Generation of surface plasmon-polariton pairs by a nonlinear nanoparticle

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Abstract. In the present paper, we study theoretically the generation of surface plasmon-polariton pairs in the process of spontaneous parametric downconversion of light by a nonlinear nanoparticle located near the metal-dielectric interface. A nanoparticle made of GaAs is considered as a particular example. The particle is assumed to be much smaller than the pump radiation wavelength and considered as a point source. We show that spatial correlations between the generated plasmons can be understood as a result of an interference of electric fields produced by dipole sources that are induced in the nanoparticle by the pump photon. A directional two-plasmon generation in certain geometries is observed.

1. Introduction

Spontaneous parametric downconversion (SPDC) is the main method for the generation of entangled photon pairs [1]. In this process, a medium with a non-zero second-order nonlinear susceptibility $\chi^{(2)}$ absorbs a pump photon at the frequency $\omega_{\text{pump}}$ and emits two photons, the idler and the signal one, with frequencies $\omega_i$ and $\omega_s$ such that $\omega_{\text{pump}} = \omega_i + \omega_s$ [1]. Entangled states of light are a subject of primary interest in the area of quantum optics. Their wavefunction can not be represented as a product of one-particle wavefunctions, thus there are correlations between the photons of the pair. Entangled pairs play a fundamental role in quantum mechanics, and are suitable for various applications such as quantum cryptography, sensing and metrology [1].

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] or plasmonic nanoantennas [4, 5] has been considered. The generation of plasmon-polariton pairs in nanostructures attracts considerable attention both experimentally and theoretically [6] from the time the entanglement of plasmons was first observed [7]. However, plasmonic systems do not possess an intrinsic nonlinearity due to the centrosymmetric lattice structure of noble metals. Thus, they act as electric field amplifiers, or perform the nonlinear generation only due to a non-symmetric geometry of resonant modes. To this end, a study of the generation of photon and plasmon-polariton pairs by dielectric nanoparticles and nanoantennas with $\chi^{(2)}$ nonlinearity is of considerable interest.
consists of the free space Green’s function \( G \). The Green’s function of the point dipole located near the metal-dielectric interface \( G \) amplitude as \( r \) signal photons of given polarizations at the given positions system which generates photon pairs. The probability of simultaneous detection of idler and in the nanoparticle. The integration in Eq.(2) is performed over the volume of the nonlinear This polarization field acts as the actual electric field responsible for the nonlinear generation a pump photon interacts with a nanoparticle, the latter polarizes homogeneously (figure 1b). Figure 1. (a): A sketch of the considered geometry. The nanoparticle is shown with grey, the metallic half-space is shown with yellow. The electric field of the pump wave is parallel to the \( x \)-axis, and the pump wave vector is perpendicular to the interface. (b): Initial polarization of a nanoparticle in the \( x \)-direction. (c): Induced dipoles in the \( z \)- and \( y \)-directions.

In the present work, we consider the generation of plasmon pairs with a nanoparticle made of a nonlinear material (figure 1a). A nanoparticle is assumed to be small enough so that the following condition is satisfied:

\[
d < \frac{\lambda}{n},
\]

where \( d \) is the typical size of a nanoparticle, \( \lambda \) is the pump radiation wavelength and \( n \) is the refractive index of the nanoparticle material. Thus, a nanoparticle can be treaten theoretically as an electric dipole. Hereafter we consider the second-order nonlinear susceptibility tensor for GaAs: \( \chi^{(2)}_{xyz} = \chi^{(2)}_{yxy} = \chi^{(2)}_{yxz} = \chi^{(2)}_{xzy} = \chi^{(2)}_{zxy} = 4d_{36} \), with other matrix elements being zero [8].

2. Theoretical description of the nonlinear generation

We apply the general theoretical framework of paper [6]. Within this approach, spatial correlations between photons or plasmons of the generated pair are described via the two-particle amplitude

\[
T_{is}(r_i, r_s) = \int d_{i,\alpha} G_{\alpha\beta}(r_i, r_0, \omega_i) \Gamma_{\beta\gamma}(r_0) G_{\gamma\delta}(r_0, r_s, \omega_s) d_{s,\delta} dr_0, \tag{2}
\]

where the subscript \( i \) denotes the first (idler) photon with the frequency \( \omega_i \), and the subscript \( s \) denotes the second (signal) one with the frequency \( \omega_s \). The dipole moments \( d_i \) and \( d_s \) of idler and signal detectors define the polarizations of the detected photons. In the equation above, \( \hat{G}(r, r_0, \omega) \) is the matrix of dyadic Green’s function of the system, and \( \hat{\Gamma}(r) \) is the generation matrix: \( \Gamma_{ij}(r) = \chi^{(2)}_{ijkl}(r) E^{pump}_k(r) e^{-i\omega_pump t} \), where \( \chi^{(2)}_{ijkl} \) is the second-order nonlinear susceptibility tensor, and \( E^{pump} \) is the pump field which causes nonlinear generation. When a pump photon interacts with a nanoparticle, the latter polarizes homogeneously (figure 1b). This polarization field acts as the actual electric field responsible for the nonlinear generation in the nanoparticle. 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 \( r_i \) and \( r_s \) is related to the two-photon amplitude as \( W(r_i, r_s) = (2\pi/\hbar)\delta(\hbar\omega_i + \hbar\omega_s - \hbar\omega_pump)|T_{is}(r_i, r_s)|^2 \) [6].

Since the condition (1) is satisfied, the nanoparticle 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 \( G_{ref} \): \( G(r, r_0, \omega) = G_0(r, r_0, \omega) + G_{ref}(r, r_0, \omega) \). The free space part has the following form [9]:

\[
G_{0\alpha\beta}(r, r_0, \omega) = \frac{\exp(ik_0 R)}{4\pi R} \left[ \left( \delta_{\alpha\beta} - \frac{R_\alpha R_\beta}{R^2} \right) - \frac{1 - i k_0 R}{k_0^2 R^2} \left( \delta_{\alpha\beta} - 3 \frac{R_\alpha R_\beta}{R^2} \right) \right], \tag{3}
\]
where $\mathbf{R} = \mathbf{r} - \mathbf{r}_0$, $R = |\mathbf{R}|$, and $k_0 = \omega/c$. The reflected part allows only an integral representation [9, 10]:

$$
G_{\text{ref}}(\rho, \varphi, z) = \frac{ik_0}{8\pi} \int_0^\infty \dot{M}(s, \rho, \varphi) \exp(i s_{1z} z) ds. \tag{4}
$$

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 $z = z_i + z_0$. The normalized wavenumber is $s = \sqrt{k_z^2 + k_y^2}/k_0$, and $s_{1z} = k_1 z/k_0$. The index 1 denotes the upper half-space filled with a dielectric. Elements of the matrix $\dot{M}$ that are relevant for the following example have the form $M_{zz} = 2\pi J_0(sp)(\omega) s^3/s_{1z}$ and $M_{yz} = -M_{zy} = -2\pi i s^2 p(\omega) J_1(sp) \sin(\varphi)$, with $J_0(sp)$ and $J_1(sp)$ being the zero order and the first order Bessel functions of the first kind, respectively, and $r_p$ being the Fresnel reflection coefficient for $p$-polarized waves. For numerical calculations we assume that the permittivity of the dielectric is $\varepsilon_d = 1$, and the permittivity of the metal is $\varepsilon(\omega) = 1 - \omega_p^2/(\omega^2 + i\gamma\omega)$, where the plasma frequency of the metal $\omega_p = 7 \cdot 10^{15}$ s$^{-1}$, and $\gamma = 0.01\omega_p$.

The generation matrix can be represented via the dyadic decomposition: $\Gamma = \sum_{\alpha, \beta} C_{\alpha \beta} |e_\alpha⟩⟨e_\beta|$, where $\alpha$ and $\beta$ take values $x$, $y$ and $z$, and coefficients $C_{\alpha \beta}$ are defined by $\chi^{(2)}$ and the pump field. Thus, the amplitude $T$ can be represented as a sum of products of two components of Green’s functions having form $⟨e_i| G |e_\alpha⟩ ⟨e_\beta| G |e_s⟩$, which corresponds to the interference of electric fields from induced dipole sources.

3. Generation of surface plasmon-polariton pairs

Figure 2 shows the two-photon detection probability $W(r_1,r_2)$ for the generation of plasmon-polaritons by a GaAs nanoparticle located at the position $x_0 = y_0 = 0$, $z_0 = 10$ nm. In the case when the pump field is polarized along the $x$-axis, the two induced sources are dipoles parallel to the $y$- and $z$-axis, correspondingly (figure 1b,c). Indeed, $\Gamma \propto \chi^{(2)} |e_x⟩ |e_y⟩ |e_z⟩ |e_s⟩$. The $z$-dipole emits plasmons in all directions in the $(xy)$ plane with the same efficiency, whereas the $y$-dipole does not emit in the direction of the $x$-axis. Combinations of the corresponding Green’s functions define the spatial structure of $W$. It is seen that if one detector is located at the angle $\varphi = \pi/2$ or $\varphi = -\pi/2$ with respect to the pump field, the probability of detection of the second plasmon is maximal in the same direction and vanishes in the opposite direction (figure 2a,b).

In the limiting case when one of the detectors is located along the $x$-axis, the second one can detect a photon in the upper or in the lower half-planes with the same probability (figure 2c). Thus, by changing the orientation of detectors with respect to the nanoparticle one can tailor the correlation between the generated plasmons up to the case when both plasmons are emitted in one direction.

In macroscopic crystals, the efficiency of the nonlinear generation is maximal when the phase matching condition is satisfied. The latter reads as [8]

$$
\mathbf{k}_{\text{pump}} = \mathbf{k}_i + \mathbf{k}_s, \tag{5}
$$

where $\mathbf{k}_{\text{pump}}$, $\mathbf{k}_i$ and $\mathbf{k}_s$ are wave vectors of the pump, idler and signal photons, respectively. As seen from figure 2, this condition is not satisfied. Thus, in our case of SPDC by a nanoparticle, the phase matching condition is replaced by some kind of geometrical selection rules for eigenmodes of the considered system.

Both of the generated surface plasmon-polaritons share the same polarizations due to the structure of the Green’s function $G_{\text{ref}}$. In particular, at an arbitrary point near the interface, the electric field of plasmon-polaritons has non-vanishing components only in the vertical $e_z$ and radial $e_\rho$ directions. Hence, the considered generation of plasmon pairs is analogous to the SPDC Type 1 process [1].
Figure 2. (a): The two-photon detection probability $W(r_i, r_s)$ for $\omega_i = \omega_s = 4.5 \cdot 10^{15} \text{ s}^{-1}$, $\rho_s = 300 \text{ nm}$ and $\varphi_s = \pi/2$. Vertical positions of the nanoparticle centre $z_0$ and detectors $z_{i,s}$ are $z_0 = z_i = z_s = 10 \text{ nm}$. The measured polarization of the electric field is directed along the $z$-axis. (b): The same as (a), but with $\rho_s = 400 \text{ nm}$ and $\varphi_s = -\pi/2$. (c): The same as (a), but with $\varphi = 0$.

4. Conclusion

In the present work, we describe the generation of surface plasmon-polariton pairs by a GaAs nanoparticle in the process of spontaneous parametric downconversion. Spatial correlations between generated plasmons can be understood in terms of an interference between the electric fields generated by induced electric dipole sources. A number and orientations of these induced dipoles are defined by the second order nonlinear dielectric susceptibility tensor of the nanoparticle material, as well as by the direction of the pump electric field. In the simple case, when two of the principal axes of the nanoparticle are parallel to the metal-dielectric interface, one can observe an emission of both plasmons in the same direction for a linearly polarized pump. The efficiency of the generation increases significantly, if the frequencies of the generated plasmons are close to the the surface plasmon resonance frequency $\omega_p/\sqrt{2}$.

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