Analytic Properties of Two-Photon Scattering Matrix in Integrated Quantum Systems Determined by the Cluster Decomposition Principle

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We consider a general class of integrated quantum systems where photon-photon interaction occurs in a quantum device that is localized in space. Using techniques that are closely related to cluster decomposition principles in quantum field theory, we provide a general constraint on the analytic properties of a two-photon S matrix in this class of systems. We also show that the photon-photon interaction in these systems inevitably leads to frequency mixing and entanglement and that frequencies of the single photons cannot be preserved in these systems.

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Creating a strong photon-photon interaction at the few-photon level is of great importance for quantum information science. While strong photon-photon interactions were first realized in atomic systems [1], in recent years there have been significant experimental efforts in seeking to realize such interactions on chip in integrated systems [2–6]. In integrated systems, few-photon states can be routed via nanophotonic waveguides or microwave transmission lines to couple with a quantum system [Fig. 1(a)], which provide new possibilities for controlling photon-photon interactions [7].

The experimental efforts, in turn, have motivated significant efforts aiming to theoretically describe photon-photon interactions in integrated systems. The interaction between two photons is fundamentally characterized by the two-photon S matrix. A number of authors have calculated two-photon S matrices for waveguides coupling to a wide variety of quantum systems, including single or multiple atoms having either two or multiple levels [8–17], a cavity with Kerr nonlinearity [18], an optomechanical cavity [19], and a cavity with an atom inside [20–22]. These calculations have provided substantial insights into the nature of the photon-photon interaction in these specific geometries.

In this Letter, complementary to all the detailed calculations on specific geometries, we consider the general analytic structures of the two-photon S matrices in integrated systems. A common characteristic of all the systems considered in Refs. [8–22] is that the quantum system that induces the photon-photon interaction is localized in space. Here, using an argument related to the cluster decomposition principle [23–25] in quantum field theory, we show that such a spatially local nature of photon-photon interaction provides strong constraints on the structure of the two-photon S matrix. We also show that such general constraints can be used to guide the design of quantum information processing devices.

To describe the general system shown in Fig. 1(a), we consider the Hamiltonian ($\hbar = 1$)

$$H = \int dk c_k^\dagger c_k + H_{\rm int}[c_k, c_k^\dagger, a], \qquad (1)$$

where c_k ($c_k^\dagger$) is the annihilation (creation) operator of the photon state in the waveguide. These operators satisfy the

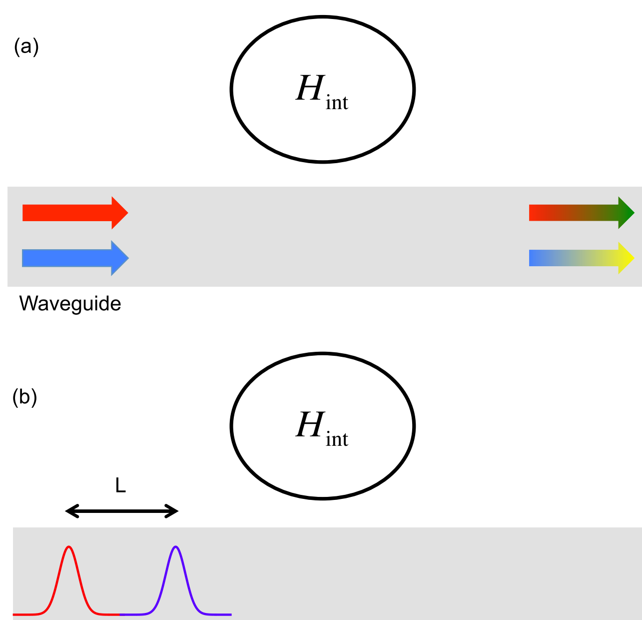

FIG. 1 (color online). A photonic waveguide coupled to a local region, as described by the Hamiltonian $H_{\rm int}$ where a photon-photon interaction occurs. (a) The first main result of this Letter: When a photon-photon interaction occurs, the frequency of the single photons, as represented by the colors here, cannot be preserved. (b) The schematic of the proof: The effect of interaction should vanish as the separation L of the two single-photon pulses goes to infinity.

standard commutation relation $[c_k, c_{k'}^\dagger] = \delta(k - k')$. The group velocity is set to be $v_g = 1$ so that the single photon's frequency is equal to its momentum. Here for simplicity we consider a waveguide consisting of only a single mode in the sense of Ref. [9]. The argument here, however, can be straightforwardly generalized to waveguides supporting multiple modes. $H_{\rm int}[c_k, c_k^\dagger, a]$ describes a *spatially localized* region, referred to here as a "device," that couples to the waveguide. The symbol a here generically represents the internal operators of the device. Since we are interested in nonlinear interactions, $H_{\rm int}$ in general is not in the bilinear form of boson operators. We consider the case where the number of photons is conserved, i.e., $[H, N] = 0$, where N is the total excitation operator. In the single excitation subspace, the scattering matrix of the system is in general of the form

$$S_{pk} = t_k \delta(p - k), \tag{2}$$

where t_k is the transmission coefficient and the δ function arises from the energy conservation. For two incident photons, with their momenta denoted by k_1 and k_2, respectively, if there is no photon-photon interaction, the two-photon scattering matrix can be straightforwardly written as

$$S^0_{p_1p_2k_1k_2} = t_{k_1} t_{k_2}[\delta(p_1 - k_1)\delta(p_2 - k_2) + \delta(p_1 - k_2)\delta(p_2 - k_1)]. \tag{3}$$

Notice in this noninteracting case that the energy of the individual photons are preserved after the scattering process. In the presence of a photon-photon interaction, the two-photon S matrix is modified as

$$S_{p_1p_2k_1k_2} = S^0_{p_1p_2k_1k_2} + iT_{p_1p_2k_1k_2}. \tag{4}$$

This equation can be considered as the definition of the T matrix.

The first main result of this Letter is as follows: We show that, for any device of the type as described by (1), the T matrix in general *must* have the form

$$T_{p_1p_2k_1k_2} = C_{p_1p_2k_1k_2}\delta(p_1 + p_2 - k_1 - k_2), \tag{5}$$

which contains only a single δ function as required by energy consideration. The T *matrix* cannot take the form

$$\frac{1}{2}C_{k_1k_2}[\delta(p_1 - k_1)\delta(p_2 - k_2) + \delta(p_1 - k_2)\delta(p_2 - k_1)]. \tag{6}$$

In other words, in the presence of a photon-photon interaction, after a scattering process, the energy of individual photons *cannot* be conserved. This is in spite of the fact that the S matrix corresponding to (6) satisfies the constraint of the energy conservation and can be made to satisfy all other symmetry constraints such as the time-reversal symmetry.

This result can be proved by using the procedure, as schematically shown in Fig. 1(b), which is closely related to the cluster decomposition principle in quantum field theory. As the start of the proof, we consider a two-photon in state consisting of two single-photon pulses spatially well separated from each other as shown in Fig. 1(b). By the identical-particle postulate, the in state therefore has the form

$$|\bar{k}_1, \bar{k}_2, L\rangle \equiv \frac{1}{\sqrt{2}}[|\bar{k}_1\rangle \otimes e^{-i\hat{p}L}|\bar{k}_2\rangle + |\bar{k}_2\rangle \otimes e^{-i\hat{p}L}|\bar{k}_1\rangle], \tag{7}$$

where $|\bar{k}\rangle = \int dk f_{\bar{k}}(k)|k\rangle$ describes a single-photon pulse with mean momentum $\bar{k}$ [26]. $\hat{p}$ is the momentum operator, and L is the spatial separation between two pulses. Since the device occupies a localized region in space, the cluster decomposition principle states that the interaction between the two photons should vanish if the spatial separation between the pulses is large enough [23], i.e.,

$$\lim_{L\to\infty}\langle \bar{p}_1, \bar{p}_2, L|T|\bar{k}_1, \bar{k}_2, L\rangle = 0. \tag{8}$$

On the other hand, if we were to assume a T matrix with the form in (6) that contains the product of two δ functions, by inserting the complete momentum basis in (8), one can compute directly that

$$\begin{aligned}\langle \bar{p}_1, \bar{p}_2, L|T|\bar{k}_1, \bar{k}_2, L\rangle = \frac{1}{8}\int dk_1 dk_2 C_{k_1k_2}\{1 + \cos[(k_1 &- k_2)L]\}[f^*_{\bar{p}_1}(k_1)f^*_{\bar{p}_2}(k_2) \\ &+ f^*_{\bar{p}_1}(k_2)f^*_{\bar{p}_2}(k_1)][f_{\bar{k}_1}(k_1)f_{\bar{k}_2}(k_2) \\ &+ f_{\bar{k}_1}(k_2)f_{\bar{k}_2}(k_1)].\end{aligned} \tag{9}$$

The result of (9) contains a term that is independent of L:

$$\frac{1}{8}\int dk_1 dk_2 C_{k_1k_2}[f^*_{\bar{p}_1}(k_1)f^*_{\bar{p}_2}(k_2) + f^*_{\bar{p}_1}(k_2)f^*_{\bar{p}_2}(k_1)] \times [f_{\bar{k}_1}(k_1)f_{\bar{k}_2}(k_2) + f_{\bar{k}_1}(k_2)f_{\bar{k}_2}(k_1)]. \tag{10}$$

This term for arbitrary pulse shape f does not vanish and, hence, violates the cluster decomposition principle of (8). [As an illustrative example, we provide an explicit evaluation of (9) assuming a spatially localized wave packet form for f in Supplemental Material [27].] Therefore, we conclude that the T matrix $T_{p_1p_2k_1k_2}$ should contain only a single δ function as that in (5). No additional δ functions are allowed, and as a result an individual photon's momentum cannot be preserved.

Intuitively, one can visualize the interaction process of the Hamiltonian (1) by imagining that the first photon enters the localized region and generates an excited state. While the region is in the excited state, the second photon then scatters inelastically off the excited state [8]. With such a picture, one should expect that the energy of individual photons is not conserved by the interaction. Our derivation above provides a rigorous support to such an intuitive picture.

As a second main result of this Letter, we now show that we can use the thought-experimental setup of Fig. 1(b) to strongly constrain the analytic properties of the two-photon S matrix. The device in Fig. 1(a) is localized in space.

Therefore, in the single excitation subspace, we generally expect that, after the injection of a single-photon pulse, the amplitude of excitation inside the device should decay in the form $\sum_m e^{-i\omega_m t-\gamma_m t}$ as controlled by a set of simple poles $\omega_m - i\gamma_m$ in the single-photon S matrix. The two-photon state of (7) comes into the same device. After the injection of the first pulse, the excitation in the device then decays into the waveguide. The interaction between the photons can occur only if there remains excitation in the device when the second pulse arrives. Therefore, we expected that, as $L \to \infty$, the outcome of such an interaction, as characterized by $\langle p_1, p_2|T|\bar{k}_1, \bar{k}_2, L\rangle$, should also decay in the form $\sum_m e^{-i\omega_m L-\gamma_m L}$.

The heuristic argument above constrains the analytic properties of the two-photon S matrix in the complex plane of momentum difference between the two incoming photons $\Delta_k = k_1 - k_2$, because the spatial separation between the pulses L is conjugate to Δ_k. To see this conjugate relation, let $E = p_1 + p_2$ be the total momentum and rewrite $C_{p_1p_2k_1k_2}$ as $C_{p_1p_2}(\Delta_k)$ in (5); a calculation in the momentum basis of the plane wave states $|k_1, k_2\rangle$ then results in

$$\langle p_1, p_2|T|\bar{k}_1, \bar{k}_2, L\rangle = \frac{1}{8}e^{-iEL/2}\int d\Delta_k C_{p_1p_2}(\Delta_k) \times (e^{-i\Delta_k L/2} + e^{i\Delta_k L/2}) \times \left[f_{\bar{k}_1}\left(\frac{E+\Delta_k}{2}\right) f_{\bar{k}_2}\left(\frac{E-\Delta_k}{2}\right) + f_{\bar{k}_1}\left(\frac{E-\Delta_k}{2}\right) f_{\bar{k}_2}\left(\frac{E+\Delta_k}{2}\right)\right]. \quad (11)$$

In order to obtain from (11) the desired exponential decay behavior of $e^{-i\omega_m L-\gamma_m L}$, the poles of $C_{p_1p_2}(\Delta_k)$ must be located at $(\Delta_k/2) = \pm((E/2) - \omega_m + i\gamma_m)$, as can be seen by the counter integral in the complex Δ_k plane. Consequently, the heuristic argument above requires that all the poles of the two-photon S matrix in the complex Δ_k plane are completely determined by the properties of single-photon excitation.

Finally, for systems with time-reversal symmetry, the two-photon S matrix is symmetric with respect to the exchange of Δ_k and $\Delta_p \equiv p_1 - p_2$. Therefore, in general, the two-photon T matrix has the form

$$T_{p_1p_2k_1k_2} = \prod_n \frac{A(E, \Delta_k, \Delta_p)}{E - E_n + i\Gamma_n} \prod_m \frac{1}{\left(\frac{\Delta_k}{2}\right)^2 - \left(\frac{E}{2} - \omega_m + i\gamma_m\right)^2} \times \frac{1}{\left(\frac{\Delta_p}{2}\right)^2 - \left(\frac{E}{2} - \omega_m + i\gamma_m\right)^2}\delta(E - k_1 - k_2), \quad (12)$$

where $A(E, \Delta_k, \Delta_p)$ is a fully analytic function with symmetries $A(E, \Delta_k, \Delta_p) = A(E, \Delta_p, \Delta_k) = A(E, |\Delta_k|, |\Delta_p|)$ and can be further constrained by the unitarity requirement of the S matrix. The analysis here completely determined the analytic properties of the two-photon S matrix in the complex Δ_k and Δ_p planes. The additional poles of the two-photon S matrix can exist only in the complex total energy E plane. These poles, if they exist, correspond to two-photon resonances of the device. The location of such a resonance in the complex E plane in many systems can be estimated by diagonalizing the Hamiltonian of the localized region alone in the two-excitation subspace.

In recent years, there have been a large number of specific calculations on photon-photon scattering in integrated systems, where the interaction Hamiltonian H_{int} considered included waveguide coupling to a two-level atom [8–10], multiple two-level atoms [12,14], a multilevel atom [15–17], a cavity with Kerr nonlinear media [18], and a cavity with embedded atoms [20,21]. Our general results, that the T matrix must have the analytic structure of (12) and cannot be of the form of (6), point to a general unifying feature of all these specific calculations. While the concept of the cluster decomposition principle is certainly well known in quantum field theory, the fact that such a principle provides a strong constraint on the photon-photon scattering matrix in integrated systems has never been explicitly pointed out before.

The general discussions above provide an important constraint when one seeks to construct quantum information processing devices. Below, as an example that serves to illustrate and confirm the results above, we consider the possibilities of a two-photon phase gate. In quantum information processing, the two-qubit conditional quantum phase gate is represented by the following unitary transformation:

$$|0\rangle|0\rangle \to |0\rangle|0\rangle, \quad |1\rangle|0\rangle \to e^{i\theta_{10}}|1\rangle|0\rangle,$$
$$|0\rangle|1\rangle \to e^{i\theta_{01}}|0\rangle|1\rangle, \quad |1\rangle|1\rangle \to e^{i(\theta_{10}+\theta_{01}+\Phi)}|1\rangle|1\rangle, \quad (13)$$

where an additional nonzero phase Φ appears due to the designed nontrivial two-qubit interaction. A polarization-based photon phase gate has been demonstrated [28,29], where $|0\rangle$ and $|1\rangle$ correspond to the polarization states of two single photons.

Instead of a polarization-based phase gate, one may speculate on the existence of a momentum-based phase gate [30,31] as defined by the following unitary transformation:

$$|0\rangle|0\rangle \to |0\rangle|0\rangle, \quad |k_1\rangle|0\rangle \to e^{i\theta_{k_1}}|k_1\rangle|0\rangle,$$
$$|0\rangle|k_2\rangle \to e^{i\theta_{k_2}}|0\rangle|k_2\rangle, \quad |k_1\rangle|k_2\rangle \to e^{i(\theta_{k_1}+\theta_{k_2}+\Phi_{(k_1,k_2)})}|k_1\rangle|k_2\rangle. \quad (14)$$

Here k_1 and k_2 are momenta of two single photons. θ_{k_1} and θ_{k_2} are the phase shifts that a single photon experiences as it passes through the device. The gate operation is described by the additional phase $\Phi_{(k_1,k_2)}$ when both photons pass through. Equivalent to (14), the S matrix of such a phase gate can be identified as

$$S_{p_1p_2k_1k_2} = S^0_{p_1p_2k_1k_2} + iT_{p_1p_2k_1k_2},$$

where

$$S^0_{p_1p_2k_1k_2} = e^{i\theta_{k_1}}e^{i\theta_{k_2}}[\delta(p_1 - k_1)\delta(p_2 - k_2) + \delta(p_1 - k_2)\delta(p_2 - k_1)],$$
$$iT_{p_1p_2k_1k_2} = e^{i\theta_{k_1}}e^{i\theta_{k_2}}(e^{i\Phi_{(k_1,k_2)}} - 1)[\delta(p_1 - k_1)\delta(p_2 - k_2) + \delta(p_1 - k_2)\delta(p_2 - k_1)]. \quad (15)$$

We immediately see that the T matrix has the form of (6) that contains two δ functions. Therefore, according to our general argument above based on the cluster decomposition principle, such a two-photon momentum-based phase gate *cannot* be achieved with any system described by the Hamiltonian (1).

To support the general argument against the momentum-based photon phase gate, since many of the phase gate proposals are based on the Kerr nonlinearity [29], we consider a concrete model of photon transport in a single-mode waveguide side-coupled to a ring resonator incorporating Kerr nonlinear media [18] with the following specific Hamiltonian in (1):

$$H_{\rm int}[c_k, c_k^\dagger, a, a^\dagger] = V\int_0^\infty dk[c_k^\dagger a + a^\dagger c_k] + \omega_c a^\dagger a + \frac{\chi}{2}a^\dagger a^\dagger aa, \quad (16)$$

where ω_c is the frequency of the resonator mode. a ($a^\dagger$) is its annihilation (creation) operator that satisfies the standard commutation relation $[a, a^\dagger] = 1$. V is the coupling constant between the waveguide and the ring resonator and is related to the decay time of the resonance $\tau = 1/(\pi V^2)$. χ describes the strength of Kerr nonlinearity in the ring resonator. The Hamiltonian (16) can be solved exactly [18], for example, by the input-output formulism [32] as adopted for calculating Fock state transport [10]. For single-photon transport, the S matrix is simply

$$S_{pk} = \frac{k - \omega_c - i/\tau}{k - \omega_c + i/\tau}\delta(p - k) \equiv e^{i\theta_k}\delta(p - k), \quad (17)$$

where the transmission amplitude of a single photon is a pure phase factor. For two-photon transport, the S matrix is

$$S_{p_1p_2k_1k_2} = S^0_{p_1p_2k_1k_2} + iT_{p_1p_2k_1k_2},$$

where

$$S^0_{p_1p_2k_1k_2} = e^{i\theta_{k_1}}e^{i\theta_{k_2}}[\delta(p_1 - k_1)\delta(p_2 - k_2) + \delta(p_1 - k_2)\delta(p_2 - k_1)],$$
$$T_{p_1p_2k_1k_2} = -\frac{4\chi}{\pi\tau^2}\frac{E - 2\omega_c + 2i/\tau}{E - 2\omega_c - \chi + 2i/\tau} \times \frac{1}{\left(\frac{\Delta_k}{2}\right)^2 - \left(\frac{E}{2} - \omega_c + i/\tau\right)^2} \times \frac{1}{\left(\frac{\Delta_p}{2}\right)^2 - \left(\frac{E}{2} - \omega_c + i/\tau\right)^2} \times \delta(E - k_1 - k_2). \quad (18)$$

We see that the T matrix indeed has the general form of (12). It has a two-photon pole of $2\omega_c + \chi - 2i/\tau$ in the complex E plane and has its analytic properties in the complex $\Delta_{k,p}$ plane exactly of the form of (12). Also, in spite of the common interpretation of such a Kerr nonlinear interaction as an intensity-dependent phase shift, the resulting S matrix does not have the form of a momentum-based phase gate of (15) and does not preserve the momentum of the individual photon.

As a final remark, the results in this Letter remain valid even if the localized region has loss, since loss can always be treated by coupling the region to an additional reservoir. More fundamentally, the validity of the result depends upon a causal relation between the output and input. For a system with gain, such a causal relation may not always exist, since a system with gain can output a photon in the absence of photon input. We expect, however, that our results here should remain valid for any system as long as there is a causal relation between input and output.

In summary, we have considered a general class of quantum-integrated systems and provided a set of general analytic constraints on its two-photon scattering matrix based on a thought experiment that is closely related to the cluster decomposition principle in quantum field theory. Our results indicate that, in general, one cannot construct nonlinear quantum devices in these systems that preserve the momenta of single photons. A similar procedure can be generalized to study the analytic properties of multiple-photon transport as well.

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