity of the emitter mode. However, the upper bound P_{ub} on the trapping probability asymptotically increases to 1 as $\gamma t_d \to \infty$—we point out that to achieve this bound with an incident two-photon wave packet, the required energy uncertainty Δ scales inversely with t_d. This effectively makes the incident wave packet increasingly unconfined in space, and consequently increase its interaction time with the bound state and compensates for the its reduced nonlinear interaction with the scattering states. From a technological standpoint, this implies the existence of a trade-off between the time taken to trap the bound state and the probability with which the bound state can be excited.

Figure 2(e) shows finite difference time domain (FDTD) simulation of two-photon scattering [42] from this system for $\gamma t_d = 2$, and we indeed see that the bound state can be excited with near unity probability as predicted by the two-photon scattering theory. Furthermore, it follows from Eqs. (14b) and (19) that the spatial profile of the optimal wave packet is governed by the inverse Fourier transform of $\xi(\omega)$, the overlap of the emitter with the scattering state at frequency ω. This can be interpreted as a physical consequence of the incident two-photon wave packet, when expressed in terms of the scattering state modes, needing to maximally excite the emitter mode to nonlinearly couple into the bound state.

While our analysis suggests that there are no fundamental barriers to exciting a bound state with near-unity trapping probabilities, a concrete experimental implementation of the bound-state trapping protocol would need emitters coupled to the waveguide with high cooperativity, as well as a method to generate the incident two-photon state. First, the tradeoff between the bound-state trapping probability and excitation times outlined above will likely place stringent constraints on the emitter cooperativities, since the emitter should not have significantly decayed into channels other than the waveguide in the time it takes to trap the bound state. Such cooperativities are conceivably achievable in circuit QED setups [27], and might possibly be within the reach of quantum optical systems in near future. Second, for an optimal excitation, the wave packet needs to be an energy-entangled two-photon state—this can be generated from a coherent state using a parametric down conversion process, which is available in both circuit QED [43,44] as well as in quantum optical systems using optical nonlinearities [45]. We also point out that directly exciting the system with a coherent state can also excite the bound state [17], although this excitation will not be optimal.

B. Exciting bound-state superpositions

Multi-emitter non-Markovian wQED systems can support more than one single-excitation bound states. An incident two-photon wave packet will, in general, excite a superposition of bound states that is controllable by engineering the two-photon wave packet. This opens up the possibility of using such systems for large quantum memories, with the number of bound states determining the size of the quantum memory.

Since the scattering amplitude in Eq. (14b) suggests that the superposition of the bound states being excited depends on the overlap of $\psi_{\text{in}}(\nu_1, \nu_2)$ with $\xi_n(\nu)$, we assume the following ansatz for $\psi_{\text{in}}(\nu_1, \nu_2)$:

$$\psi_{\text{in}}(\nu_1, \nu_2) = f_{\Delta,\Omega_0}(\nu_1 + \nu_2) \sum_{n=1}^{N} c_n^{\text{in}} \xi_n^*(\nu_1)\xi_n^*(\nu_2), \tag{21}$$

where $f_{\Delta,\Omega_0}(\nu) = (\pi\Delta^2)^{-1/4} \exp[-(\nu-\Omega_0)^2/2\Delta^2]$ determines the distribution of the two-photon energy and the coefficients c_n^{in} specify the spectral distribution of the two photons. Under the assumption of negligible energy uncertainty ($\Delta \to 0$), an application of the scattering matrix in Eq. (14a) yields the following state:

$$|\psi_{\text{out}}\rangle = \sum_{\alpha=1}^{N_b} c_\alpha^{\text{out}} \int_{-\infty}^{\infty} f_{\Delta,\Omega_0}(\omega+\omega_\alpha)\psi_\omega^\dagger \phi_\alpha^\dagger |\text{G}\rangle \, d\omega, \tag{22}$$

where $\mathbf{c}_{\text{out}} = \mathbf{S}(\Omega_0)\mathbf{c}_{\text{in}}$ with $\mathbf{c}_{\text{in}}$ being a vector of c_n^{in}, $\mathbf{c}_{\text{out}}$ being a vector of c_α^{out}, and $\mathbf{S}(\Omega)$ being a matrix given by

$$[\mathbf{S}(\Omega)]_{\alpha,n} = -2\sqrt{2}\pi \sum_{m=1}^{N} \varepsilon_m^{\alpha^*} \xi_m^*(\Omega-\omega_\alpha) \times [\mathbf{T}^{-1}(\Omega+i0^+)\mathbf{X}(\Omega)]_{m,n}, \tag{23a}$$

where

$$[\mathbf{X}(\Omega)]_{m,n} = \int_{-\infty}^{\infty} \xi_m(\nu)\xi_m(\Omega-\nu)\xi_n^*(\nu)\xi_n^*(\Omega-\nu)d\nu. \tag{23b}$$

The matrix $\mathbf{S}(\Omega_0)$ maps the quantum state of an incoming two-photon wave packet expressed on the basis of the scattering state overlaps $[\xi_n^*(\nu_1)\xi_n^*(\nu_2)$ for $n \in \{1, 2, \ldots, N\}]$ to the trapped state expressed on the bound-state basis—its inverse allows us to design the incident two-photon state [Eq. (21)] to excite a specific bound-state superposition. Furthermore, if the bound states are degenerate, i.e., $\omega_\alpha = \omega_b$ for all α, then $|\psi_{\text{out}}\rangle$, is separable into this bound superposition and a single-photon in the scattering state mode with spectrum $f_{\Delta,\Omega_0}(\omega+\omega_b)$. This allows heralding of a successful trapping process by detecting the scattered single-photon with a photon-number resolving detector.

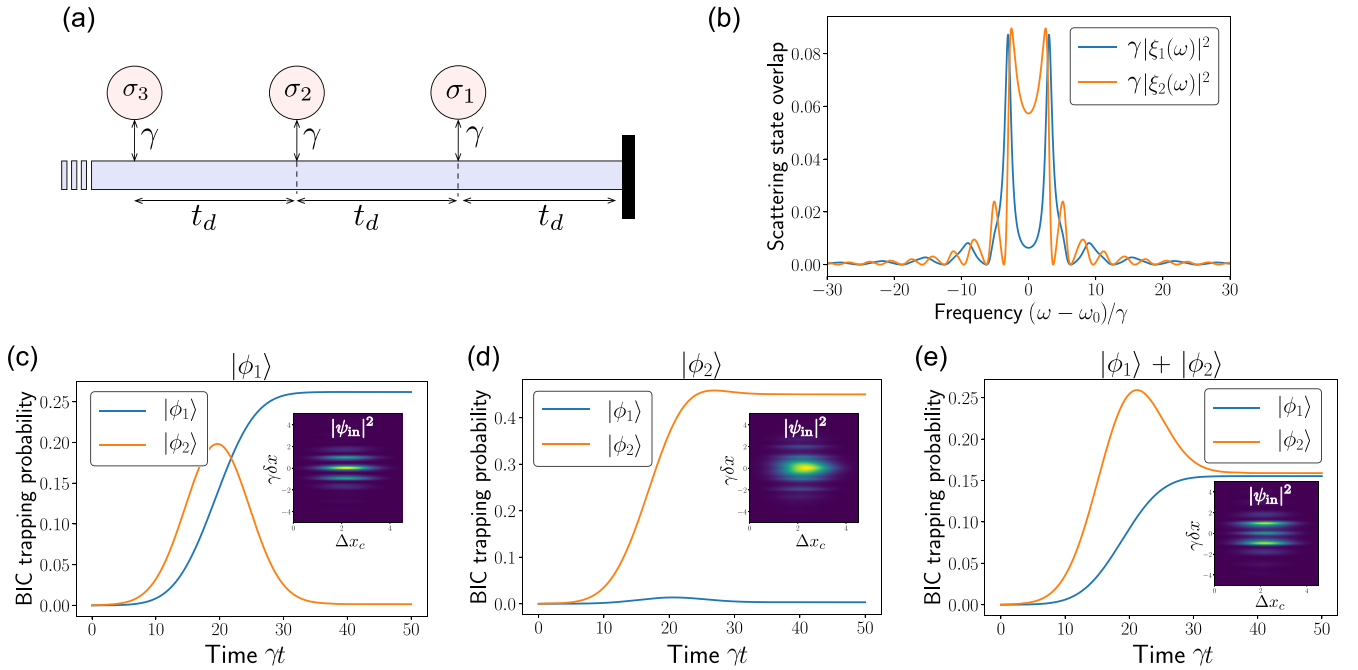

FIG. 3. Excitation of bound-state superpositions in time-delayed feedback system. (a) Schematic of a time-delayed feedback system with two emitters connected to a waveguide mode terminated by a mirror. (b) Overlap of the scattering states with the two emitters as a function of the scattering state mode frequency. Finite-difference time-domain simulations showing trapping (c) first bound state, (d) second bound state, and (e) equal superposition of the two with appropriately chosen two-photon wave packets (shown as insets). In all the simulations, the time-delay t_d is assumed to be $0.5/\gamma$, the central frequency Ω_0 of the wave packet is chosen to be 2.4γ, and the two-photon energy uncertainty Δ is chosen to be 0.15γ.

As a concrete example of exciting bound-state superpositions, we consider a time-delayed feedback system with two emitters [Fig. 3(a)]. As is shown in Appendix, assuming that both the emitters have the same resonance frequency ω_0 and that $\omega_0 t_d = n\pi$ for some integer n, this system supports two bound states. Figure 3(b) shows $\xi_1(\omega)$ and $\xi_2(\omega)$, the overlap of the scattering states with the two emitters. Figures 3(c)–3(e) shows FDTD simulations of the response of this multi-emitter system to two-photon wave packets that are designed using Eqs. (21) and (23) to excite either of the two bound states individually [Figs. 3(c)–3(d)] and an equal superposition of the two bound states [Fig. 3(e)]. We point out that the probability of trapping the individual bound states are different [Fig. 3(c)–3(d)] due to the different overlap of the bound states with the waveguide field.

IV. CONCLUSION

Using a scattering matrix formalism, we comprehensively studied the two-photon excitation of bound states in general non-Markovian wQED systems. We provided upper limits on the two-photon excitation probability of bound states, as well as the wave packet that can achieve this upper limit. Furthermore, we also considered systems with multiple bound states and provided a formalism for constructing wave packets that can excite various superpositions of the bound states. The results in this paper not only further our understanding of bound-state excitation in wQED systems but also provide concrete quantum memory storage protocols using these systems.

ACKNOWLEDGMENTS

We thank Francesco Ciccarello and Leo Fang for insightful discussions. R.T. acknowledges support from the Kailath Graduate Fellowship and the Germany Research and Internship Programme (GRIP) award. D.M. acknowledges funding from ERC Advanced Grant QENOCOBA under the EU Horizon 2020 program (Grant Agreement No. 742102). R.T., S.F., and J.V. acknowledge funding from the Air Force Office of Scientific Research under AFOSR MURI programme (Award No. FA9550-17-1-0002).

APPENDIX: COMPUTING BOUND-STATE AND SCATTERING-STATE OVERLAP FOR TIME-DELAYED FEEDBACK SYSTEMS

In this section, we consider the calculation of bound states and scattering states of multi-emitter time-delayed feedback systems. We consider system shown in Fig. 4—N emitters with lowering operators $\sigma_1, \sigma_2, \ldots, \sigma_N$ are coupled to the forward- and backward-propagating modes of the waveguide with decay rates $\gamma_1, \gamma_2, \ldots, \gamma_N$. The waveguide mode is terminated with a perfect mirror which is at a distance t_n from the nth emitter. The quadratic part of the Hamiltonian for this system, H_0, can be expressed as

$$H_0 = \sum_{n=1}^{N} \omega_n \sigma_n^\dagger \sigma_n + \int_{-\infty}^{\infty} \omega s_\omega^\dagger s_\omega d\omega - 2i \int_{-\infty}^{\infty} \sum_{n=1}^{N} \sqrt{\gamma_n} \sin(\omega t_n)(s_\omega \sigma_n^\dagger - s_\omega^\dagger \sigma_n) \frac{d\omega}{\sqrt{2\pi}}, \quad \text{(A1)}$$

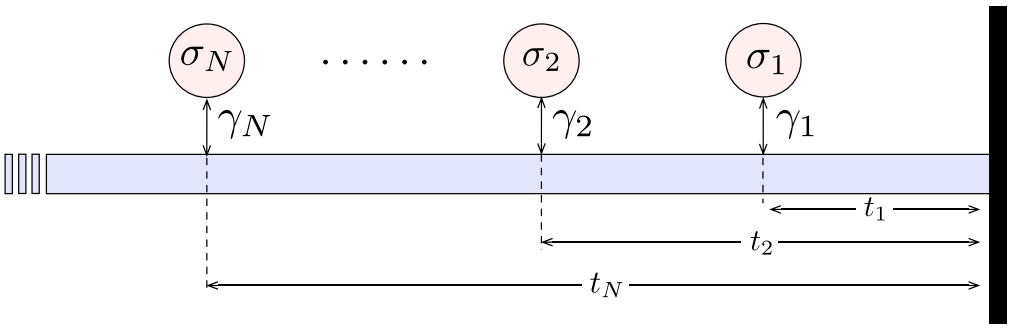

FIG. 4. Schematic of a multi-emitter waveguide-QED system with time-delayed feedback.

where ω_n is the resonance frequency of the nth emitter. Alternatively, this Hamiltonian can be expressed in terms of the position domain annihilation operator, $s_x = \int_{-\infty}^{\infty} s_\omega e^{i\omega x} d\omega/\sqrt{2\pi}$:

$$H_0 = \sum_{n=1}^{N} \omega_n \sigma_n^\dagger \sigma_n - i \int_{-\infty}^{\infty} s_x^\dagger \frac{\partial s_x}{\partial x} dx + \sum_{n=1}^{N} \sqrt{\gamma_n}\left[\left(s_{x=-t_n} - s_{x=t_n}\right)\sigma_n^\dagger + \text{H.c.}\right]. \tag{A2}$$

We first consider the calculation of the scattering states for this system. We assume the following ansatz for ψ_ω:

$$\psi_\omega = \sum_{n=1}^{N} \beta_n(\omega)\sigma_n + \int_{-\infty}^{\infty} \Psi_\omega(x) s_x dx, \tag{A3}$$

where $\beta_n(\omega)$ and $\Psi_\omega(x)$ are to be determined. Using $[\psi_\omega, H] = \omega\psi_\omega$, we obtain

$$i\frac{\partial \Psi_\omega(x)}{\partial x} + \sum_{n=1}^{N} \sqrt{\gamma_n}[\delta(x+t_n) - \delta(x-t_n)]\beta_n(\omega) = \omega\Psi_\omega(x), \tag{A4a}$$

$$\omega_n\beta_n(\omega) + \sqrt{\gamma_n}[\Psi_\omega(-t_n) - \Psi_\omega(t_n)] = \omega\beta_n(\omega). \tag{A4b}$$

With the boundary condition $\Psi_\omega(x) \to e^{-i\omega x}/\sqrt{2\pi}$ as $x \to -\infty$, the solution to Eq. (A4a) can be expressed as

$$\Psi_\omega(x) = \frac{e^{-i\omega x}}{\sqrt{2\pi}} \begin{cases} 1 & \text{for } x < -t_N \\ C_n^- & \text{for } -t_{n+1} < x < -t_n, \\ & n \in \{1, 2, \ldots, N-1\} \\ C_0 & \text{for } -t_1 < x < t_1 \\ C_n^+ & \text{for } t_n < x < t_{n+1}, \\ & n \in \{1, 2, \ldots, N-1\} \\ C_N^+ & \text{for } t_{n+1} < x. \end{cases} \tag{A5}$$

Furthermore, at the discontinuities at $x = \pm t_n$, we can set $\Psi_\omega(x) = [\Psi_\omega(x^+) + \Psi_\omega(x^-)]/2$. Integrating Eq. (A4a) across the discontinuities at $x = \pm t_n$ we obtain

$$C_n^- = 1 + \sum_{m=n+1}^{N} i\sqrt{\gamma_m}\beta_m(\omega)e^{-i\omega t_m} \quad \text{for } n \in \{1, 2 \ldots, N-1\}, \tag{A6a}$$

$$C_0 = 1 + \sum_{m=1}^{N} i\sqrt{\gamma_m}\beta_m(\omega)e^{-i\omega t_m}, \tag{A6b}$$

$$C_n^+ = 1 + \sum_{m=1}^{N} i\sqrt{\gamma_m}\beta_m(\omega)e^{-i\omega t_m} - \sum_{m=1}^{n} i\sqrt{\gamma_m}\beta_m(\omega)e^{i\omega t_m} \quad \text{for } n \in \{1, 2, \ldots, N\}. \tag{A6c}$$

Finally, using Eq. (A4b), we obtain the following system of equations for the coefficients $\beta_1(\omega), \beta_2(\omega), \ldots, \beta_N(\omega)$:

$$\begin{bmatrix} \omega - \omega_1 + 2\gamma_1 \sin(\omega t_1)e^{-i\omega t_1} & 2\sqrt{\gamma_1\gamma_2}\sin(\omega t_1)e^{-i\omega t_2} & \ldots & 2\sqrt{\gamma_1\gamma_N}\sin(\omega t_1)e^{-i\omega t_N} \\ 2\sqrt{\gamma_2\gamma_1}\sin(\omega t_1)e^{-i\omega t_2} & \omega - \omega_2 + 2\gamma_2\sin(\omega t_2)e^{-i\omega t_2} & \ldots & 2\sqrt{\gamma_2\gamma_N}\sin(\omega t_2)e^{-i\omega t_N} \\ \vdots & \vdots & \ddots & \vdots \\ 2\sqrt{\gamma_N\gamma_1}\sin(\omega t_1)e^{-i\omega t_N} & 2\sqrt{\gamma_N\gamma_2}\sin(\omega t_2)e^{-i\omega t_N} & \ldots & \omega - \omega_N + 2\gamma_N\sin(\omega t_N)e^{-i\omega t_N} \end{bmatrix} \begin{bmatrix} \beta_1(\omega) \\ \beta_2(\omega) \\ \vdots \\ \beta_N(\omega) \end{bmatrix} = \begin{bmatrix} 2i\sqrt{\gamma_1}\sin(\omega t_1) \\ 2i\sqrt{\gamma_2}\sin(\omega t_2) \\ \vdots \\ 2i\sqrt{\gamma_N}\sin(\omega t_N) \end{bmatrix}, \tag{A7}$$

which can be solved to obtain $\beta_1(\omega), \beta_2(\omega), \ldots, \beta_N(\omega)$. Finally, we note from Eq. (4) that $\xi_n(\omega) = [\sigma_n, \psi_\omega^\dagger] = \beta_n^*(\omega)$ for $n \in \{1, 2, \ldots, N\}$—consequently, $\xi_1(\omega), \xi_2(\omega), \ldots, \xi_N(\omega)$ can easily be computed once $\beta_1(\omega), \beta_2(\omega), \ldots, \beta_N(\omega)$ are known.

Next, we consider the bound states for this system. We will restrict ourselves to time-delayed feedback systems where all the emitters are at the same frequency $\omega_1 = \omega_2 = \cdots = \omega_N = \omega_0$ and the time-delays $t_1, t_2, \ldots, t_N$ all satisfy $\omega_0 t_k = n_k\pi$ for some integer n_k and for all $k \in \{1, 2, \ldots, N\}$. Under these

conditions, as is shown below, this system supports N bound states all at frequency ω_0. We assume the following ansatz for the bound-state annihilation operator ϕ_α:

$$\phi_\alpha = \sum_{n=1}^{N} v_n^\alpha \sigma_n + \int_{-\infty}^{\infty} \Phi_\alpha(x)\psi_x dx. \tag{A8}$$

Since this is a bound-state at frequency ω_0, using $[\phi_\alpha, H_0] = \omega_0\phi_\alpha$ we obtain

$$i\frac{\partial \Phi_\alpha(x)}{\partial x} + \sum_{n=1}^{N} \sqrt{\gamma_n}[\delta(x+t_n) - \delta(x-t_n)]v_n^\alpha = \omega_0 \Phi_\alpha(x). \tag{A9a}$$

$$\Phi_\alpha(t_n) = \Phi_\alpha(-t_n). \tag{A9b}$$

Furthermore, $\Phi_\alpha(x)$ is zero as $|x| \to \infty$ for it to be a bound state, and consequently the solution to this equation can be written as

$$\Phi_\alpha(x) = e^{-i\omega_0 x}\begin{cases} 0 & \text{for } x < -t_N \\ B_n^- & \text{for } -t_{n+1} < x < t_n, \\ & n \in \{1, 2, \ldots, N-1\} \\ B_0 & \text{for } -t_1 < x < t_1 \\ B_n^+ & \text{for } t_n < x < t_{n+1}, \\ & n \in \{1, 2, \ldots, N-1\} \\ B_N^+ & \text{for } x > t_{n+1}, \end{cases} \tag{A10}$$

with $\Phi_\alpha(x) = [\Phi_\alpha(x^+) + \Phi_\alpha(x^-)]/2$ at $x = \pm t_n$. It follows from integration of Eq. (A9) across the discontinuities at $x = \pm t_n$ that

$$B_n^- = \sum_{m=n+1}^{N} i\sqrt{\gamma_m} v_m^\alpha e^{-i\omega_0 t_m} \text{ for } n \in \{1, 2, \ldots, N-1\}, \tag{A11a}$$

$$B_0 = \sum_{m=1}^{N} i\sqrt{\gamma_m} v_m^\alpha e^{-i\omega_0 t_m}, \tag{A11b}$$

$$B_n^+ = \sum_{m=1}^{N} i\sqrt{\gamma_m} v_m^\alpha e^{-i\omega_0 t_m} - \sum_{m=1}^{n} i\sqrt{\gamma_m} v_m^\alpha e^{i\omega_0 t_m} \text{ for } n \in \{1, 2, \ldots, N\}. \tag{A11c}$$

We note that if $\omega_0 t_k = n_k \pi$, then $B_N^+ = 0$, indicating that $\Phi_\alpha(x) \neq 0$ only if $|x| \leqslant t_n$. Furthermore, under the assumption that all the emitters have frequency ω_0, Eq. (A9) requires that $\Phi_\alpha(-t_n) = \Phi_\alpha(t_n)$ for $n \in \{1, 2, \ldots, N\}$. This condition is already satisfied if $\Phi_\alpha(x)$ is given by Eqs. (A10) and (A11). Therefore, any choice of $v_1^\alpha, v_2^\alpha, \ldots, v_N^\alpha$ will yield a valid bound state—we thus obtain N (linearly independent) degenerate bound states at frequency ω_0.

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