Spontaneous parametric downconversion of light by a dielectric nanoparticle

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Abstract. In the present paper, we theoretically consider the generation of entangled photon pairs in the process of spontaneous parametric downconversion of light by a nonlinear dielectric nanoparticle. This nanoparticle is the only source of nonlinearity in the considered model. We assume that the particle is small compared to the wavelength of light so that the process is non-resonant. An intuitive theoretical description is developed for the case when the nonlinear susceptibility tensor of the material of a nanoparticle has a form of the Levi-Civita symbol. In this case, a two-photon amplitude which characterizes a spatial distribution of entangled photon pairs can be interpreted as a result of interference between two induced dipoles, both being orthogonal to the direction of polarization of the pump photon. A generation of entangled surface plasmon-polaritons by a dielectric nanoparticle located near the metal-dielectric interface is also considered.

1. Introduction

Entangled states are a subject of primary interest in the area of quantum optics. The wavefunction of these many-particle states cannot be represented as a product of the one-particle wavefunctions. Thus, there are correlations between the particles. Entangled photon pairs are an example of such states. They are widely used in experiments testing the fundamentals of quantum mechanics, as well as for applications such as quantum cryptography and sensing. The main source of entangled photon pairs is the spontaneous parametric downconversion of light (SPDC) [1]. In this process, a medium with non-zero second-order nonlinear susceptibility \( \chi^{(2)} \) absorbs a pump photon and produces two photons, namely idler and signal ones, with half the frequency.

Nowadays, the generation of quantum states with nanosystems attracts considerable attention in the areas of nanophotonics and quantum optics. For example, the generation of entangled photon pairs in semiconductor waveguides of subwavelength cross sections [2] as well as by quantum dots [3] has been considered. Also, the generation of entangled photon and plasmon-polariton pairs in hyperbolic metamaterials has been studied theoretically in [4]. However, SPDC has a very low efficiency since it is a spontaneous nonlinear process. This considerably limits the possibility of observing entangled pair creation in solitary nanostructures or in small groups of nanoscale objects, which are of special interest for the development of on-chip nanophotonic devices.
Various strategies have been considered to enhance SPDC efficiency in nanostructures. The first is the engineering of quasi-phase-matching in waveguides. Another strategy is the use of the resonant processes in nanoparticles and nanoantennas. In order to study this possibility, the production of photon pairs by a quantum dot located near a plasmonic nanoparticle [5] or a bimodal plasmonic nanoantenna [6] has been considered. However, plasmonic systems do not have an intrinsic nonlinearity due to the centrosymmetric lattice structure of noble metals. Thus, they only act as electric field amplifiers. On the other hand, all-dielectric nanophotonics is one of the most rapidly developing areas of modern nanostructure optics. Dielectric structures demonstrate electric as well as magnetic resonances and very low losses that are unattainable in metals. In addition, they possess considerable \( \chi^{(2)} \) nonlinearity and, thus, are widely used for second harmonic generation. For example, in a series of recent papers [7, 8, 9] a resonant enhancement of second harmonic generation assisted by Mie resonances in dielectric nanoparticles has been reported. Secondary harmonic generation is the opposite process to SPDC. Nevertheless, spontaneous parametric downconversion in dielectric nanostructures, such as nanoparticles or nanoantennas, has been the subject of a very limited number of studies. Namely, photon pair creation in a semiconductor microresonator has been considered numerically in [10].

In the present paper, we consider spontaneous parametric downconversion by a dielectric nanoparticle within the Rayleigh approximation, when the following condition is satisfied:

\[
\frac{\lambda}{n} \gg d, \tag{1}
\]

where \( \lambda \) is the wavelength of the incident radiation, \( n \) is the index of the material of a nanoparticle and \( d \) is the size of a nanoparticle. Thus, a nanoparticle can be considered as a point electric dipole. This simple non-resonant model is of basic interest for the following study of SPDC in dielectric nanostructures. This model provides an analytical solution in the case of a nanoparticle located in vacuum or in the dielectric bulk as well as a semi-analytical solution in the case of a nanoparticle located near a metal-dielectric interface. To our knowledge, such a basic model has not yet been considered.

2. Theoretical description of nonlinear generation

We start with a general theoretical framework of paper [4]. Within this approach, spatial correlations between photons of the generated photon pair are described by the two-photon amplitude

\[
T_{is}(\mathbf{r}_i, \mathbf{r}_s) = \int d_\alpha \mathbf{G}^{\alpha\beta}(\mathbf{r}_i, \mathbf{r}_0, \omega_i) \Gamma_{\beta\gamma}(\mathbf{r}_0, \mathbf{r}_s, \omega_s) d_s d_\beta d_\gamma d_\delta d_\epsilon \mathbf{d}_s \mathbf{d}_\alpha d_\delta \mathbf{d}_\epsilon \mathbf{r}_0, \tag{2}
\]

where the subscript \( i \) denotes the first (idler) photon with a frequency \( \omega_i \), and the subscript \( s \) denotes the second (signal) one with a frequency \( \omega_s \). The dipole moments \( \mathbf{d}_i \) and \( \mathbf{d}_s \) of idler and signal detectors define the polarizations of the detected photons. In the equation above, \( \mathbf{G}(\mathbf{r}, \mathbf{r}_0, \omega) \) is the matrix of the dyadic Green’s function of the generating system, and \( \Gamma(\mathbf{r}) \) is a generation matrix. The latter in the case of the SPDC process is defined as

\[
\Gamma_{ij}(\mathbf{r}) = \chi_{ijk}^{(2)}(\mathbf{r}) E^{\text{pump}}_{k}(\mathbf{r}) e^{-i\omega_{\text{pump}} t}, \tag{3}
\]

where \( \chi_{ijk}^{(2)} \) is the second-order nonlinear susceptibility tensor and \( E^{\text{pump}} \) at frequency \( \omega_{\text{pump}} \) is the pump field which causes nonlinear generation. The integration in Eq.(2) is performed over the volume of the nonlinear system which generates photon pairs. The probability of simultaneous detection of idler and signal photons of given polarizations at the given positions \( \mathbf{r}_i \) and \( \mathbf{r}_s \) is related to the two-photon amplitude as [4]:

\[
W(\mathbf{r}_i, \mathbf{r}_s) = \frac{2\pi}{\hbar} \delta(\hbar \omega_i + \hbar \omega_s - \hbar \omega_{\text{pump}}) |T_{is}(\mathbf{r}_i, \mathbf{r}_s)|^2. \tag{4}
\]
Figure 1. Sketch of photon pair generation in the process of SPDC by a nonlinear dielectric nanoparticle. (a): Initial polarization of a nanoparticle. Incident photon is $z$-polarized. (b): Induced dipoles. (c): Field pattern of a radiating dipole. Electric field vectors are shown with red, and magnetic field vectors are shown with blue.

3. SPDC by a point dipole

When a pump photon interacts with a nanoparticle, the latter is homogeneously polarizes (figure 1a). This polarization field acts as the actual electric field responsible for nonlinear generation in the nanoparticle. Let us define a coordinate system with the $z$-axis being directed along the polarization. Then, the pump field can be expressed as

$$E_{\text{pump}}(r) = \chi E_\delta(r) |e_z\rangle.$$  

(5)

In the equation above, $|e_z\rangle$ is the unit vector in the $z$-direction, $\chi = \chi^{(1)}$ is the scalar linear susceptibility of the nanoparticle, and $E$ is the electric field of the pump photon. In the case of a spherical particle, susceptibility is related to the permittivity of the nanoparticle $\varepsilon$ by the Lorentz-Lorenz equation:

$$\chi(\omega) = \frac{3}{4\pi} \frac{\varepsilon(\omega) - 1}{\varepsilon(\omega) + 2}.$$  

(6)

We consider the second-order nonlinear susceptibility tensor in the form of the Levi-Civita symbol

$$\chi^{(2)}_{ijk} = \epsilon_{ijk}$$  

(7)

as an example. This case corresponds to a crystal of cubic $O$-class symmetry (see [11]). Let us recall that the considered tensor has 6 nonzero components $\epsilon_{xyz} = \epsilon_{zxy} = \epsilon_{yzx} = 1$, $\epsilon_{yxz} = \epsilon_{zyx} = \epsilon_{xzy} = -1$. Then, the generation matrix $\Gamma$ (3) can be expressed as follows:

$$\Gamma_{ij} = \epsilon_{ijk} E_k = \epsilon_{ijz} E_z = E(|e_x\rangle \langle e_y| - |e_y\rangle \langle e_x|),$$  

(8)

with $|e_x\rangle$ and $|e_y\rangle$ being unit vectors in the directions $x$ and $y$, correspondingly. We consider the dyadic Green’s function of a point electric dipole in the following form:

$$G_{\alpha\beta}(r, r', \omega) = \frac{\exp(ik_0 R)}{R} \left[ \left( \delta_{\alpha\beta} - \frac{R_{\alpha} R_{\beta}}{R^2} \right) - \frac{1 - ik_0 R}{k_0^2 R^2} \left( \delta_{\alpha\beta} - 3 \frac{R_{\alpha} R_{\beta}}{R^2} \right) \right],$$  

(9)

where $R = r - r'$, $R = |R|$, $k_0 = \omega/c$.

We start with the simplest case when both of the detectors and the pump field are polarized in the $z$-direction. Then, the following expression for the two-photon amplitude can be obtained:

$$|T_{1s}^z(r_1, r_s)|^2 = \frac{z^2 z_s^2 |r_1 \times r_s|^2}{r_1^2 r_s^2} \left( \frac{9}{r_1} + \frac{3}{r_1^3} \left( \frac{\omega_1}{c} \right)^2 + \left( \frac{\omega_1}{c} \right)^4 \right) \left( \frac{9}{r_s^2} + \frac{3}{r_s^4} \left( \frac{\omega_s}{c} \right)^2 + \left( \frac{\omega_s}{c} \right)^4 \right).$$  

(10)
Figure 2. Simultaneous detection probability \( W(\mathbf{r}_i, \mathbf{r}_s) \) as a function of the position of the idler detector \( x_i, y_i \) for a fixed position of the signal detector. Pump polarization is aligned along the \( z \) axis at the position \( x_0 = y_0 = z_0 = 0 \) nm, and \( \omega_i = \omega_s = 3 \cdot 10^{11} \text{s}^{-1} \).

First panel: Idler and signal detectors are polarized in the \( z \) direction. (a) \( x_s = 500 \) nm, \( y_s = 0 \) nm, \( z_i = z_s = 1 \) \( \mu \text{m} \). (b): \( x_s = y_s = 500 \) nm, \( z_i = z_s = 1 \) \( \mu \text{m} \). (c): \( x_s = y_s = 500 \) nm, \( z_i = 1 \) \( \mu \text{m} \), \( z_s = 0.5 \) \( \mu \text{m} \). (d): \( x_s = y_s = 500 \) nm, \( z_i = 1 \) \( \mu \text{m} \), \( z_s = 3 \) \( \mu \text{m} \).

Second panel: Idler and signal detectors are polarized in the \( x \) direction. Top row: \( x_s = 500 \) nm, \( z_i = z_s = 1 \) \( \mu \text{m} \). (a): \( y_s = 0 \) nm. (b): \( y_s = 100 \) nm. (c): \( y_s = 200 \) nm. (d): \( y_s = 500 \) nm. Bottom row: \( x_s = y_s = 500 \) nm, \( z_i = 1 \) \( \mu \text{m} \). (e): \( z_s = 0.5 \) \( \mu \text{m} \). (f): \( z_s = 1 \) \( \mu \text{m} \). (g): \( z_s = 2 \) \( \mu \text{m} \). (h): \( z_s = 4 \) \( \mu \text{m} \).

Hereafter, we define the two-photon amplitude as divided by the factor \(|\mathbf{d}_i \mathbf{d}_s E_{\text{pump}}|\). It is seen that the detection probability (4) vanishes if the \( z \)-component of the cross product \([\mathbf{r}_i \times \mathbf{r}_s]\) is zero, i.e. if both the signal and idler detectors are located in the plane that is orthogonal to the \( xy \) plane. This is clearly seen in figure 2. At the same time, the amplitude at the given distances between detectors and a point dipole is maximal if the idler and signal detectors are located in the perpendicular directions. Thus, we can say that the photons emitted in the perpendicular directions are correlated, whereas the photons emitted in the parallel directions are anticorrelated.

However, there are several possible alignments of detector polarizations with respect to the pump field in the considered problem. Namely, if the pump is polarized along the \( z \)-axis, after obviously equivalent variants have been excluded, the following combinations remain: \( \mathbf{d}_i, \mathbf{d}_s \parallel z; \)
Figure 3. Simultaneous detection probability $W(r_i, r_s)$ calculated numerically as a function of the in-plane position of the idler detector $x_i$, $y_i$ for $\omega_i = \omega_s = 1.55$ eV. The position of a nonlinear dipole is $x_0 = y_0 = 0$ nm, $z_0 = 10$ nm. The vertical position of the idler detector is $z_i = 10$ nm. The position of the signal detector is $x_s = 200$ nm, $y_s = 0$ nm, $z_s = 10$ nm.

d_i, d_s|\parallel x; d_i, d_s|\parallel y. The expressions for the two-photon amplitude look more complicated in the last three cases due to the absence of the axial symmetry. In figure 2, the first and second situations are presented as well. As a result, it is seen that one can sufficiently tailor the spatial distribution of entangled photon pairs by selecting polarizations of the detectors. Generalization of the obtained results to the case of materials with the second-order nonlinear susceptibility tensor $\chi^{(2)}_{xyz} = \chi^{(2)}_{zxy} = \chi^{(2)}_{yzx} = \chi^{(2)}_{yxz} = \chi^{(2)}_{zyx} = \chi^{(2)}_{xzy}$ (such as GaAs) is straightforward.

4. SPDC by a point dipole located near a metal-dielectric interface

In the present section, we consider SPDC by a dielectric nanoparticle located near the planar metal-dielectric interface. There is a principal difference between this model and the previous one. Indeed, surface plasmon-polaritons as well as photons are generated in such a system. Entanglement of plasmons was observed for the first time in [12]. Since then, the generation of entangled plasmons in nanostructures attracts considerable attention both experimentally and theoretically [4].

As in the previous section, we assume that the second order susceptibility tensor has a form $\chi^{(2)}_{ijk} = \epsilon_{ijk}$, and the condition $\lambda/n \gg d$ is valid. Thus, a particle can be described as a point electric dipole. The Green’s function of the point dipole located near the metal-dielectric interface consists of the free space Green’s function $G_0$ and the reflected part $G_{\text{ref}}$:

$$G(r, r_0, \omega) = G_0(r, r_0, \omega) + G_{\text{ref}}(r, r_0, \omega).$$

We consider the simplest case, when a pump photon is polarized in the $z$-direction along with the signal and idler detectors. In this case, the reflected part of the Green’s function $G_{\text{ref}}$ includes only $p$-polarized waves and can be expressed as follows [13]:

$$G_{\text{ref}}(\rho, \varphi, z) = \frac{i k_0^3}{2} \int_0^{\infty} \hat{M}(s, \rho, \varphi) \exp(isl_1z) ds.$$

In the relation above, we use the polar representation with $\rho = \sqrt{(x-x_0)^2 + (y-y_0)^2}$, $\varphi = \arctg((y-y_0)/(x-x_0))$ and $z = z_i, z_s + z_0$. The normalized wavenumber $s = \sqrt{k_x^2 + k_y^2}/k_0$, where $k_x = k_0 \sin(l_1 x_i), k_y = k_0 \sin(l_1 y_i)$ and $k_0$ is the wavenumber of the pump beam.
and $s_{1z} = k_1 z / k_0$. The index 1 denotes an upper half-space filled with a dielectric. Elements of the matrix $M$ that are relevant for us have the form $M_{xx} = -M_{zz} = -2 \pi i s^2 r_p(\omega) J_1(s \rho) \cos(\varphi)$ and $M_{yz} = -M_{zy} = -2 \pi i s^2 r_p(\omega) J_1(s \rho) \sin(\varphi)$, with $J_1(s \rho)$ being the Bessel function of the first kind and of the first order and $r_p$ being the Fresnel reflection coefficient for $p$-polarized waves. The resulting probability of simultaneous detection $W(r_1, r_s)$ is shown in figure 3. The permittivity of the dielectric is $\varepsilon_d = 1$ and the permittivity of metal is $\varepsilon(\omega) = 1 - \omega_p^2 / (\omega^2 + i \gamma \omega)$, where the plasma frequency of metal $\omega_p = 5$ eV and $\gamma = 0.01 \omega_p$.

We can interpret the obtained results in the same way as in the previous case. Indeed, surface plasmons are generated by a superposition of two dipoles arranged in the $x$ and $y$ directions, correspondingly. Thus, one dipole does not emit any plasmons in the $y$-direction, whereas the other one does not emit in the $x$-direction. As a result, the two-photon amplitude shows mirror symmetry with respect to the plane that includes a line connecting a dipole and one of the detectors and is orthogonal to the interface. At this plane $|T_{is}(r_1, r_s)|^2 = 0$ (see figure 3). Hence, surface plasmon-polaritons emitted by a nonlinear particle in a non-resonant SPDC process are correlated in the orthogonal angular directions and anticorrelated in the parallel directions, if nonlinearity of the nanoparticle has a form $\chi_{ijk}^{(2)} = \epsilon_{ijk}$ and all of the excitations in the system are polarized in the $z$-direction.

5. Conclusion

In the present paper, we have considered spontaneous parametric downconversion of light by a dielectric nanoparticle within a theoretical framework of paper [4]. The particle is described as a point electric dipole. Such an approximation is valid for particles of a size much smaller than the production of the wavelength of incident light and the refractive index of a nanoparticle. Even this simple model demonstrates rich behavior, which depends on the polarizations of signal and idler photons. The obtained results can be interpreted in the interference of the fields produced by two induced dipoles, if the nonlinear susceptibility tensor of the nanoparticle and idler photons. The obtained results can be interpreted in terms of the interference of the waves. The resulting probability of simultaneous detection $W$ is proportional to the Levi-Civita symbol, i.e. for materials of the $O$ symmetry group. The same is true in the case when a dipole is placed near a planar metal-dielectric interface, and surface plasmon-polaritons are excited. This interpretation is supported by the results of numerical calculations.

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