Numerical approach to statistical properties of resonance fluorescence

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We formulate a numerical scheme to investigate the correlation of photons scattered by a two-level atom coupled to a waveguide, and compute the second-order correlation function. We also show that a three-dimensional resonance fluorescence experiment can be mapped onto the one-dimensional situation studied here. The correlation functions for both cases show excellent agreement. © 2014 Optical Society of America

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Photon antibunching, which manifests the nonclassical signature of light, was predicted in the resonance fluorescence of a driven two-level atom by a resonant laser field [1,2], and experimentally confirmed by Kimble et al. in 1977 using sodium atoms [3]. Progress has been made, so as to isolate and measure the radiation from single quantum emitters such as individual ions [4]. While the measured correlations from such experiments can be fit by a theoretical treatment given in Refs. [2] and [3], the calculation is rather involved, and the extension of this method to more complicated geometries is not immediately clear.

On a separate front, in recent years, photons confined in a one-dimensional waveguide have been used as the probe in cavity quantum electrodynamic (QED) systems. Due to the potential applications, it is increasingly important to compute the statistical properties of resonance fluorescence in such one-dimensional systems. Although computational schemes using density matrix methods have been developed [6,7], such a scheme assumes a weak coherent excitation, and is not applicable for scenarios with a Fock state input. It is also not transparent to use density matrix methods to investigate the dependence of the photon statistics on the waveform of the incoming photons. In this Letter, we present a different numerical approach to compute the statistical properties of photons interacting with a two-level atom. This approach is based on a real-space Hamiltonian formalism [8], which facilitates a direct numerical computation using the wavefunction of the system. We validate this approach by comparing to experimental data, finding the results to be in excellent agreement.

Figure 1 shows the schematics of the system: a two-level atom coupled to a waveguide. This system represents the most fundamental situation for photon–atom interaction, and serves as a starting point for other important geometries in cavity QED. This system can be described by the real-space Hamiltonian [8]

\[ H = H_w + H_a + H_i. \]

(1)

\( H_w \) is the free Hamiltonian of photons propagating in the waveguide and is given by

\[ H_w = \hbar \int dx \left[ c^\dagger_h(x)(-i\partial_x)c_h(x) + c^\dagger_e(x)(i\partial_x)c_e(x) \right] \]

+ \( \hbar \int dx \left[ c^\dagger_L(x)(i\delta_x)c_L(x) + c^\dagger_R(x)(-i\delta_x)c_R(x) \right], \]

(2)

where \( c^\dagger_h(x) \) and \( c_h(x) \) are creation and annihilation operators for a right-moving photon at position \( x \); \( c^\dagger_L(x) \) and \( c_L(x) \) are creation and annihilation operators for a left-moving photon at position \( x \); \( v \) is the group velocity of photons in the waveguide. The waveguide has a single transverse mode for each operating frequency.

\( H_a \) is the Hamiltonian for the atom and is given by

\[ H_a = \hbar \omega_g a^\dagger_g a_g + \hbar \omega_e a^\dagger_e a_e, \]

(3)

where \( a^\dagger_g \) and \( a_g \) (\( a^\dagger_e \) and \( a_e \)) are creation and annihilation operators for the atomic ground (excited) state; \( \hbar \omega_g \) and \( \hbar \omega_e \) are the energies of the atomic ground state \( |g\rangle \) and excited state \( |e\rangle \) respectively; and \( \omega_a = \omega_e - \omega_g \) is the atomic transition frequency.

\( H_i \) describes the interaction between guided photonic modes and the atom and is given by

\[ H_i = \hbar \int dx V\delta(x)[c^\dagger_R(x)a^\dagger_g a_e + c^\dagger_L(x)a^\dagger_e a_g \]

+ \( a^\dagger_e a_g c_R(x) + a^\dagger_g a_e c_L(x) \].

(4)

\( V \) gives the coupling between the atom (at \( x = 0 \)) and the photonic field in the waveguide, with \( \Gamma = V^2/v \) (\( \Gamma \) gives the spontaneous emission rate of the atom). \( H_i \) describes all emission and absorption processes between the atom and photons. For example, the term proportional to \( c^\dagger_R(x)a^\dagger_g a_e \) describes an atomic transition from the excited \( |e\rangle \) state to the \( |g\rangle \) ground state and the spontaneous emission of a right-moving photon.

Fig. 1. Two-level atom coupled to a waveguide; \( \omega_a \) is the atomic transition frequency; \( \Gamma \) is the atom–photon interaction strength (\( \Gamma \) also gives the spontaneous emission rate of the atom).
To represent a weak coherent input, we use a truncated coherent input that contains the vacuum state, a one-photon state, and a two-photon state. A coherent state $|\alpha\rangle$ with expected photon number $\bar{n} = |\alpha|^2$ is given by [9]

$$|\alpha\rangle = \sum_{n=0}^{\infty} c_n |n\rangle,$$

where

$$c_n = e^{-\bar{n}/2} \frac{\alpha^n}{\sqrt{n!}}$$

and $|n\rangle$ is a photon number (Fock) state. For a weak coherent state, the amplitudes $c_n$ will be vanishingly small for $n > 2$, so the state can be approximated by

$$|\alpha\rangle' = c_0 |0\rangle + c'_1 |1\rangle + c'_2 |2\rangle,$$

where the amplitudes $c'_n$ are the same as the full coherent state amplitudes $c_n$ but normalized such that $|c_0|^2 + |c'_1|^2 + |c'_2|^2 = 1$. We inject into the system a weak coherent pulse, represented by Eq. (7), as the input state, and the state is numerically evolved in time according to the equations of motion until the atomic response has died down.

The equations of motion are given by expanding the Schrödinger equation $i\hbar \partial_t |\alpha(t)\rangle' = H |\alpha(t)\rangle'$ in each photon-number Fock space, where $|\alpha(t)\rangle'$ represents the state of the system with the initial condition $|\alpha(0)\rangle' = |\alpha\rangle'$. For example, the fully interacting two-photon dynamics for this system are described by

$$\dot{\phi}_{RR}(x, t) = -v(\partial_x + \partial_{x'}^c) \phi_{RR}(x, x', t) - \frac{i}{\sqrt{2}} [\delta(x) e_R(x, t) + \delta(x') e_R(x', t)] e^{-i\omega_0 t},$$

$$\dot{\phi}_{RL}(x, t) = -v(\partial_x - \partial_{x'}^c) \phi_{RL}(x, x', t) - \frac{i}{\sqrt{2}} [\delta(x) e_R(x, t) + \delta(x') e_L(x', t)] e^{-i\omega_0 t},$$

$$\dot{\phi}_{LR}(x, t) = -v(-\partial_x + \partial_{x'}^c) \phi_{LR}(x, x', t) - \frac{i}{\sqrt{2}} [\delta(x) e_R(x, t) + \delta(x') e_L(x', t)] e^{-i\omega_0 t},$$

$$\dot{\phi}_{LR}(x, t) = +v(\partial_x + \partial_{x'}^c) \phi_{LR}(x, x', t) - \frac{i}{\sqrt{2}} [\delta(x) e_R(x, t) + \delta(x') e_L(x', t)] e^{-i\omega_0 t},$$

$$\dot{\phi}_{LR}(x, t) = +v(\partial_x - \partial_{x'}^c) \phi_{LR}(x, x', t) - \frac{i}{\sqrt{2}} [\delta(x) e_R(x, t) + \delta(x') e_L(x', t)] e^{-i\omega_0 t},$$

where $\phi_{RR}(x, x', t)$, $\phi_{RL}(x, x', t)$, $\phi_{LR}(x, x', t)$, and $\phi_{LL}(x, x', t)$ are amplitudes corresponding to two photons in the waveguide and the atom in the ground state; $e_R(x, t)$ and $e_L(x, t)$ are amplitudes corresponding to one photon in the waveguide and the atom in the excited state. The equations of motion are evolved numerically in time to trace out the full spatiotemporal dynamics of the scattering process, to which end we have developed efficient pseudospectral codes [10]. The Hamiltonian in Eq. (1) can be easily extended to include, for example, an optical cavity [11] or a ring resonator [12]. The derivation of the equations of motion and the calculation of the statistical properties of the scattered light proceed in exactly the same manner.

For a coherent excitation, the photon statistics for the output are described by the second-order correlation function $g^{(2)}(\tau)$, which is given by [9]

$$g^{(2)}(\tau) = \frac{\langle E^+(-t) E^-(-t + \tau) E^+(-t + \tau) E^+(t + \tau) \rangle}{\langle E^+(-t) E^+(t) \rangle \langle E^+(-t + \tau) E^+(-t + \tau) \rangle},$$

where $\langle \cdot \rangle$ indicates the expectation, and $E^-$ and $E^+$ are the standard electric field operators. Evaluated within the real-space framework corresponding to the Hamiltonian in Eq. (1), this expression reduces to

$$g^{(2)}_{ii}(\tau) = \frac{\langle c^\dagger_i(x_m) c^\dagger_i(x_m + \tau \sigma) c_i(x_m + \tau \sigma) c_i(x_m) \rangle}{\langle c^\dagger_i(x_m) c^\dagger_i(x_m) \rangle \langle c^\dagger_i(x_m + \tau \sigma) c_i(x_m + \tau \sigma) \rangle},$$

where $i = R$ is used for the input or the transmitted state (i.e., when the photons are right-moving), $i = L$ is used for the reflected part (i.e., when the photons are left-moving), and $x_m$ is a reference position for calculating the correlation. While the resulting correlation should not in principle depend on the choice of $x_m$, we find that choosing $x_m$ to correspond to the center of the scattered pulse yields better numerical accuracy.

The injected state at $t = 0$ consists of vacuum, one-photons, and two-photon parts. The shape of $g^{(2)}(\tau)$ is, however, entirely determined by the two-photon part, while the vacuum and one-photon parts influence only the normalization. In the following, we therefore focus on the two-photon part, which consists of two identically overlapping Gaussian wavepackets with spatial width $\sigma$ and central frequency $\omega_0$. 

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\[ \phi_{\text{in}}(x_1, x_2, 0) = \frac{1}{\sqrt{2\pi\sigma^2}} e^{-\left(x_1-x_0\right)^2/4\sigma^2 - \left(x_2-x_0\right)^2/4\sigma^2} e^{i\left(\omega_0/v\right)x_1 + i\left(\omega_0/v\right)x_2}. \]

where \( x_1 \) and \( x_2 \) correspond to the location of each photon in the input port of the waveguide, and \( x_0 \) is the center position of the input pulse (the result will not depend on the choice of \( x_0 \) so long as the initial pulse is entirely in the third quadrant). Such an input is a product state, meaning that the two photons are completely uncorrelated. The square amplitude \( |\phi_{\text{in}}|^2 \) is the probability density and is normalized to unity when integrated from \(-\infty \) to \( \infty \) in both \( x_1 \) and \( x_2 \). It can be shown both numerically and analytically that for a state consisting only of the two-photon part, one has \( g^{(2)}_{\text{input}}(\tau) = 1/2 \) for all \( \tau \), which is expected for a number (Fock) state [13]. On the other hand, when this two-photon part is included as part of a truncated coherent state, one has \( g^{(2)}_{\text{input}}(\tau) = 1 \) for all \( \tau \) as expected for a coherent state. We note that for a single-photon input with the same width, essentially the entire pulse is reflected \((R = 0.999)\) [8].

Figure 2 shows a graphic representation of the two-photon scattering dynamics for the on-resonance case \((\delta = 0)\), where \( \delta \equiv \omega_0 - \omega_a \) gives the detuning of the input frequency from the atomic transition frequency. In this colormap representation, the two-photon input described by Eq. (16) appears as a disk in the third quadrant \((x_1, x_2 < 0)\) as shown in Fig. 2(a). After the two photons are injected into the system at \( t = 0 \), the pulse will move parallel to the \( x_1 = x_2 \) line toward the origin and maintain its shape (i.e., the photons will propagate freely along the input branch of the waveguide) until the pulse reaches the coordinate axes, where the photons are scattered by the atom. At the origin \((x_1 = x_2 = 0)\), the two photons interact with the scatterer at the same time, so saturation effects can dominate, and the correlation between the two photons can be generated. Away from the origin, each photon interacts with the atom individually.

For the output state in Fig. 2(b), the center cut along the \( x_1 = x_2 \) line in the third quadrant (i.e., in the reflection mode) indicates antibunching, as the probability density is zero to numerical precision, so the two photons will always be separate from one another. Conversely, in the first quadrant, the output wavefunction is localized along the \( x_1 = x_2 \) line, indicating bunching in the transmission mode. The localization of the wavefunction in the second and fourth quadrants along the \( x_1 = -x_2 \) line corresponds to the two photons being found at the same distance from the atom but on opposite sides. From this output state, \( g^{(2)}(\tau) \) is calculated by evaluating Eq. (15), and is proportional to the projection of the wavefunction onto the relative coordinate axis (i.e., the \( x_1 = -x_2 \) line).

While \( g^{(2)}(\tau) \) is generated with a particular choice of input width \( \sigma \), we find the resulting \( g^{(2)}(\tau) \) to be insensitive to the chosen value. Figure 2(c) plots \( g^{(2)}(\tau) \) for the input and the reflected part of the output (LL branch). While \( g^{(2)}(\tau) \) is unity for all \( \tau \) for the weak coherent input, corresponding to the photons being completely uncorrelated, the reflected signal produces a dip in the small-\( \tau \) region with \( g^{(2)}(\tau) \) going all the way to zero at \( \tau = 0 \), which along with \( g^{(2)}(0) < g^{(2)}(\tau) \) corresponds to a sub-Poissonian and antibunched signal.

The numerical scheme presented here also facilitates the computation of the statistical properties of photons scattered by an atom in full three-dimensional resonance fluorescence experiments. In the system described in
Ref. [4], a single $^{24}\text{Mg}^+$ ion is trapped and excited by low-intensity laser light, and the fluorescent radiation is detected by collecting emitted photons over a large solid angle. Here we show that such a three-dimensional experiment is effectively a one-dimensional situation, allowing us to apply our formulation to calculate $g_2(\tau)$ for the scattered field. The detected fluorescent photons in the nonforward directions do not interfere with the input and collectively are mapped to the backward scattered photons in one dimension as shown in Fig. 3 to yield identical $g_2(\tau)$. The angular dependence in freespace affects the total photonic flux but not the correlation between photons.

To substantiate this picture, Fig. 4 plots the numerically obtained $g_2(\tau)$ alongside experimental data from Ref. [4] for $\delta/\Gamma = -1, -2.2, -4.6$. As the detuning is moved farther from resonance (Fig. 4, top to bottom), oscillatory features gradually develop. For example, when the detuning is low ($\delta/\Gamma = -1$), the shape of $g_2(\tau)$ is nearly the same as for the on-resonance case, but the transition from zero to unity is no longer monotonic, but slowly rises to peaks near $\Gamma \tau = \pm 1.6$. As the detuning increases, the initial peaks grow in magnitude and move toward lower values of $|\tau|$, and additional ripples form that decay in amplitude as $|\tau|$ increases. In all cases, the numerical results fully capture the features of the experimental data. We emphasize that there has been no free parameter used to fit the experimental data. For the simplest situation studied here, an analytical expression for $g_2(\tau)$ for the one-dimensional case can be derived [14], which is also in excellent agreement with the numerical results.

Fig. 3. Three-dimensional to one-dimensional correspondence. (a) Scattering in free space. (b) Corresponding modes in a one-dimensional waveguide.

Fig. 4. Comparison of experimental and numerical results for $g_2(\tau)$ for $\delta/\Gamma = -1, -2.2, -4.6$. Note that the three-dimensional spontaneous rate in Ref. [4] is 1/2 of that in the one-dimensional case, due to the mapping between the radial direction and the reflected contribution from a bi-directional geometry.

In summary, we have presented a new numerical method for investigating the correlation of photons scattered by a two-level atom coupled to a waveguide. The treatment applies directly, however, to other input states, e.g., different entanglement between photons or different photonic waveforms. Moreover, the theoretical framework can easily be extended to describe more complicated geometries such as an atom embedded in an optical cavity or networks of multiple atom-cavity systems.

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