Generation of correlated photon pairs in different frequency ranges

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The feasibility of generating correlated photon pairs at variable frequencies is investigated. For this purpose we consider the interaction of an off-resonant laser field with a two-level system possessing broken inversion symmetry. We show that the system generates nonclassical photon pairs exhibiting strong intensity-intensity correlations. The intensity of the applied laser tunes the degree of correlation while the detuning controls the frequency of one of the photons, which can be in the terahertz domain. Furthermore, we observe the violation of a Cauchy-Schwarz inequality characterizing these photons.

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I. INTRODUCTION

The nature of light has always intrigued mankind and its study has nowadays culminated in the field of quantum optics investigating matter-field interaction [1,2]. With the first measurement of an intensity-intensity correlation function by Hanbury-Brown and Twiss [3] and the theoretical basis for the characterization of light by Glauber [4,5] scientists have efficient tools in their hands to probe light fields for quantum signatures [6]. In the past few decades the interest in nonclassical light has grown significantly with the advent of quantum computation and information science [7]. Entangled photon pairs turn out to be indispensable for many quantum protocols [8] and quantum algorithms [9]. Currently, there is a series of experimental techniques available to produce entangled photons such as parametric down-conversion [10–12], four-wave mixing [13–15], electromagnetically induced transparency [16,17], and cavity QED [18,19]. Furthermore, an atomic memory for correlated photon states has been realized experimentally, playing an essential role for quantum communication over long distances [20–22]. Recently, a heralded entanglement source of great practical importance has been demonstrated [23,24]. In addition, theoretical considerations have predicted the generation of a correlated photon pair in the x-ray regime from strongly driven atomic ensembles [25]. Very recently, a communication network for quantum information processing has been proposed [26], which consists of numerous different nodes and channels. Since such different nodes may have different characteristic frequencies, there is great interest in investigating nonclassical pairs of photons of different frequencies [27]. As an important milestone in this direction, entangled photons of different but close frequencies limited to the microwave or optical ranges have been generated and detected experimentally [28,29].

Based on this background, we investigate here a two-level system with broken inversion symmetry that is driven by an off-resonant laser field. By means of adjusting the laser frequency ω_L, one can spontaneously generate a photon at an approximate frequency $\omega_L - \omega_0$ and a subsequent photon with transition frequency ω_0. With the parameters of, e.g., gamma-globulin macromolecules, those frequencies can be in the terahertz and optical regimes, respectively (see Fig. 1). We find that this photon pair of different frequencies is both of nonclassical character and entangled because it violates a Cauchy-Schwarz inequality. The advantage of our scheme lies in the fact that the frequency of the longer-wavelength photon can be manipulated by an appropriate selected detuning. This is quite useful in driving a quantum network composed of different nodes of various frequencies including quantum wells or dots of terahertz transition frequencies. Furthermore, the high flexibility distinguishes our model from a cascade three-level system or other down-conversion processes.

II. MODEL

In particular we consider a two-level system (see Fig. 1) with the transition frequency ω_0 described by the orthonormal ground state $|1\rangle$ and excited state $|2\rangle$ with broken inversion symmetry, meaning that the diagonal parts of the dipole operator satisfy the condition $|\boldsymbol{\wp}_{11}| \neq |\boldsymbol{\wp}_{22}|$, where we define $\boldsymbol{\wp}_{ij} = e\langle i|\boldsymbol{r}|j\rangle$ for $\{i,j\} \in \{1,2\}$. The system is driven by a classical off-resonant laser field given by a linearly polarized monochromatic plane-wave field in the dipole approximation $\boldsymbol{E} = \boldsymbol{E}_0 \cos(\omega_L t)$ with laser frequency ω_L and amplitude $\boldsymbol{E}_0$. The sample is surrounded by a quantized environment that accounts for the processes of spontaneous emission [30].

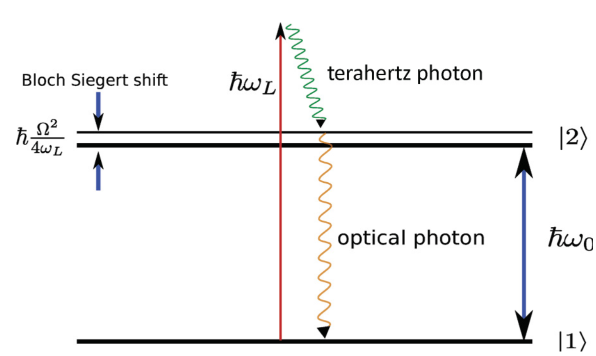

FIG. 1. (Color online) Emission of the nonclassical photon pair. The nonresonant laser excites the two-level system with broken inversion symmetry and induces the emission of a terahertz photon and the subsequent spontaneously emitted optical photon.

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The Hamiltonian H describing the system takes into account the energy of the environment H_E and of the two-level system H_T, the interaction energy between the laser and the two-level system H_{I1}, and the interaction energy between the environment and the two-level system H_{I2}, $H = H_E + H_T + H_{I1} + H_{I2}$, or

$$H = \sum_{\boldsymbol{k}} \hbar\omega_{\boldsymbol{k}} a_{\boldsymbol{k}}^\dagger a_{\boldsymbol{k}} + \hbar\omega_0 S_z + \hbar\Omega(S^+ + S^-)\cos(\omega_L t) + \hbar G S_z \cos(\omega_L t) + i\sum_{\boldsymbol{k}} (\boldsymbol{g_k}\cdot\boldsymbol{d})(a_{\boldsymbol{k}}^\dagger - a_{\boldsymbol{k}})(S^+ + S^-), \tag{1}$$

where we define the Rabi frequency $\Omega = \boldsymbol{\wp}_{12}\cdot\boldsymbol{E}_0/\hbar$ and $G = (\boldsymbol{\wp}_{11} - \boldsymbol{\wp}_{22})\cdot\boldsymbol{E}_0/\hbar$ leads to broken inversion symmetry [31]. Here we have introduced the usual atomic operators $S^+ = |2\rangle\langle 1|$, $S^- = |1\rangle\langle 2|$ and $S_z = (|2\rangle\langle 2| - |1\rangle\langle 1|)/2$ and $a_{\boldsymbol{k}}^\dagger$ and $a_{\boldsymbol{k}}$ denote the creation and annihilation electromagnetic field operators of the $\boldsymbol{k}$th mode of frequency $\omega_{\boldsymbol{k}}$. The coupling constant $\boldsymbol{g_k}$ is defined as $\boldsymbol{g_k} = \sqrt{2\pi\hbar\omega_{\boldsymbol{k}}/V}\hat{\epsilon}_\lambda$, where $\hat{\epsilon}_\lambda$ is the photon polarization vector, $\lambda \in \{1,2\}$, and V is the electromagnetic field quantization volume. The electromagnetic atom-field interaction is given in the usual dipole approximation. We stress the fact that we do not work in the rotating-wave approximation, but rather choose a perturbative approach to account for nonlinear effects. For this purpose we first perform a unitary transformation on H with $H_0 = \sum_{\boldsymbol{k}} \hbar\omega_L a_{\boldsymbol{k}}^\dagger a_{\boldsymbol{k}} + \hbar\omega_L S_z$,

$$\tilde{H} = e^{(i/\hbar)H_0 t}(H - H_0)e^{-(i/\hbar)H_0 t}, \tag{2}$$

which may be separated ($\tilde{H} = \tilde{H}' + \tilde{H}''$) into a time-independent part

$$\tilde{H}' = \sum_{\boldsymbol{k}} \hbar(\omega_{\boldsymbol{k}} - \omega_L)a_{\boldsymbol{k}}^\dagger a_{\boldsymbol{k}} + \hbar(\omega_0 - \omega_L)S_z + \frac{\hbar\Omega}{2}(S^+ + S^-) + i\sum_{\boldsymbol{k}} (\boldsymbol{g_k}\cdot\boldsymbol{d})(a_{\boldsymbol{k}}^\dagger S^- - a_{\boldsymbol{k}} S^+) \tag{3}$$

and a time-dependent part containing fast oscillating terms

$$\tilde{H}'' = \frac{\hbar G}{2} S_z(e^{i\omega_L t} + e^{-i\omega_L t}) + \frac{\hbar\Omega}{2}(S^+ e^{2i\omega_L t} + S^- e^{-2i\omega_L t}) + i\sum_{\boldsymbol{k}} (\boldsymbol{g_k}\cdot\boldsymbol{d})(a_{\boldsymbol{k}}^\dagger S^+ e^{2i\omega_L t} - a_{\boldsymbol{k}} S^- e^{-2i\omega_L t}). \tag{4}$$

The time-dependent part can be regarded as a perturbation to the time-independent part and we can thus apply the second-order perturbation theory [32,33] since $G < \omega_L$, $\Omega < \omega_L$, and $(\boldsymbol{g_k}\cdot\boldsymbol{d}) < \omega_L$:

$$H_{\text{pert}} = -\frac{i}{\hbar}\tilde{H}''\int dt\,\tilde{H}''. \tag{5}$$

Our final Hamiltonian $H_f = \tilde{H}' + H_{\text{pert}}$ acquires the shape

$$H_f = \sum_{\boldsymbol{k}} \hbar(\omega_{\boldsymbol{k}} - \omega_L)a_{\boldsymbol{k}}^\dagger a_{\boldsymbol{k}} + \hbar\left(\omega_0 - \omega_L + \frac{\Omega^2}{4\omega_L}\right)S_z + \frac{\hbar\Omega}{2}(S^+ + S^-) + i\sum_{\boldsymbol{k}} (\boldsymbol{g_k}\cdot\boldsymbol{d})(a_{\boldsymbol{k}}^\dagger S^- - a_{\boldsymbol{k}} S^+) + \frac{3G}{8i\omega_L}\sum_{\boldsymbol{k}} (\boldsymbol{g_k}\cdot\boldsymbol{d})(a_{\boldsymbol{k}}^\dagger S^+ e^{i\omega_L t} - a_{\boldsymbol{k}} S^- e^{-i\omega_L t}) + \frac{\Omega}{2i\omega_L}\sum_{\boldsymbol{k}} (\boldsymbol{g_k}\cdot\boldsymbol{d})(a_{\boldsymbol{k}} - a_{\boldsymbol{k}}^\dagger)S_z, \tag{6}$$

where we keep the slowest-oscillating time-dependent terms only. We notice that the time-dependent terms are proportional to G and are thus important for the description of a system with broken inversion symmetry. The ratios G/ω_L and Ω/ω_L are small for optical frequencies ω_L such that higher orders are negligible in the Hamiltonian. Our perturbative approach also reveals an effect of strong driving fields: the Bloch-Siegert shift $\hbar\Omega^2/4\omega_L$ [34] of the upper state of the two-level system (see Fig. 1). Finally, the two-level approximation applies because $\Omega/\omega_L \ll 1$ and $|\omega_0 - \omega_L|/\omega_L \ll 1$.

In what follows we shall derive the master equation by employing the Hamiltonian in Eq. (6) and the Heisenberg picture. We assume that the matter-field interaction is weak in the sense that an emitted photon does not react with the atom and use the well-known Born-Markov approximation. Thus the time evolution of an arbitrary atomic operator $Q(t)$ is governed by the Heisenberg equation $\frac{d}{dt}\langle Q(t)\rangle = \frac{i}{\hbar}\langle [H_f, Q]\rangle$. By inserting the final Hamiltonian we obtain

$$\frac{d}{dt}\langle Q(t)\rangle = \frac{i}{\hbar}\langle[\tilde{H}_0, Q]\rangle - \sum_{\boldsymbol{k}} \frac{\boldsymbol{g_k}\cdot\boldsymbol{d}}{\hbar}\Big\{\langle a_{\boldsymbol{k}}^\dagger[S^-, Q]\rangle + \langle[Q, S^+]a_{\boldsymbol{k}}\rangle - \frac{3G}{8\omega_L}(\langle a_{\boldsymbol{k}}^\dagger[S^+, Q]\rangle e^{i\omega_L t} + \langle[Q, S^-]a_{\boldsymbol{k}}\rangle e^{-i\omega_L t}) + \frac{\Omega}{2\omega_L}(\langle a_{\boldsymbol{k}}^\dagger[S_z, Q]\rangle + \langle[Q, S_z]a_{\boldsymbol{k}}\rangle)\Big\}, \tag{7}$$

where $\tilde{H}_0 = \hbar(\omega_0 - \omega_L + \Omega^2/4\omega_L)S_z + \hbar\Omega(S^+ + S^-)/2$. To further simplify the analytical formalism we have to express the annihilation and creation operators as a function of atomic operators in the Born-Markov approximation. First, we insert $a_{\boldsymbol{k}}^\dagger(t)$ in the Heisenberg equation and obtain the general solution for the linear inhomogeneous differential equation of first order. Then we consider the leading order in the coupling and neglect the Lamb shift, so the creation operator acquires the shape

$$a_{\boldsymbol{k}}^\dagger(t) = a_{\boldsymbol{k}}^\dagger(0)e^{i\Delta_{\boldsymbol{k}} t} + \frac{\pi\Omega}{2\hbar\omega_L}(\boldsymbol{g_k}\cdot\boldsymbol{d})S_z(t)\delta(\Delta_{\boldsymbol{k}}) - \frac{3\pi G}{8\hbar\omega_L}(\boldsymbol{g_k}\cdot\boldsymbol{d})S^-(t)\delta\left(\Delta_{\boldsymbol{k}} + \omega_0 + \frac{\Omega^2}{4\omega_L}\right)e^{-i\omega_L t} + \pi\frac{\boldsymbol{g_k}\cdot\boldsymbol{d}}{\hbar}S^+(t)\delta\left(\omega_{\boldsymbol{k}} - \omega_0 - \frac{\Omega^2}{4\omega_L}\right), \tag{8}$$

where $\Delta_{\boldsymbol{k}} = \omega_{\boldsymbol{k}} - \omega_L$. We notice that for the annihilation operator $a_{\boldsymbol{k}}$ we only have to take the Hermitian conjugate of the above formula. If we further define the different decay rates of the system

$$\gamma_R = \pi\sum_{\boldsymbol{k}} \frac{(\boldsymbol{g_k}\cdot\boldsymbol{d})^2}{\hbar^2}\delta\left(\omega_{\boldsymbol{k}} - \omega_0 - \frac{\Omega^2}{4\omega_L}\right), \tag{9a}$$

$$\gamma_L = \pi \sum_{\mathbf{k}} \frac{(\mathbf{g_k} \cdot \mathbf{d})^2}{\hbar^2} \delta(\omega_{\mathbf{k}} - \omega_L), \tag{9b}$$

$$\gamma_T = \pi \sum_{\mathbf{k}} \frac{(\mathbf{g_k} \cdot \mathbf{d})^2}{\hbar^2} \delta\left(\omega_{\mathbf{k}} - \omega_L + \omega_0 + \frac{\Omega^2}{4\omega_L}\right) \tag{9c}$$

and insert Eq. (8) in Eq. (7), we may write our final master equation

$$\begin{aligned}
&\frac{d}{dt}\langle Q(t)\rangle - \frac{i}{\hbar}\langle[\tilde{H}_0, Q]\rangle \\
&\quad = -\gamma_R(\langle S^+[S^-,Q]\rangle + \langle[Q,S^+]S^-\rangle) \\
&\quad\quad - \frac{\Omega}{2\omega_L}\gamma_L(\langle S_z[S^-,Q]\rangle + \langle[Q,S^+]S_z\rangle) \\
&\quad\quad - \left(\frac{3G}{8\omega_L}\right)^2 \gamma_T(\langle S^-[S^+,Q]\rangle + \langle[Q,S^-]S^+\rangle) \\
&\quad\quad - \frac{\Omega}{2\omega_L}\gamma_R(\langle S^+[S_z,Q]\rangle + \langle[Q,S_z]S^-\rangle) \\
&\quad\quad - \left(\frac{\Omega}{2\omega_L}\right)^2 \gamma_L(\langle S_z[S_z,Q]\rangle + \langle[Q,S_z]S_z\rangle),
\end{aligned} \tag{10}$$

which may be interpreted as follows. The first term accounts for the spontaneous emission at resonance $\omega_0 + \Omega^2/4\omega_L$, taking into account the Bloch-Siegert shift. The second term describes the spontaneous emission at the laser frequency ω_L preceded by an excitation. The third term corresponds to the emission at frequency $\omega_L - \omega_0 - \Omega^2/4\omega_L$ preceded by an excitation of the two-level system. With the used parameters, later on it has terahertz frequency while the main resonance is optical. The fourth term accounts for a spontaneous emission at resonance preceded by an excitation (off resonant as always). The last term contributes to the dephasing of the system. We are interested in correlations between the processes of the first and third summands that are illustrated in Fig. 1. For this purpose we need to define these correlations and their time-dependent behaviors.

In order to probe the quantum nature of our generated photons, we calculate its intensity-intensity correlation function $g_{ij}^{(2)}$ defined as [1,2]

$$g_{ij}^{(2)}(\tau) = \frac{\langle E_i^{(-)}(t)E_j^{(-)}(t+\tau)E_j^{(+)}(t+\tau)E_i^{(+)}(t)\rangle}{\langle E_i^{(-)}(t)E_i^{(+)}(t)\rangle\langle E_j^{(-)}(t)E_j^{(+)}(t)\rangle}. \tag{11}$$

We know from the definition of the quantized electric field [1] that $\mathbf{E}^{(-)} \propto a_{\mathbf{k}}^\dagger$ and $\mathbf{E}^{(+)} \propto a_{\mathbf{k}}$. In our case we also know from Eq. (8) that for terahertz emission $a_{\mathbf{k}}^\dagger \propto S^-$ and for optical emission $a_{\mathbf{k}}^\dagger \propto S^+$. Therefore, the probability to detect an optical photon after a terahertz photon as a function of atomic operators is given by

$$g_{12}^{(2)}(0) = \frac{\langle S^-(t)S^+(t)S^-(t)S^+(t)\rangle}{\langle S^-(t)S^+(t)\rangle\langle S^+(t)S^-(t)\rangle} \tag{12}$$

and the probability to detect an optical photon followed by a terahertz photon reads

$$g_{21}^{(2)}(0) = \frac{\langle S^+(t)S^-(t)S^+(t)S^-(t)\rangle}{\langle S^+(t)S^-(t)\rangle\langle S^-(t)S^+(t)\rangle}. \tag{13}$$

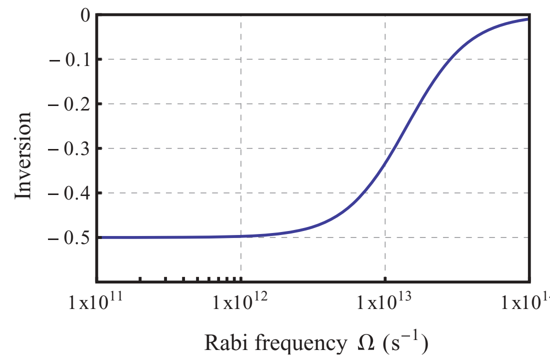

FIG. 2. (Color online) Steady-state inversion operator as a function of the Rabi frequency with a transition frequency $\omega_0 = 5.0 \times 10^{15}$ s^{-1}, laser frequency $\omega_L = \omega_0 + 10^{13}$ s^{-1}, detuning $\Delta = 10^{13}$ s^{-1}, and decay rate $\gamma_0 = 3 \times 10^6$ s^{-1} with respect to ω_0.

III. RESULTS

As a concrete system we consider gamma-globulin macromolecules [35] with the parameters $|\omega_2 - \omega_1| \cong 4.8 \times 10^{15}$ s^{-1}, $|\wp_{21}| \cong 1$ D, and $|\wp_{22} - \wp_{11}| \cong 100$ D. We notice that the transition frequency is optical and we do observe the necessary broken inversion symmetry. We choose the laser detuning such that the long-wavelength photon is in the terahertz domain. Alternative systems are quantum dots, which are zero-dimensional quantum systems having an electron confined in all three space dimensions [36]. Gallium nitride devices, for example, show broken inversion

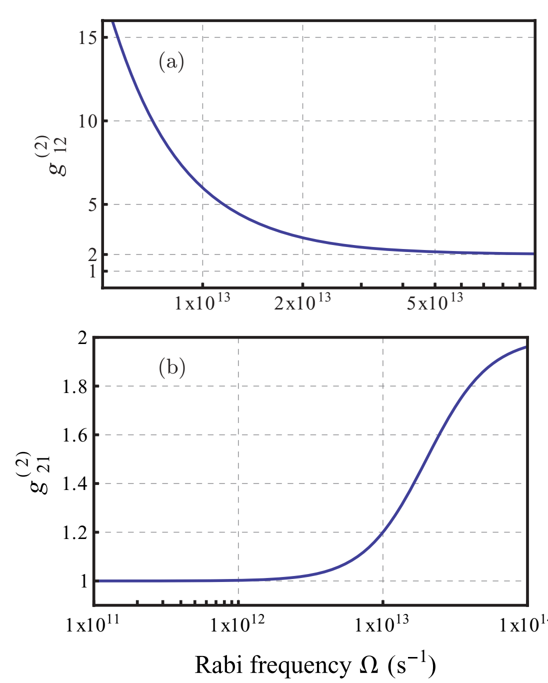

FIG. 3. (Color online) Steady-state second-order intensity-intensity correlation function describing the probability of (a) terahertz emission followed by an optical emission and (b) optical emission followed by terahertz emission as a function of the Rabi frequency Ω. Other parameters are the same as in Fig. 2.

symmetry and have typical values of $|\wp_{22} - \wp_{11}| \cong 10$ D, $|\wp_{12}| \cong 10$ D, and $\omega_0 = 4.92 \times 10^{15}$ s^{-1} [37–40].

First, we display the population inversion $\langle S_z(t)\rangle$ as a function of the Rabi frequency Ω in Fig. 2. We observe that for low Rabi frequencies Ω the population remains in the ground state. At a frequency of about 10^{12} s^{-1} we notice an increase of the population and at 10^{13} s^{-1} we see that there is a nonvanishing probability of finding the system in the excited state. Now we turn to the plot in Fig. 3(a) of the second-order correlation function $g^{(2)}_{12}(0)$ as a function of the Rabi frequency Ω describing the probability of the emission of a terahertz photon and the subsequent emission of an optical photon. We observe a strong correlation that decreases with rising Rabi frequency. To induce the emission of a terahertz photon the system has to be excited from the ground state to the upper state, where it may spontaneously emit an optical photon. Thus, at low Rabi frequencies, the emission of an optical photon is almost always preceded by the emission of a terahertz photon. This explains the high degree of correlation of the photon pair. As Ω increases there is a nonvanishing probability of finding the system in the excited state and an optical emission that is not preceded by a terahertz photon is possible. This means that the correlation decreases. Finally, we discuss the intensity-intensity correlation function $g^{(2)}_{21}(0)$ in Fig. 3(b) describing the probability of detecting a terahertz photon right after an optical photon. It turns out that this probability is very low, as expected. It slowly rises with increasing Rabi frequency Ω.

In this context we also investigate the violation of the Cauchy-Schwarz inequality

$$g^{(2)}_{11}(0)g^{(2)}_{22}(0) \geqslant \left[g^{(2)}_{12}(0)\right]^2. \quad (14)$$

The correlations $g^{(2)}_{11}(0)$ and $g^{(2)}_{22}(0)$ vanish trivially and in Figs. 3(a) and 3(b) we notice nonvanishing cross correlations violating Eq. (14). Thus we are dealing with a nonclassical pair of correlated and entangled photons.

IV. SUMMARY

We have investigated the interaction of a two-level system with broken inversion symmetry and an off-resonance laser field. Using the parameters of, e.g., gamma-globulin macromolecules or certain quantum dots, we have found the possibility of generating a long-wavelength photon in the terahertz regime followed by a photon in the optical frequency range. Furthermore, we have observed a high degree of correlation between these photons and even a violation of a Cauchy-Schwarz inequality. This proves the nonclassical character and entanglement of the photon pair. In the emerging field of quantum information science, nonclassical correlated or even entangled photon pairs of different frequencies are of great interest, finding applications in the realization of a quantum network.

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