Analytic and geometric properties of scattering from periodically modulated quantum-optical systems

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We study the scattering of photons from periodically modulated quantum-optical systems. For excitation-number-conserving quantum-optical systems, we connect the analytic structure of the frequency-domain \( N \)-photon scattering matrix of the system to the Floquet decomposition of its effective Hamiltonian. Furthermore, it is shown that the first-order contribution to the transmission or equal-time \( N \)-photon correlation spectrum with respect to the modulation frequency is completely geometric in nature, i.e., it depends only on the Hamiltonian trajectory and not on the precise nature of the modulation being applied.

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I. INTRODUCTION

Quantum information processing and communication systems rely heavily on the generation and manipulation of nonclassical states of light [1–12]. Implementing quantum systems for such applications often involves interfacing a localized quantum system (e.g., a few-level system such as a quantum dot or color center) with bosonic baths (such as optical fibers or waveguides). Significant control over the states of light emitted by the localized system into the bosonic bath can be gained by engineering the coupling between the two [13–15] and by controlling the excitation of the localized system [16–18]. Recently, the ability to modulate the localized system on frequency scales comparable to or exceeding the decay rate of the localized system into the bosonic bath has been demonstrated in various quantum-optical platforms such as quantum dots [19] and color centers [20,21]. This has opened up the possibility of controlling the spectral content of the photons scattered by the localized system into the bosonic bath by engineering the modulation applied on the localized system. Such spectral engineering could enable quantum networks of localized systems that are robust to variations in their physical characteristics, unlock quantum information protocols relying on high-dimensional entangled photon states [22], and realize nonreciprocal photon transport [23].

From a theoretical standpoint, it has opened up the question of how to calculate and understand the scattering properties of the modulated localized system. The scattering properties of time-independent (unmodulated) localized systems can be completely described by its scattering matrix. Significant progress has been made in developing single- and two-photon scattering matrices for specific localized systems (e.g., two-level systems and Jaynes-Cummings systems) by adapting a variety of different techniques from quantum field theory [24–27]. The problem of systematically calculating scattering and emission from a general time-independent Markovian localized system was addressed in Refs. [28,29] and it was shown that the computation of scattering matrices only required diagonalization of an effective non-Hermitian Hamiltonian that is completely restricted to the Hilbert space of the localized system. The formalism introduced can be used to derive explicit relationships between the few-photon scattering properties of the localized system and the spectrum of its effective Hamiltonian [30,31] and this has been employed to understand a number of experimentally relevant quantum systems [31,32].

While most of the efforts in calculating and understanding scattering matrices have been restricted to time-independent localized systems, a procedure for calculating the propagator from pulsed localized systems (i.e., systems whose Hamiltonian has a time dependence only within a finite-time window) was developed recently [33,34]. It was shown that it is possible to define a scattering matrix for a time-dependent system provided it is asymptotically time independent, and a recipe for its computation was provided [33]. This procedure was applied to understand scattering of a single-photon from a two-level system driven by a pulsed laser, and the scattering matrix was shown to have significantly different structure from that of a time-independent two-level system [33].

In this paper, we consider the problem of calculating the scattering matrix for a periodically modulated localized system. We focus exclusively on localized systems which are excitation-number conserving even in the presence of periodic modulation and relate the scattering matrices to the Floquet decomposition of the non-Hermitian effective Hamiltonian of the localized system. Special attention is paid to the difference in the analytic properties of the resulting scattering matrix from the scattering matrix of time-independent systems. Finally, we consider the slow-modulation regime and study the properties of the equal-time \( N \)-photon correlation function. It is shown that this correlation function, to the zeroth order in the modulation frequency, is equal to the time average of the instantaneous correlation function obtained by assuming the system to be time independent and that the first-order correction is completely geometric in nature.

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This paper is organized into three major sections. Section II introduces the mathematical model of the system under consideration along with a review of the frequency-domain scattering matrix. Section III presents the construction and general properties of the N-photon scattering matrix for a periodically modulated quantum system. As an example, single- and two-photon scattering from a cavity with Kerr nonlinearity is studied. In Sec. IV B we study the properties of the equal-time N-photon correlation function in the slow-modulation regime.

II. MODEL AND PRELIMINARIES

We consider a general class of time-dependent systems which have a periodic localized system interacting with two bosonic baths schematically depicted in Fig. 1(a). The localized system can be completely optical (e.g., nonlinear optical systems and its derivatives) and the bosonic baths is described by frequency-dependent annihilation operators $a_\omega$ and $b_\omega$. These operators satisfy the bosonic commutation relations $[a_\omega, a_{\omega'}] = 0, [b_\omega, b_{\omega'}] = 0, [a_\omega, b_{\omega'}] = \delta(\omega - \omega')$, and $[a_\omega, b_{\omega')] = [a_{\omega'}, b_\omega] = 0$. The dynamics of the system is governed by the following time-dependent Hamiltonian:

$$H(t) = H_0(t) + \sum_{\omega \in \Lambda(a, b)} \int_{-\infty}^{\infty} \omega \phi^*_\omega s_\omega(t) d\omega + \sum_{\omega \in \Lambda(a, b)} \int_{-\infty}^{\infty} i(L s_{\omega}^* - s_\omega L^\dagger) \frac{d\omega}{\sqrt{2\pi}}.$$  

Here $L$ is the operator through which the localized system couples to the bosonic baths. This model of the bosonic bath and its interaction with the localized system in Eq. (1) assumes several standard approximations made in quantum optics [35]. First, we make the Markovian approximation, which assumes that the localized system interacts equally with all the frequencies in the bosonic bath. Second, we assume that the low-dimensional system only interacts strongly with excitations close to its resonance frequency; this allows us to artificially include bosonic modes with negative frequencies in the Hamiltonian and exploit a number of mathematical simplifications that follow from this inclusion. Throughout this paper, we will consider the bath described by $a_\omega$ as the input bath and that described by $b_\omega$ as the output bath. For simplicity, we assume that the two baths couple equally to the localized system.

We will also assume the localized system and its coupling to the bosonic baths to be excitation-number conserving; this is often the consequence of a rotating-wave approximation made on the interaction between the bosonic baths and the localized system and is an excellent approximation for most quantum-optical systems [35] wherein the resonance frequencies of the localized system are much larger than its decay rates. More rigorously, this requires that (a) the Hilbert space of the localized system can be described as a direct sum of subspaces $H_0 = H_0^0 \oplus H_0^1 \oplus H_0^2 \oplus \cdots$ such that each subspace $H_0^n$ is invariant under evolution with respect to the system Hamiltonian $H_0(t)$ and (b) $L$ maps the subspace $H_0^n$ to $H_0^{n-1}$ for $n \geq 1$, $L^\dagger$ maps the subspace $H_0^n$ to $H_0^{n+1}$ for $n \geq 0$, and $L$ annihilates $H_0^0$, i.e., $H_0^0$ is within the null space of $L$. Throughout this paper, we will refer to $H_0^n$ as the $n$th-excitation subspace and associate with it an excitation number $n$. The operator $L$ then decreases the excitation number of the localized system’s state by 1 and $L^\dagger$ increases it by 1. Furthermore, evolving a state in $H_0^n$ with respect to $H_0(t)$ is identical to evolving it with respect to $H_0(t)$ without interacting with the bosonic bath; $H_0^n$ is therefore the space of the ground states of the localized system. In this paper, we will restrict ourselves to systems with a single ground state $|g\rangle$, i.e., $H_0^0 = \{ |g\rangle \}$, but make no assumption about the dimensionality of the higher-excitation subsystems.

As is shown in Appendix A, for an excitation-number-conserving system with a single ground state, $N$ photons incident on the localized system can only scatter into $N$ outgoing photons and consequently the scattering properties of the system can be described by the $N$-photon scattering matrix

$$S(\omega_1, \omega_2, \ldots, \omega_N; \nu_1, \nu_2, \ldots, \nu_N) = \left\langle \text{vac}; g | \left( \prod_{j=1}^{N} b_{\omega_j} \right) \hat{S} \left( \prod_{j=1}^{N} a_{\nu_j}^\dagger \right) \right| \text{vac}; g \right\rangle,$$

where $\hat{S}$ is the scattering-matrix defined via [36]

$$\hat{S} = \lim_{t_0 \to -\infty} U_0(t_0, t) \hat{U}(t, t_\nu) U_0(t_\nu, t_0).$$

Here $U(\cdot, \cdot)$ is the propagator corresponding to the Hamiltonian $H(t)$ and $U_0(\cdot, \cdot)$ is the propagator corresponding to the Hamiltonian $H_0(t)$ corresponding to the uncoupled localized system and bosonic baths:

$$H_0(t) = H_0(t) + \sum_{\omega \in \Lambda(a, b)} \int_{-\infty}^{\infty} \omega \phi^*_\omega s_\omega(t) d\omega.$$
Additionally, $t_0$ is a time reference that is used for defining the input and output asymptotes corresponding to the states incident and scattered from the localized system [36]. While this time reference does not affect the scattering matrix of a time-independent system, it encodes the time of arrival of the incident photon wave packet and thus is relevant for time-dependent systems. Using the input-output formalism [29,37], it can easily be shown that the scattering matrix element in Eq. (2) is related to the Heisenberg-picture system operator $L(t) = U(t_0, t)L(t, t_0)U(t, t_0)\rangle$ via (refer to Appendix B for derivation)

$$S(\omega_1, \omega_2, \ldots, \omega_N; \nu_1, \nu_2, \ldots, \nu_N) = (-1)^N e^{-i\phi(t_0)} \int_{-\infty}^{\infty} \cdots \int_{-\infty}^{\infty} G(t_1, t_2, \ldots, t_N; \nu_1, \nu_2, \ldots, \nu_N) \times \sum_{j=1}^{N} e^{i \omega(t_j - \nu_j)} \frac{dt_j ds_j}{2\pi},$$

(5)

where $\phi(t_0) = \sum_{j=1}^{N} (\omega_j - \nu_j) t_0$ and we have introduced the time-domain system Green’s function

$$G(t_1, t_2, \ldots, t_N; \nu_1, \nu_2, \ldots, \nu_N) = \langle \text{vac}; g \mid T \prod_{j=1}^{N} L(t_j) \prod_{j=1}^{N} L^\dagger(s_j) \rangle \mid \text{vac}; g \rangle,$$

(6)

where $T[\cdot]$ indicates time ordering in its arguments. Since the interaction between the localized system and the bosonic baths is Markovian in nature, an application of the quantum regression theorem [28,33] can allow us to evaluate these Green’s functions entirely within the Hilbert space of the localized system; while it conserves the total photon frequency modulo $\Omega$, this implies that the scattering matrix $S(\omega_1, \omega_2, \ldots, \omega_N; \nu_1, \nu_2, \ldots, \nu_N)$ can be written as a sum of terms proportional to $\delta(\sum_{j=1}^{N} \omega_j - \sum_{j=1}^{N} \nu_j - k\Omega)$:

$$S(\omega_1, \omega_2, \ldots, \omega_N; \nu_1, \nu_2, \ldots, \nu_N) = \sum_{k=-\infty}^{\infty} e^{-i\pi\frac{k\Omega}{2}} S_k(\omega_1, \omega_2, \ldots, \omega_N; \nu_1, \nu_2, \ldots, \nu_N) \times \delta \left( \sum_{j=1}^{N} \omega_j - \sum_{j=1}^{N} \nu_j - k\Omega \right),$$

(8)

Note that here we have explicitly shown the dependence on the time reference $t_0$ used for defining the scattering matrix (3) that enters Eq. (5) as a phase factor depending on the difference between the total input and output frequencies under consideration; the discrete time-translation symmetry of the Hamiltonian ensures that the scattering matrix is periodic in $t_0$ with period $2\pi/\Omega$. This general form of the scattering matrix can be contrasted with the scattering matrix for time-independent systems, which would be proportional to $\delta(\sum_{j=1}^{N} \omega_j - \sum_{j=1}^{N} \nu_j)$ since it conserves the total photon frequency and consequently be independent of the time reference $t_0$.

The functions $S_k(\omega_1, \omega_2, \ldots, \omega_N; \nu_1, \nu_2, \ldots, \nu_N)$ in Eq. (8) can in general be further decomposed into a sum of a nonsingular function, denoted by $S_k^C(\omega_1, \omega_2, \ldots, \omega_N; \nu_1, \nu_2, \ldots, \nu_N)$, and terms with $\delta$-function singularities. The connected part of the $N$-photon scattering matrix $S_k^C(\omega_1, \omega_2, \ldots, \omega_N; \nu_1, \nu_2, \ldots, \nu_N)$ can then be defined as

$$S_k^C(\omega_1, \omega_2, \ldots, \omega_N; \nu_1, \nu_2, \ldots, \nu_N) = \sum_{k=-\infty}^{\infty} e^{-i\pi\frac{k\Omega}{2}} S_k^C(\omega_1, \omega_2, \ldots, \omega_N; \nu_1, \nu_2, \ldots, \nu_N) \times \delta \left( \sum_{j=1}^{N} \omega_j - \sum_{j=1}^{N} \nu_j - k\Omega \right).$$

(9)

From a physical standpoint, $S_k^C(\omega_1, \omega_2, \ldots, \omega_N; \nu_1, \nu_2, \ldots, \nu_N)$ accounts for all the nonlinear interactions between the $N$ incident photons that are induced by the localized quantum system; while it conserves the total photon frequency modulo $\Omega$, it can in general lead to a change in the individual photon frequencies. Furthermore, an application of the cluster decomposition principle allows us to construct the full $N$-photon scattering matrix from its connected part and the

III. SCATTERING MATRICES

In this section, we explore the systematic construction of the frequency-domain scattering matrices (5) of the modulated quantum system. Special attention is paid to the similarities and differences that arise in these scattering matrices relative to the time-independent case. Explicit results are provided for single- and two-photon scattering matrices.
connected part of fewer-photon scattering matrices \[29,30\]
\[S(\omega_1, \omega_2, \ldots, \omega_N; v_1, v_2, \ldots, v_N) = \sum_{B} \sum_{P} \prod_{k=1}^{[N]} S^C(\omega_{B_1(P_1)}, \omega_{B_2(P_2)}; \ldots; \nu_{B_1(P_1)}, \nu_{B_2(P_2)}; \ldots),\]  
(10)

where \( B \) is an ordered partition of \( \{1, 2, 3, \ldots, N\} \) into smaller subsets, \( P \) is a permutation of \( \{1, 2, 3, \ldots, N\} \), and \( BP \) is the partition \( B \) applied on \( \{P(1), P(2), \ldots, P(N)\} \).

For a time-independent localized system, it can be shown that the frequency-domain scattering matrix is completely determined by the spectral decomposition of the effective Hamiltonian. In particular, the position and linewidth of resonances in the \( N \)-photon scattering matrix are determined by the complex eigenvalues of the effective Hamiltonian, and the amplitude of the scattering matrix at these resonances is determined by its eigenvectors. For periodically modulated localized systems, a similar relationship can be established between the scattering matrices and the Floquet decomposition of the effective Hamiltonian. Since the effective Hamiltonian is non-Hermitian, its Floquet decomposition along with Eq. (7) can be used to relate the frequency-domain scattering matrices to the Floquet decomposition of the effective Hamiltonian.

Due to the periodic time dependence of the Floquet states, the frequency-domain scattering matrices have resonances at \( \epsilon_n^\pm + p\Omega \) for \( p \in \mathbb{Z} \) with linewidths \( \kappa_n^\pm \). Furthermore, the amplitudes of these resonances are determined by the Fourier components of the periodic Floquet eigenstates \( |\phi_n^\pm(t)\rangle \) and \( |\chi_n^\pm(t)\rangle \). This is made more explicit for single- and two-photon scattering matrices in the following section.

### B. Single- and two-photon scattering matrices

Of particular interest are the single- and two-photon scattering matrices, since they can often be easily probed experimentally with transmission and two-photon correlation experiments. Following the procedure outlined above for the single-photon scattering matrix, we obtain (details in Appendix C)

\[S(\omega; v) = \sum_{k \in \mathbb{Z}} e^{-i\Delta t\Omega} S_k(v) \delta(\omega - \nu - k\Omega),\]
(13a)

with

\[S_k(v) = \sum_{m \in \mathbb{Z}} L_k^{1-0;m} \left( \frac{1}{i(\epsilon^1 + m\Omega - \nu + \kappa^1/2)} \right) L_m^{0-1}.\]
(13b)

Here \( \epsilon^1 \) and \( \kappa^1 \) are vectors of \( \epsilon_n^1 \) and \( \kappa_n^1 \), respectively, and \( D(\cdot) \) constructs a diagonal matrix from a vector that is passed as its argument. In addition, \( L_k^{1-0} \) is a row vector and \( L_k^{0-1} \) is a column vector and their elements are given by

\[
\begin{align*}
L_k^{1-0} &= \int_0^T \langle g| L|\phi_n^1(t)\rangle e^{i\Delta t\Omega} dt/m, \\
L_k^{0-1} &= \int_0^T \langle \chi_n^0(t)| L^\dagger| g\rangle e^{-i\Delta t\Omega} dt/T.
\end{align*}
(14)

Clearly, the form of the single-photon scattering matrix implies that a photon at frequency \( \nu \) is in general scattered into photons at frequencies differing from \( \nu \) by an integer multiple of \( \Omega \). Furthermore, the amplitude of transmission at these sidebands would in general depend on the Fourier-series components of the Floquet states \( |\phi_n^1(t)\rangle \) and \( |\chi_n^0(t)\rangle \).

A similar procedure can be followed for the computation of the two-photon scattering matrix. As is shown in Appendix C, the connected part of the two-photon scattering matrix \( S^C(\omega_1, \omega_2; v_1, v_2) \) can be expressed as a sum of two components \( S_{k}^{C,1}(\omega_1, \omega_2; v_1, v_2) \), which is completely determined by the Floquet decomposition of \( H_{\text{eff}}^1(t) \), and \( S_{k}^{C,2}(\omega_1, \omega_2; v_1, v_2) \), which depends on the Floquet decomposition of \( H_{\text{eff}}^2(t) \).

\[
\begin{align*}
S_{k}^{C,1}(\omega_1, \omega_2; v_1, v_2) &= \frac{1}{2\pi i} \sum_{P, Q, p, m, n \in \mathbb{Z}} \left[ L_p^{1-0;m} D\left( \frac{1}{i(\epsilon^1 - \omega_p(1) + p\Omega + \kappa^1/2)} \right) L_{m+p}^{0-1} D\left( \frac{1}{i(\epsilon^2 - \nu(2) + n\Omega + \kappa^2/2)} \right) \\
&\quad \times L_{m+p}^{1-0;n} D\left( \frac{1}{i(\epsilon^2 - \nu(2) + m\Omega + \kappa^2/2)} \right) L_{m+n}^{0-1}. \\
S_{k}^{C,2}(\omega_1, \omega_2; v_1, v_2) &= \frac{1}{2\pi i} \sum_{P, Q, p, m, n \in \mathbb{Z}} \left[ L_p^{1-0;m} D\left( \frac{1}{i(\epsilon^1 - \omega_p(1) + p\Omega + \kappa^1/2)} \right) L_{m+p}^{2-1} D\left( \frac{1}{i(\epsilon^2 - \nu(2) + n\Omega + \kappa^2/2)} \right) \\
&\quad \times L_{m+p}^{1-2;m} D\left( \frac{1}{i(\epsilon^2 - \nu(2) + m\Omega + \kappa^2/2)} \right) L_{m+n}^{0-1}. 
\end{align*}
(15a)

(15b)
where \( P \) and \( Q \) are permutations of the two-element set \{1, 2\}, \( \mathcal{P} \) indicates the principal part, \( \epsilon^2 \) and \( \kappa^2 \) are vectors of \( \epsilon_\alpha^2 \) and \( \kappa_\alpha^2 \), respectively, and \( L_n^{2-1} \) and \( L_n^{1-2} \) are matrices whose elements are given by

\[
[L_n^{2-1}]_{m,n} = \int_0^T \langle \chi_m(t) | L_n^{1}(t) | \phi_\alpha^1(t) \rangle e^{i\omega t} dt, \\
[L_n^{1-2}]_{m,n} = \int_0^T \langle \chi_m(t) | L_n^{2}(t) | \phi_\alpha^2(t) \rangle e^{i\omega t} dt.
\]

The full two-photon scattering matrix can be constructed from the connected parts in Eqs. (13) and (15) by an application of the cluster decomposition principle (10). We note that while it appears that \( S_n^{\mathcal{C}}(\omega_1, \omega_2; v_1, v_2) \) has singularities corresponding to principal parts, as shown in Appendix C, a proper evaluation of the summation removes these singularities.

As an illustrative example of this procedure, we consider the computation of the single- and two-photon scattering matrices for a cavity with Kerr nonlinearity and a periodically modulated resonance frequency. The Hamiltonian of the localized system under consideration here is given by

\[
H(t) = \Delta(t) a \dagger a + \chi(a \dagger)^2 a^2, 
\]

with a coupling operator \( L = \sqrt{\kappa/2a} \). Here \( \Delta(t) \) is the periodic modulation applied on the cavity mode, \( \chi \) is the photon-photon repulsion in the cavity due to the Kerr nonlinearity, and \( \kappa \) is the decay rate of the cavity. We assume that the mean of \( \Delta(t) \) over one period is 0. Since the \( N \)-excitation subspace for this system has dimensionality 1, there is only one solution to the Floquet problem in Eq. (11),

\[
|\phi_\alpha^N(t)\rangle = |\chi_\alpha^N(t)\rangle = e^{-iN\epsilon(t)}(a^\dagger)^N |g\rangle, \\
\epsilon^N = -\frac{iN\kappa}{2} + \chi N(N-1),
\]

where \( \psi(t) = \int_0^t \Delta(t')dt' \). With this choice of Floquet states, \( L_k^{1-0}, L_k^{0-1}, L_k^{2-1}, \) and \( L_k^{1-2} \) in Eqs. (14) and (16) reduce to scalars given by

\[
L_k^{1-0} = \left( \frac{\kappa}{2} \right)^{1/2} \alpha_k, \\
L_k^{2-1} = \sqrt{\kappa} \alpha_k, \\
L_k^{1-2} = \sqrt{\kappa} \alpha_k^*,
\]

where \( \alpha_k \) are the Fourier-series components of \( e^{-i\psi(t)} \):

\[
e^{-i\psi(t)} = \sum_{k \in \mathbb{Z}} \alpha_k e^{-i\kappa t}.
\]

Therefore, the single-photon scattering matrix (13) evaluates to

\[
S_k(v) = \sum_{m \in \mathbb{Z}} \frac{\alpha_{k+m}^{\mathcal{C}} a^{\dagger}_m}{i(m\Omega - v) + \kappa/2}.
\]

Similarly, using Eqs. (15), the two-photon scattering matrices evaluate to

\[
S_n^{\mathcal{C},1}(\omega_1, \omega_2; v_1, v_2) = \frac{\kappa^2}{2\pi} \sum_{P \subset Q, P \in \mathbb{N}, n \in \mathbb{Z}} \left[ \frac{\alpha_{P_1} a^{\dagger}_{P_1+\kappa} a^{\dagger}_{P_2-n\Omega}}{i(p\Omega - \omega_{P_1}) + \kappa/2} \right],
\]

\[
S_n^{\mathcal{C},2}(\omega_1, \omega_2; v_1, v_2) = \frac{\kappa^2}{\pi} \sum_{P \subset Q, P \in \mathbb{N}, n \in \mathbb{Z}} \left[ \frac{\alpha_{P_1} a^{\dagger}_{P_1+\kappa} a^{\dagger}_{P_2-n\Omega} a^{\dagger}_{P_2-n\Omega}}{i(p\Omega - \omega_{P_1}) + \kappa/2} \right] P^{1/2} \omega_{P_2}^{-1} - v_{Q_2}^{-1} - n\Omega.
\]

Numerical studies of the single- and two-photon transport through a modulated Kerr cavity with \( \Delta(t) = \Delta_0 \sin \Omega t \) are shown Fig. 2. Figure 2(a) shows the total single-photon transmission \( T(v) = \sum_{k \in \mathbb{Z}} |S_k(v)|^2 \) through the cavity for slow modulation (\( \Omega \ll \kappa \)) and fast modulation (\( \Omega \gg \kappa \)) of its resonant frequency. In the fast-modulation regime, the transmission spectrum shows resonances at integer multiples of \( \Omega \) with the transmission being smaller than the resonant transmission for an unmodulated cavity. In the slow-modulation regime, moderate transmissions are achieved if the input photon is within the resonant frequencies achieved by the periodic modulation (\( \pm \Delta_0, \Delta_0 \)). The amplitude \( |S_k(v)| \) of a photon...
at frequency \( \nu \) scattering into a photon at frequency \( \nu + k \Omega \) within the slow- and fast-modulation regimes is shown in Fig. 2(b). We point out that the single-photon transmissions obtained here are unaffected by the nonlinearity \( \chi \) in the cavity mode; they are identical to the classical transmission that would be obtained through a linear cavity with the same cavity mode; they are identical to the classical transmission provided in the previous section.

The connected part of the two-photon scattering matrix corresponding to the \( k \)th sideband under excitation by two photons at frequencies \( \nu_1 = \nu_2 = 0, S^C(\nu_1, \nu_2; \nu_1 = 0, \nu_2 = 0) \), is shown in Fig. 3. Since the output frequencies \( \omega_1 \) and \( \omega_2 \) of the two photons emitted into this sideband are constrained to satisfy \( \omega_1 + \omega_2 = k \Omega \), they can be completely parametrized by their frequency difference \( \delta = \omega_1 - \omega_2 \). As can be seen from Fig. 3, the amplitude of the connected part increases on increasing the nonlinearity \( \chi \). This is intuitively expected since the connected part captures the photon-photon interactions induced by the localized system. Furthermore, we note that there is an asymmetry in the amplitudes of the connected part corresponding to \( k = 1 \) and \( k = -1 \); this can be attributed to the fact that the nonlinearity \( \chi \) results in an increase in the cavity resonant frequency with the number of photons in the cavity and thus has larger contribution to one sideband as opposed to the other. Indeed, in the two-level system limit (\( \chi \to \infty \)), it can be seen from Fig. 3 that both sidebands have identical connected part amplitudes.

IV. SLOW-MODULATION REGIME

In a number of physical systems, the modulation period is significantly smaller than the timescale of evolution of the localized system [40,41]. Such systems are considered to be in the slow-modulation regime and have been the subject of significant theoretical interest. In this section, we consider the limiting case of a slowly modulated localized system and study the form of the scattering matrices as well as their geometric properties.

A. Limiting form of the scattering matrices

For incident wave packets that interact with the localized systems for time durations much smaller than the modulation period, it is well known that the scattering matrices for the time-dependent system reduces to the scattering matrix of a time-independent localized system with the Hamiltonian being equal to the time-dependent Hamiltonian at the time of arrival of the incident wave packet [42]. In this section, we show that the general expressions for the single- and two-photon scattering matrices provided in the previous sections reduce to the scattering matrices for a time-independent system in the limit of slow modulation.

For the purpose of analyzing the slowly modulated limit, it is convenient to work with the scattering matrix element \( S(\omega_1, \omega_2, \ldots, \omega_N; \nu_1, \nu_2, \ldots, \nu_N) \) in which the incident state is described in the time domain and the outgoing state is described in the frequency domain. This is related to the frequency-domain scattering matrix \( S(\omega_1, \omega_2, \ldots, \omega_N; \nu_1, \nu_2, \ldots, \nu_N) \) via a Fourier transform over the input frequencies \( \nu_1, \nu_2, \ldots, \nu_N \),

\[
S(\omega_1, \omega_2, \ldots, \omega_N; \nu_1, \nu_2, \ldots, \nu_N) = \int \cdots \int S(\omega_1, \omega_2, \ldots, \omega_N; \nu_1, \nu_2, \ldots, \nu_N) \times \prod_{n=1}^{N} e^{i\nu_n t_n} \frac{d\nu_n}{\sqrt{2\pi}}.
\] (23)

where \( t_0 \) is the time reference in the definition of the scattering matrix introduced in Eq. (3). This matrix element can be related to the \( N \)-excitation Green’s function using Eq. (5),

\[
S(\omega_1, \omega_2, \ldots, \omega_N; \nu_1, \nu_2, \ldots, \nu_N) = (-1)^N e^{-i\phi(t)} \int \cdots \int G(t_1, t_2, \ldots, t_N; \nu_1, \nu_2, \ldots, \nu_N) \times \prod_{n=1}^{N} e^{i\nu_n t_n} \frac{d\nu_n}{\sqrt{2\pi}},
\] (24)

where \( \phi(t) = \sum_{n=1}^{N} \omega_n t_n \). As was pointed out in Sec. II, the \( N \)-excitation Green’s function can be determined completely by the non-Hermitian effective Hamiltonian. In the slow-modulation regime, the propagator corresponding to this effective Hamiltonian can be approximated to zeroth order in the modulation frequency by the instantaneous biorthogonal eigenstates of the time-dependent effective Hamiltonian. More concretely, the propagator \( U_{\text{df}}(t, s) \) within the \( n \)-excitation subspace can be expressed as

\[
U_{\text{df}}^n(t, s) = \sum_k (\Phi_k(t)) (\Phi_k(s) e^{-i \int_s^t \mathcal{A}_k^Z(s') ds'} + O(\Omega),
\] (25)

FIG. 3. Two-photon scattering from the cavity with Kerr nonlinearity: the connected part of the two-photon scattering matrix for the \( k = -1, 0, 1 \) sidebands with the two input photons at \( \nu_1 = \nu_2 = 0 \) as a function of the frequency difference \( \delta \) between the output photons. Note that the two output photons scattered into the \( k \)th sideband are constrained and have a mean frequency of \( k \Omega/2 \). Parameter values of \( \Omega = 2.5\kappa \) and \( \Delta_0 = 3\kappa \) have been assumed in all simulations.
where $|\Phi_n^p(t)\rangle$ and $|X_n^p(t)\rangle$ are the biorthogonal eigenstates of $H_{\text{eff}}^p(t)$ and $\Lambda_k^p(t)$ is the corresponding complex eigenvalue:

$$H_{\text{eff}}^p(t) |\Phi_n^p(t)\rangle = \Lambda_k^p(t) |\Phi_n^p(t)\rangle,$$

$$\left[H_{\text{eff}}^p(t)\right] |X_n^p(t)\rangle = \left[\Lambda_k^p(t)\right] |X_n^p(t)\rangle.$$  

(26)

Furthermore, since $H_{\text{eff}}^p(t)$ is non-Hermitian, $\Lambda_k^p(t)$ is complex; it can thus be split into the instantaneous frequency $\mathcal{E}_n^p(t)$ and decay rate $K_n^p(t): \Lambda_k^p(t) = \mathcal{E}_n^p(t) - iK_n^p(t)/2$.

Using Eqs. (24) and (25), the single- and two-photon scattering matrices in the slow-modulation regime to the zeroth order in the modulation frequency can be explicitly evaluated. As is shown in Appendix D, the single-photon scattering matrix $S(\omega, s)$ is given by

$$S(\omega, s) = -\sum_k \frac{(gL|\Phi_1(s)\rangle|X_1(s)\rangle g^\dagger|g\rangle \delta(\omega - \omega_s)}{i[\epsilon_1(s) - \omega_s] + K_1(s)/2} + O(\Omega),$$

(27)

and the connected part of the two-photon scattering matrix $S^{C}(\omega_1, \omega_2; s_1, s_2) = S^{C,1}(\omega_1, \omega_2; s_1, s_2) + S^{C,2}(\omega_1, \omega_2; s_1, s_2)$ for $s_1 \geq s_2$ is given by

$$S^{C,1}(\omega_1, \omega_2; s_1, s_2) = \sum_p \frac{e^{i\omega_1 s_1} e^{-i\omega_2 s_2}}{2\pi} \times \sum_{m,n} \frac{(gL|\Phi_m^1(s_1)\rangle|X_m^1(s_1)\rangle g^\dagger|\Phi_n^1(s_1)\rangle L_1^\dagger|g\rangle}{i[\omega_1(s_1) - \epsilon_n^1(s_1)] - K_n^1(s_1)/2} + O(\Omega),$$

$$S^{C,2}(\omega_1, \omega_2; s_1, s_2) = -\frac{e^{i(\omega_1 + \omega_2 s_1 - s_2)}}{2\pi} \times \sum_p \sum_{m,n} \frac{(gL|\Phi_m^2(s_1)\rangle|X_m^2(s_1)\rangle L_2^\dagger|\Phi_n^1(s_1)\rangle L_1^\dagger|\Phi_n^1(s_1)\rangle L_2^\dagger|g\rangle}{i[\omega_1 + \omega_2 - \epsilon_n^1(s_1)] - K_n^1(s_1)/2} + O(\Omega),$$

(28)

where $P$ is a permutation of $[1, 2]$. From Eqs. (27) and (28) it immediately follows that if the incident photonic state is localized in arrival time, then it is scattered as per the time-independent scattering matrix corresponding to the instantaneous Hamiltonian at its arrival time. We point out that this is only true to the zeroth order in the modulation frequency; to the first order in the modulation frequency, the time variation in the Hamiltonian of the system results in an energy shift in the scattered state relative to the incident photon state which depends on the derivative of the instantaneous scattering matrix with respect to the arrival time [42]. Furthermore, the expressions for the scattering matrices in Eqs. (27) and (28) reduce to those of time-independent scattering matrices [29] when the system Hamiltonian is time independent.

B. Geometric properties in the slow-modulation regime

For slowly modulated quantum systems, several experimentally observable properties depend only on the geometry of the modulation applied on the Hamiltonian and are independent of the modulation’s specific time dependence. For closed quantum systems, observables such as the Berry phase [43] can be defined which only depend on the geometry of the Hamiltonian being modulated and are independent of the modulation being applied to the Hamiltonian. For single-particle fermionic scattering problems, it has been shown that scattered currents from time-modulated scatterers are geometric in nature [44,45]. Here we consider the geometric properties of scattering from a slowly modulated quantum-optical system. In particular, it is shown that the equal-time $N$-photon correlation function, to the zeroth order in the modulation frequency, is equal to the time average of the instantaneous correlation function obtained by assuming the system is time independent. Furthermore, we show that the first-order correction to the $N$-photon correlation function is purely geometric in nature, i.e., it is independent of the precise form of the modulation applied on the Hamiltonian. We point out that the results derived in this section are distinct from those known for fermionic scattering since they hold for multiparticle scattering scenarios (i.e., $N > 1$) and account for the particle-particle interactions that are induced by the nonlinear nature of the localized system.

We consider a localized system with the Hamiltonian defined in terms of the $\mathcal{C}$, $\mathcal{N}$-excitation Green’s function, to the zeroth order in the modulation frequency, is equal to the time average of the instantaneous correlation function obtained by assuming the system is time independent. Furthermore, we show that the first-order correction to the $N$-photon correlation function is purely geometric in nature, i.e., it is independent of the precise form of the modulation applied on the Hamiltonian. We point out that the results derived in this section are distinct from those known for fermionic scattering since they hold for multiparticle scattering scenarios (i.e., $N > 1$) and account for the particle-particle interactions that are induced by the nonlinear nature of the localized system.

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For \( N = 1 \), from Eq. (13) this correlation function is identical to the total transmission \( \sum_{\kappa=0}^{\infty} \left| S_\kappa(v) \right|^2 \) through the localized system. For \( N \geq 2 \), this correlation function can be measured with \( N \)-photon coincidence counts on the emission from the localized system.

We now consider the calculation of a perturbative expansion for \( G_N(v) \) with respect to \( \Omega \). As is shown in Appendix E, it follows from the definition of the \( N \)-excitation Green’s function that

\[
\frac{1}{N!} \int_{-\infty}^{\infty} \ldots \int_{-\infty}^{\infty} G(t, t, \ldots; t; s_1, s_2, \ldots, s_N) \prod_{i=1}^{N} e^{-i\nu \tau} ds_i
\]

\[
= e^{-i\nu \tau} \left[ G_N^{(0)}(p; v) + G_N^{(1)}(p; v) \frac{dp(t)}{dt} + O(\Omega^2) \right].
\]

(31a)

where \( G_N^{(0)}(p; v) \) is zeroth order in the modulation frequency \( \Omega \) and is given by

\[
G_N^{(0)}(p; v) = (-i)^N \langle g | L^N \left[ \prod_{n=0}^{N} \left[ H_{\text{eff}}(p) - n\nu \right]^{-1} L \right] | g \rangle.
\]

(31b)

and \( G_N^{(1)}(p; v) \), also zeroth order in \( \Omega \), is given by

\[
G_N^{(1)}(p; v) = (-i)^{N-1} \sum_{k=1}^{N} \langle g | L^N \left[ \prod_{n=0}^{N} \left[ H_{\text{eff}}(p) - n\nu \right]^{-1} L \right] \langle k \rangle.
\]

(31c)

Here \( H_{\text{eff}}(p) \) is the \( n \)-excitation effective Hamiltonian as a function of the parameters \( p \). The equal-time \( N \)-photon correlation function can now be expanded into a perturbative series in \( \Omega \): \( G_N(v) = G_N^{(0)}(v) + \Omega G_N^{(1)}(v) + O(\Omega^2) \) where both \( G_N^{(0)}(v) \) and \( G_N^{(1)}(v) \) are zeroth order in \( \Omega \). It follows from Eqs. (30) and (31) that the zeroth-order contribution \( G_N^{(0)}(v) \) is given by

\[
G_N^{(0)}(v) = \int_0^T \left| G_N^{(0)}(p(t); v) \right|^2 dt / T.
\]

(32)

It should be noted that \( |G_N^{(0)}(p; v)|^2 \) is the equal-time \( N \)-photon correlation function that would be measured from the emission of a time-independent localized system with Hamiltonian \( H(p) \) and consequently to zeroth order \( G_N(v) \) is simply a time average of the instantaneous correlation function \( |G_N^{(0)}(p; v)|^2 \). Furthermore, \( G_N^{(0)}(v) \) is dynamical in nature, i.e., it is dependent on the precise modulation of the
on the loop C trace during modulation. A.W. acknowledges support from Herb and Jane Dwight in the parameter space that the parameters p trace during modulation.

As an illustrative example, we consider scattering from a Jaynes-Cummings system formed by coupling a cavity with resonant frequency \( \omega_c \) to a two-level system (TLS) at frequency \( \omega_o \),

\[
H_o(g) = \omega_o \sigma^+ \sigma + \omega_o a^+ a + (g a \sigma^+ + g^* a \sigma),
\]

where we modulate the complex cavity-TLS coupling strength \( g \) periodically as a function of time to obtain a time-dependent Hamiltonian. We assume that this system coupled to the bosonic bath through the cavity mode, i.e., \( L = \sqrt{\kappa/2a} \), where \( \kappa \) is the decay rate of the cavity. We consider three different modulations of \( g \) as depicted in Fig. 4(a), which traverse the same loop in the complex plane per period. The shaded regions in Fig. 4(a) indicate the rate of change of \( g \) with time at different points on the loop. Figure 4(b) shows the zeroth- and first-order contributions to the transmission spectrum \( T(v) = G_1(v) \) for the three different choices of \( g(t) \). We can see clearly that the zeroth-order contribution \( T^{(0)}(v) \) is dependent on the time dependence of the modulation applied on \( g \) whereas the first-order contribution \( T^{(1)}(v) \) is identical for the three different modulation schemes, i.e., it is completely geometric in nature. A similar behavior can be seen for the zeroth- and first-order contributions to the equal-time two-photon correlation.

V. CONCLUSION

In this paper, we have studied scattering of photons from periodically modulated quantum systems. A procedure for constructing \( N \)-photon scattering matrices and relating them to the Floquet decomposition of the effective Hamiltonian of the quantum system was outlined. Furthermore, we studied the properties of the equal-time \( N \)-photon correlation function in the slow-modulation regime and showed that the first-order correction with respect to the modulation frequency is completely geometric in nature. The formalism and results presented in this paper are of fundamental interest in the study of time-dependent open systems as well as for simulating quantum systems relevant for building quantum information processing systems.

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APPENDIX A: PHOTON-NUMBER CONSERVATION BY THE SCATTERING MATRIX

Let \( \Pi_n^s \) be the projector onto the \( n \)-th-excitation subspace \( \mathcal{H}_n^s \). The excitation-number operator \( \mu_s \) can be constructed from \( \Pi_n^s \) via

\[
\mu_s = \sum_{n=0}^{\infty} n \Pi_n^s.
\]

By construction, \( \mu_s = \mu_s^+ \) and \( \mu_s|\phi\rangle = n|\phi\rangle \) for \( |\phi\rangle \in \mathcal{H}_n^s \). Additionally,

\[
[L, \mu_s] = L.
\]

To see this, suppose \( |\phi\rangle \in \mathcal{H}_n^s \) for any \( n \geq 1 \); then \( L|\phi\rangle \in \mathcal{H}_n^{s-1} \). Therefore,

\[
\mu_s|\phi\rangle = n L|\phi\rangle, \quad \mu_s L|\phi\rangle = (n - 1)L|\phi\rangle \Rightarrow [L, \mu_s]|\phi\rangle = L|\phi\rangle.
\]

Furthermore, for \( |\phi\rangle \in \mathcal{H}_n^s \), since \( \mu_s|\phi\rangle = 0 \) and \( L|\phi\rangle = 0 \) it follows that \( [L, \mu_s]|\phi\rangle = 0 = L|\phi\rangle \). This shows that the operators \( L \) and \( \mu_s \) satisfy Eq. (A2).

Finally, consider the excitation-number operator \( \mu \) for the full system constructed by adding \( \mu_s \) with the photon-number operator for the two baths,

\[
\mu = \mu_s + \sum_{l \in \{a, b\}} \int_{-\infty}^{\infty} \tilde{L}_l d\omega.
\]

From the commutator \( [L, \mu_s] = L \) it follows that \( [H(t), \mu] = 0 \), i.e., the observable corresponding to \( \mu \) is a conserved quantity. Since a state with \( N \) photons in the bosonic baths and the system in \( |g\rangle \) is an eigenstate of \( \mu \) with eigenvalue \( N \), this conservation law immediately implies that it can only scatter into a state with \( N \) photons in the bosonic baths.

APPENDIX B: RELATING THE SCATTERING MATRIX ELEMENTS TO THE GREEN’S FUNCTION

In this Appendix, we derive the relationship between the Green’s function \( G(t_1, t_2, \ldots, t_N; s_1, s_2, \ldots, s_N) \) and the scattering matrix element \( S(\omega_1, \omega_2, \ldots, \omega_N; v_1, v_2, \ldots, v_N) \) [Eq. (5)]. Using the fact that \( H_o(t)|g\rangle = 0 \) and \( L|g\rangle = 0 \), it follows from Eq. (1) that \( H(t)|g; \text{vac}\rangle = 0 \). Noting that the propagator \( U_0(t_f, t_i)|g\rangle \) with respect to the Hamiltonian \( H_o(t) \) [Eq. (4)] satisfies \( U_0(t_f, t_i)|g\rangle = U_0(t_f, t_i) = e^{-it_o(t_f-t_i)} \forall l \in \{a, b\} \), the scattering matrix element \( S(\omega_1, \omega_2, \ldots, \omega_N; v_1, v_2, \ldots, v_N) \) can be expressed as

\[
S(\omega_1, \omega_2, \ldots, \omega_N; v_1, v_2, \ldots, v_N) = e^{-i\phi(t_o)} \lim_{t_o \to +0} \frac{\exp \left( \sum_{l=1}^{N} (\omega_l t_l - v_l t_l) \right)}{T} |g; \text{vac}\rangle \times \left[ \prod_{l=1}^{N} b_{\omega_l}(t_l) \right] \left( \prod_{l=1}^{N} a_{v_l}(t_l) \right) |g; \text{vac}\rangle.
\]
where \( a_i(t) = U(t_0, t_i) a U(t_i, t_0) \) and \( b_i(t) = U(t_0, t_i) b U(t_i, t_0) \), with \( U(\cdot, \cdot) \) the propagator with respect to the Hamiltonian \( H(t) \) [Eq. (1)], and \( \mathcal{T}[\cdot] \) indicates a time ordering with respect to its arguments. Note that since \( t_i \geq t_j \), this time ordering is effectively an identity operation in Eq. (B1). Next we use the Heisenberg equations of motion for \( a_i(t) \) and \( b_i(t) \); from Eq. (1) it follows that

\[
\frac{d}{dt} (a_i(t) \langle b_i(t) \rangle) = -i \left( [a_i(t), \omega b_i(t)] + \frac{1}{\sqrt{2\pi}} \left( L(t) - \tilde{L}(t) \right) \right),
\]

(B2)

These equations of motion can easily be integrated from \( t_i \) to \( t_i \) to yield the following:

\[
a_i(t_i) = a_i(t_i) e^{-i\tilde{L}(t_i-t_i)} - \int_{t_i}^{t_i} L(s) e^{-i\tilde{L}(t_i-s)} \frac{ds}{\sqrt{2\pi}}, \quad (B3a)
\]

\[
b_i(t_i) = b_i(t_i) e^{-i\tilde{L}(t_i-t_i)} + \int_{t_i}^{t_i} L(t) e^{-i\tilde{L}(t_i-t)} \frac{dt}{\sqrt{2\pi}}, \quad (B3b)
\]

Substituting Eq. (B3b) into Eq. (B1) and noting that any term with \( b_i(t) \) goes to 0 since the time-ordering operator places it to the right of \( L(t) \) for \( t \in (t_i, t_i) \) and \( b_i(t) \) commutes with \( a_i(t) \) and annihilates \( \langle g; \text{vac} \rangle \), we obtain

\[
S(\omega_1, \omega_2, \ldots, \omega_N, v_1, v_2, \ldots, v_N) = e^{-i\phi(t_0)} \lim_{t_i \to -\infty} \int_{t_i}^{t_i} \cdots \int_{t_i}^{t_i} \langle g; \text{vac} \rangle [\prod_{i=1}^{N} L(t_i)]
\times \left( \prod_{i=1}^{N} a_i(t_i) \right) \langle g; \text{vac} \rangle \prod_{i=1}^{N} e^{i\omega_i t_i} \frac{dt_i}{\sqrt{2\pi}}.
\]

(B4)

Similarly, substituting Eq. (B3a) into Eq. (B4) and noting that any term with \( a_i(t) \) goes to 0 since the time-ordering operator places it to the left of \( L(t) \) for \( t \in (t_i, t_i) \), and \( a_i(t) \) annihilates \( \langle g; \text{vac} \rangle \), we obtain the result in Eq. (5).

**APPENDIX C: SCATTERING MATRIX CALCULATION**

1. **Single-photon scattering matrix**

   The starting point for the calculation of the single-photon scattering matrix is the evaluation of the single-photon Green’s function, which is given by

\[
G(t; s) = \langle g|\mathcal{T}[\tilde{L}(t)\tilde{L}^{\dagger}(s)]|g\rangle = \langle g|LU_{\text{eff}}(t, s)L^\dagger|g\rangle \Theta(t \geq s).
\]

(C1)

Using the Floquet decomposition of \( U_{\text{eff}}(t, s) \) [Eq. (12)], this can be expressed as

\[
G(t; s) = (\mathbf{L}^{1-0}(t)D[e^{-i\tilde{L}(t-s)}]\mathbf{L}^{0-1}(s))\Theta(t \geq s),
\]

(C2)

where \( \mathbf{L}^{1-0}(t) \) is a row vector, \( \mathbf{L}^{0-1}(s) \) is a column vector, and their elements are given by

\[
[\mathbf{L}^{1-0}(t)]_n = \langle g|L|b_n(t)\rangle, \quad [\mathbf{L}^{0-1}(s)] = \langle \chi_n^s(s)|L^\dagger|g\rangle.
\]

(C3)

We note that \( \mathbf{L}^{1-0}_k \) and \( \mathbf{L}^{0-1}_k \) defined in Eq. (14) are simply the Fourier-series coefficients of \( \mathbf{L}^{1-0}(t) \) and \( \mathbf{L}^{0-1}(s) \), respectively:

\[
\mathbf{L}^{1-0}(t) = \sum_{k \in \mathbb{Z}} \mathbf{L}^{1-0}_k e^{-i\Omega_k t}, \quad \mathbf{L}^{0-1}(s) = \sum_{k \in \mathbb{Z}} \mathbf{L}^{0-1}_k e^{i\Omega_k s}.
\]

(C4)

From Eqs. (5), (C1), and (C4) it follows that the single-photon scattering matrix \( S(\omega; \nu) \) is given by Eq. (13).

2. **Two-photon scattering matrix**

   The two-excitation Green’s function \( G(t_1, t_2; s_1, s_2) \), given by Eq. (7) with \( N = 2 \), is symmetric under the Swap operations \( t_1 \leftrightarrow t_2 \) and \( s_1 \leftrightarrow s_2 \). Defining \( \mathcal{G}(t_1, t_2; s_1, s_2) = G(t_1, t_2; s_1, s_2)\Theta(t_1 \geq t_2; s_1 \geq s_2) \), it follows that

\[
\mathcal{G}(t_1, t_2; s_1, s_2) = \sum_{P,Q} \mathcal{G}(t_{P(1)}, t_{P(2)}; s_{Q(1)}, s_{Q(2)}),
\]

(C5)

where \( P \) and \( Q \) are permutations of the two-element set \( \{1, 2\} \). It thus follows from Eq. (5) that

\[
S(\omega_1, \omega_2; v_1, v_2) = \sum_{P,Q} e^{-i\phi(t_0)} S(\omega_{P(1)}, \omega_{P(2)}; v_{Q(1)}, v_{Q(2)}),
\]

(C6)

where

\[
S(\omega_1, \omega_2; v_1, v_2) = \int_{-\infty}^{\infty} \cdots \int_{-\infty}^{\infty} \mathcal{G}(t_1, t_2; s_1, s_2) \prod_{i=1}^{2} e^{i(\omega_{0i} - v_i s_i)} ds_i \frac{dt_1 ds_1}{2\pi}.
\]

(C7)

From Eq. (7) it follows that

\[
\mathcal{G}(t_1, t_2; s_1, s_2) = \mathcal{G}^1(t_1, t_2; s_1, s_2) + \mathcal{G}^2(t_1, t_2; s_1, s_2),
\]

(C8)

where

\[
\mathcal{G}^1(t_1, t_2; s_1, s_2) = \langle g|L(t_1)L^\dagger(s_1)L(t_2)L^\dagger(s_2)|g\rangle \Theta(t_1 \geq s_1 \geq t_2 \geq s_2)
\]

\[
= \langle g|LU_{\text{eff}}^1(t_1, s_1)L^\dagger|g\rangle \langle g|LU_{\text{eff}}^1(t_2, s_2)L^\dagger|g\rangle \Theta(t_1 \geq s_1 \geq t_2 \geq s_2),
\]

(C9a)
\[ G^2(t_1, t_2; s_1, s_2) = (g|L(t_1)L(t_2)L^\dagger(s_1)L^\dagger(s_2)|g)\Theta(t_1 \geq t_2 \geq s_1 \geq s_2) \]  
\[ = (g|LU_{\text{eff}}^1(t_1, t_2)L^2 U_{\text{eff}}^1(t_2, s_1)L_{\text{eff}}^1(s_1, s_2)L^\dagger|g)\Theta(t_1 \geq t_2 \geq s_1 \geq s_2). \]  
\[ \text{(C9b)} \]
\[ \text{(C9c)} \]

Using the Floquet decomposition of \( U_{\text{eff}}^1(t, s) \) [Eq. (12)], it follows that
\[ G^1(t_1, t_2; s_1, s_2) = [L^{1-0}(t_1)D[e^{-\bar{i}(t_1-s_1)}]L^{0-1}(s_1)] [L^{1-0}(t_2)D[e^{-\bar{i}(t_2-t_1)}]L^{0-1}(s_2)] \Theta(t_1 \geq s_1 \geq t_2 \geq s_2), \]  
\[ G^2(t_1, t_2; s_1, s_2) = [L^{1-0}(t_1)D[e^{-\bar{i}(t_1-t_2)}]L^{2-1}(t_2)] [L^{1-0}(s_1)D[e^{-\bar{i}(s_1-t_2)}]L^{1-2}(s_2)] \Theta(t_1 \geq t_2 \geq s_1 \geq s_2), \]  
\[ \text{(C10a)} \]
\[ \text{(C10b)} \]

where \( L^{1-0}(t) \) and \( L^{0-1}(s) \) are defined in Eq. (C3) and \( L^{2-1}(t) \) and \( L^{1-2}(s) \) are matrices with elements
\[ [L^{2-1}(t)]_{m,n} = \langle \chi^1_m(t)|\phi^2_n(t) \rangle, \quad [L^{1-2}(s)]_{m,n} = \langle \chi^2_m(s)|\phi^1_n(s) \rangle. \]  
\[ \text{(C10c)} \]

It should be noted that \( L_{k}^{2-1} \) and \( L_{k}^{1-2} \), defined in Eq. (16), are simply the Fourier-series coefficients of \( L^{2-1}(t) \) and \( L^{1-2}(s) \):
\[ L^{2-1}(t) = \sum_{k \in \mathbb{Z}} L_{k}^{2-1} e^{-i\Omega t}, \quad L^{1-2}(s) = \sum_{k \in \mathbb{Z}} L_{k}^{1-2} e^{i\Omega t}. \]  
\[ \text{(C11)} \]

Using Eq. (C10b) to evaluate the integral in Eq. (C7), we obtain
\[ S(\omega_1, \omega_2; v_1, v_2) = \sum_{k \in \mathbb{Z}} S_k^1(\omega_1, \omega_2; v_1, v_2) \delta(\omega_1 + \omega_2 - v_1 - v_2 - k\Omega), \]  
\[ \text{(C12a)} \]

where
\[ S_k^1(\omega_1, \omega_2; v_1, v_2) = \frac{1}{2\pi i} \sum_{p,m,n \in \mathbb{Z}} \left[ L_{p}^{1-0}D(1) \left( \frac{1}{i(\lambda^1 - \omega_1 - p\Omega)} \right) \right] \]
\[ \times \left[ L_{m}^{0-1}D(1) \left( \frac{1}{i(\lambda^1 - \omega_2 - n\Omega)} \right) \right], \]  
\[ \text{(C12b)} \]
\[ S_k^2(\omega_1, \omega_2; v_1, v_2) = \frac{1}{2\pi} \sum_{p,m,n \in \mathbb{Z}} \left[ L_{p}^{1-0}D(1) \left( \frac{1}{i(\lambda^1 - \omega_1 + p\Omega)} \right) \right] \]
\[ \times \left[ L_{m}^{0-1}D(1) \left( \frac{1}{i(\lambda^1 - \omega_2 + m\Omega)} \right) \right], \]  
\[ \text{(C12c)} \]

The full two-photon scattering matrix can be constructed from Eqs. (C6) and (C12). To explicitly extract the connected part of the two-photon scattering matrix, we note that \( 1/(\kappa - i0^+) = \Re(1/\kappa) + \pi \delta(\kappa) \); the connected part of the scattering matrix can thus be obtained by replacing \( 1/(\kappa - i0^+) \) by \( \Re(1/\kappa) \) in the resulting expressions for the scattering matrix. This yields the results in Eq. (15).

Finally, we show that \( S_{k}^{C,1}(\omega_1, \omega_2; v_1, v_2) \) defined in Eq. (15) is not singular despite containing the principal parts. We begin by rewriting it as
\[ S_{k}^{C,1}(\omega_1, \omega_2; v_1, v_2) = \sum_{P,Q,n \in \mathbb{Z}} S_{k-n}[\omega_P(1) - (k - n)\Omega]S_n(v_Q(2))P \frac{1}{\omega_P(2) - v_Q(2) - n\Omega} \]  
\[ = \sum_{n \in \mathbb{Z}} \sum_{l=1}^{2} \left[ S_{k-n}[\bar{\omega}_l - (k - n)\Omega]S_n(\bar{v}_l)P \frac{1}{\bar{\omega}_l - \bar{v}_l - n\Omega} + S_{k-n}[^2\bar{\omega}_l - (k - n)\Omega]S_n(^2\bar{v}_l)P \frac{1}{^2\bar{\omega}_l - ^2\bar{v}_l - n\Omega} \right] \]  
\[ = \sum_{n \in \mathbb{Z}} \sum_{l=1}^{2} \left[ S_n(\omega_l - n\Omega)S_{k-n}(\bar{v}_l)P \frac{1}{\omega_l - \bar{v}_l - (k - n)\Omega} + S_{k-n}[\bar{\omega}_l - (k - n)\Omega]S_n(\bar{v}_l)P \frac{1}{\bar{\omega}_l - \bar{v}_l - n\Omega} \right], \]  
\[ \text{(C13)} \]

where \( \bar{\lambda}_{1,2} = \lambda_{2,1} \) for \( \lambda \in \{\omega, v\} \) and \( S_k(v) \) is the scattering amplitude of a single photon at frequency \( v \) into frequency \( v + k\Omega \) defined in Eq. (13). We note that since the arguments of \( S_{k}^{C,1} \) are constrained to satisfy \( \omega_1 + \omega_2 - v_1 - v_2 = k\Omega \), it follows that
\[ S_{k}^{C,1}(\omega_1, \omega_2; v_1, v_2) = \sum_{n \in \mathbb{Z}} \sum_{l=1}^{2} \left[ S_n(\bar{v}_l - (v_l - \omega_l + n\Omega))S_{k-n}[^2\bar{v}_l - (v_l - \omega_l + n\Omega)]S_n(\bar{v}_l)P \frac{1}{v_l - \omega_l + n\Omega} \right]. \]  
\[ \text{(C14)} \]
We can then evaluate the Fourier transform of \( M_{n,m}(v,\bar{v},\delta) \), defined below, is a smooth and finite function of its arguments

\[
M_{n,m}(v,\bar{v},\delta) = \sum_{i,j} \sum_{p,q\in\mathbb{Z}} \left[ \frac{[L^0_{p\rightarrow q}][L^0_{p+q \rightarrow q}][L^0_{q\rightarrow p}]}{\lambda_i^2 + (p+q)\Omega - \bar{v} - v} \right] \cdot \frac{1}{\lambda_j^2 + p\Omega - (v - \delta) \left[ (\lambda_j^2 + q\Omega - \bar{v}) \left[ (\lambda_j^2 + p\Omega - iv) \left[ (\lambda_j^2 + q\Omega - (v + \delta)) \right] \right] \right]}
\]

it follows that

\[
S_{k}^{(1)}(\omega_1, \omega_2; v_1, v_2) = \sum_{n\in\mathbb{Z}} \sum_{i=1}^{2} M_{n,k-n}(v,\bar{v},v_i - \omega_{i} + n\Omega).
\]

This shows that \( S_{k}^{(1)}(\omega_1, \omega_2; v_1, v_2) \) is indeed a well-defined and finite function of the input and output frequencies subject to the constraint \( \omega_1 + \omega_2 - v_1 - v_2 = k\Omega \).

**APPENDIX D: SINGLE- AND TWO-PHOTON SCATTERING MATRIX IN THE SLOW MODULATION REGIME**

1. Single-photon scattering matrix

With the decomposition of the effective propagator in Eq. (25), the single-excitation Green’s function can be expressed as

\[
G(t,s) = \sum_{k} \langle g| \Phi_{k}(t) \rangle \langle X(s)L^\dagger_{k} | g \rangle \exp \left( -i \int_{s}^{t} \Lambda_{k}^{1}(\tau)d\tau \right) \Theta(t - s).
\]

We can then evaluate the Fourier transform of \( G(t,s) \) with respect to \( t \),

\[
G(\omega;s) = \int_{-\infty}^{\infty} G(t,s) \exp(i\omega t) dt = \sum_{k} \left\{ \int_{-\infty}^{\infty} \langle g| \Phi_{k}(t) \rangle \exp \left[ i(\omega t - \int_{s}^{t} \Lambda_{k}^{1}(\tau)d\tau) \right] \right\} \langle X(s)L^\dagger_{k} | g \rangle
\]

\[
= \sum_{k} \left\{ \int_{-\infty}^{\infty} \frac{\partial}{\partial(\omega - \Lambda_{k}^{1}(\tau))} \exp \left[ i(\omega t - \int_{s}^{t} \Lambda_{k}^{1}(\tau)d\tau) \right] \right\} \langle X(s)L^\dagger_{k} | g \rangle
\]

\[
= \sum_{k} \frac{i\langle g| \Phi_{k}(s) \rangle \langle X(s)L^\dagger_{k} | g \rangle}{\omega - \Lambda_{k}^{1}(s)} \exp(i\omega t) + O(\Omega),
\]

where in the last step we have used integration by parts and only retained terms that are zeroth order in the modulation frequency. Substituting this into Eq. (24), we obtain Eq. (27).

2. Two-photon scattering matrix

We consider computing the Fourier transform of the two-excitation Green’s function \( G(t_1, t_2; s_1, s_2) \) with respect to \( t_1 \) and \( t_2 \). Noting that since \( G(t_1, t_2; s_1, s_2) \) is symmetric under the SWAP operations \( t_1 \leftrightarrow t_2 \) and \( s_1 \leftrightarrow s_2 \), we can restrict ourselves to \( s_1 \geq s_2 \) and

\[
\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} G(t_1, t_2; s_1, s_2) \exp(i(\omega_{t_1} + \omega_{t_2})t_1) dt_1 dt_2 = \sum_{p} \int_{-\infty}^{\infty} \int_{t_2}^{\infty} G(t_1, t_2; s_1, s_2) \exp(i(\omega_{t_1} + \omega_{t_2})t_1) dt_1 dt_2.
\]

Recalling that \( G(t_1, t_2; s_1, s_2) = \langle g| T[L(t_1)L(t_2)L^\dagger(s_1)L^\dagger(s_2)] | g \rangle \), we obtain

\[
\int_{-\infty}^{\infty} \int_{t_2}^{\infty} G(t_1, t_2; s_1, s_2) \exp(i(\omega_{t_1} + \omega_{t_2})t_1) dt_1 dt_2
\]

\[
= \int_{s_1}^{\infty} \int_{s_2}^{\infty} \langle g| LU_{\text{eff}}(t_1, t_2)LU_{\text{eff}}^\dagger(t_2, s_1)L^\dagger s_1 L^\dagger(s_2) | g \rangle \exp(i(\omega_{t_1} + \omega_{t_2})t_1) dt_1 dt_2
\]

\[
+ \int_{s_1}^{\infty} \int_{s_2}^{\infty} \langle g| LU_{\text{eff}}(t_1, s_1)L^\dagger | g \rangle \langle g| LU_{\text{eff}}(t_2, s_2)L^\dagger | g \rangle \exp(i(\omega_{t_1} + \omega_{t_2})t_1) dt_1 dt_2.
\]
The two integrals in Eq. (D4) can be evaluated individually to the zeroth order in the modulation frequency using the decomposition of the effective propagator from Eq. (25). Consider the first term

\[
\sum_{m,n} \int_{s_1}^{s_2} \frac{i(gL)[Phi_m(t_1)]|X_m^{(1)}(s)|L^1 U^1_{eff}(s_1, s_2)L^1|g|}{\omega_{m}(t_1) - \Lambda^2_{m}(t_2)} \exp \left[ i \left( \omega_{m}(t_1) - \int_{s_1}^{t_1} \Lambda^1_{m}(t)dt \right) \right] e^{i\omega_{m}t_2} dt_1 dt_2
\]

Proceeding with the second term, we obtain

\[
\sum_{m,n} \int_{s_1}^{s_2} \frac{i(gL)[Phi_m(t_2)]|X_m^{(2)}(t)|L^1 U^1_{eff}(s_1, s_2)L^1|g|}{\omega_{m}(t_2) - \Lambda^2_{m}(t_2)} \exp \left[ i \left( \omega_{m}(t_2) - \int_{s_1}^{t_2} \Lambda^1_{m}(t)dt \right) \right] e^{i\omega_{m}t_1} dt_1 dt_2
\]

(D5)

Throughout this calculation, we have only retained terms which are zeroth order in the modulation frequency. Similarly proceeding with the second term, we obtain

\[
\sum_{m,n} \int_{s_1}^{s_2} \frac{i(gL)[Phi_m(t_1)]|X_m^{(1)}(s)|L^1 U^1_{eff}(s_1, s_2)L^1|g|}{\omega_{m}(t_1) - \Lambda^2_{m}(t_1)} \exp \left[ i \left( \omega_{m}(t_1) - \int_{s_1}^{t_1} \Lambda^1_{m}(t)dt \right) \right] e^{i\omega_{m}t_2} dt_1 dt_2
\]

Using Eqs. (D6) and (D5) along with Eq. (24), we obtain

\[
S(\omega_1, \omega_2; s_2) = S(\omega_1, s_1)S(\omega_2, s_2) + S(\omega_1; s_2)S(\omega_2; s_1) + S^C(\omega_1, \omega_2; s_1, s_2) \tag{D7}
\]

where \(S^C(\omega_1, \omega_2; s_1, s_2) = S^{C,-1}(\omega_1, \omega_2; s_1, s_2) + S^{C,-2}(\omega_1, \omega_2; s_1, s_2)\), with \(S^{C,-1}(\omega_1, \omega_2; s_1, s_2)\) and \(S^{C,-2}(\omega_1, \omega_2; s_1, s_2)\) given by Eq. (28).
APPENDIX E: EQUAL-TIME N-PHOTON CORRELATION FUNCTION IN THE SLOW MODULATION REGIME

In this Appendix, we provide a derivation of Eq. (31). Noting that the $N$-excitation Green’s function $G(t_1, t_2, \ldots, N; s_1, s_2, \ldots, s_N)$ is symmetric under permutation of the times $s_1, s_2, \ldots, s_N$, it follows that

$$\frac{1}{N!} \int_{-\infty}^{\infty} \cdots \int_{-\infty}^{\infty} G(t_1, \ldots, t; s_1, s_2, \ldots, s_N) \prod_{i=1}^{N} e^{-i\nu s_i} ds_i = \int_{-\infty}^{\infty} \cdots \int_{-\infty}^{\infty} G(t_1, \ldots, t; s_1, s_2, \ldots, s_N) \prod_{i=1}^{N} e^{-i\nu s_i} ds_i. \quad (E1)$$

Furthermore, from Eq. (7) we obtain that if $s_1 \leq s_2 \cdots \leq s_N$ then

$$G(t, \ldots, t; s_1, s_2, \ldots, s_N) = \langle g | U_n^{n} (s_{n+1}, s_n) L^1 \rangle \langle g \rangle. \quad (E2)$$

To proceed further, we consider the evaluation of

$$\int_{-\infty}^{t} U_{\text{eff}}^n(t, s) O(s) e^{-is\nu} ds, \quad (E3)$$

where $O(s)$ is a time-dependent operator which is assumed to be slowly varying. Since $U_{\text{eff}}^n(t, s)$ is the propagator corresponding to the Hamiltonian $H_{\text{eff}}^n(t)$, it follows that

$$\int_{-\infty}^{t} U_{\text{eff}}^n(t, s) O(s) e^{-is\nu} ds = -i \int_{-\infty}^{t} \left[ \frac{\partial}{\partial s} U_{\text{eff}}^n(t, s) e^{-is\nu} \right] \left[ H_{\text{eff}}^n(s) - v \right]^{-1} O(s) ds. \quad (E4)$$

Applying integration by parts, we obtain

$$\int_{-\infty}^{t} U_{\text{eff}}^n(t, s) O(s) e^{-is\nu} ds = -i \left[ H_{\text{eff}}^n(t) - v \right]^{-1} O(t) e^{-it\nu} + i \int_{-\infty}^{t} U_{\text{eff}}^n(t, s) \frac{\partial}{\partial s} \left[ H_{\text{eff}}^n(s) - v \right]^{-1} O(s) \right] e^{-is\nu} ds. \quad (E5)$$

Repeating a similar calculation for the integral on the right-hand side of Eq. (E5) and neglecting terms that are second order in the derivatives of $H_{\text{eff}}^n(t)$ and $O(t)$, we obtain

$$\int_{-\infty}^{t} U_{\text{eff}}^n(t, s) O(s) e^{-is\nu} ds \approx -i \left[ H_{\text{eff}}^n(t) - v \right]^{-1} O(t) e^{-it\nu} + \left[ H_{\text{eff}}^n(t) - v \right]^{-1} \left[ \frac{\partial}{\partial t} \left[ H_{\text{eff}}^n(s) - v \right]^{-1} O(s) \right] e^{-it\nu}. \quad (E6)$$

Repeated application of this to the integral in Eq. (E1) together with neglecting any terms second order or higher in the derivatives of the effective Hamiltonian, we obtain the result in Eq. (31).

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