Directional photon pairs generation by dielectric nanoparticles

Anna Nikolaeva[1], Kristina Frizyuk[1], Nikita Olekhno[1], Alexander Solntsev[2] and Mihail Petrov[1]

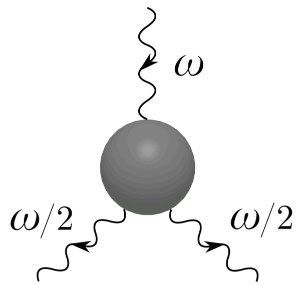
[1]ITMO University, St. Petersburg 197101, Russia

[2]University of Technology Sydney, 15 Broadway, Ultimo NSW 2007, Australia

department of PHYSICS

Introduction

Creation of correlated photon pairs is one of the key topics in contemporary quantum optics. Here, we theoretically describe the generation of photon pairs in the process of spontaneous parametric down-conversion in a resonant spherical nanoparticle made of a dielectric material with bulk $\chi^{(2)}$ nonlinearity. We pick the nanoparticle size that satisfies the condition of resonant eigenmodes described by Mie theory. We reveal that highly directional photon-pair generation can be observed utilizing the nonlinear Kerker-type effect, and that this regime provides useful polarization correlations.



Spontaneous parametric downconversion (SPDC) is an important nonlinear process, during this process one pump photon at frequency ω_p is absorbed and two photons, the idler and the signal, are generated at frequencies ω_i and ω_s. Energy conservation law is satisfied: $\hbar\omega_p = \hbar\omega_i + \hbar\omega_s$, we consider degenerate process assuming $\omega_i = \omega_s = \omega_p/2$.

Mie resonances and Kerker's-type scattering

According to the Mie theory [1], expression for the scattered electric field $\boldsymbol{E}_s$ and the field inside nanoparticle $\boldsymbol{E}_p$ at a fundamental frequency ω_p can be expressed as

$$\boldsymbol{E}_s(k_1,\boldsymbol{r}) = E_0\sum_{n=1}^{\infty} i^n \frac{2n+1}{n(n+1)}\left(ia_n\boldsymbol{N}^{(1)}_{e1n}(k_1,\boldsymbol{r}) - b_n\boldsymbol{M}^{(1)}_{o1n}(k_1,\boldsymbol{r})\right)$$

$$\boldsymbol{E}_p(k_2,\boldsymbol{r}) = E_0\sum_{n=1}^{\infty} i^n \frac{2n+1}{n(n+1)}\left(c_n\boldsymbol{M}_{o1n}(k_2,\boldsymbol{r}) - id_n\boldsymbol{N}_{e1n}(k_2,\boldsymbol{r})\right).$$

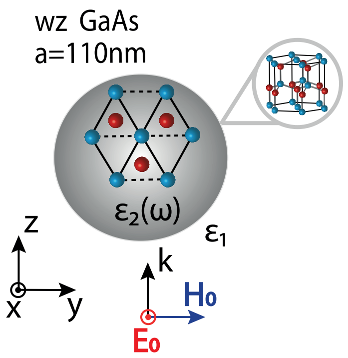


Fig. 1. Geometry of the considered problem. Spherical particle of wurtzite GaAs with a radius $a = 110$ nm, the incident plane wave propagates along z-axis, the electric field oscillates along x-axis, the figure also shows the orientation of the crystal lattice relative to the incident pane wave.

Firstly, we need to understand linear Kerker-type scattering that was done in [2]. The scattering intensity is described by Poynting vector $\boldsymbol{S}$, which in the dipole approximation in the forward ($\theta = 0$) and backward ($\theta = \pi$) direction has the form

$$S(\theta = 0, \pi) \sim |E_1|^2\left[|a_1|^2 + |b_1|^2 \pm 2|a_1^* b_1|\cos\left(\varphi_{b_1} - \varphi_{a_1}\right)\right]$$

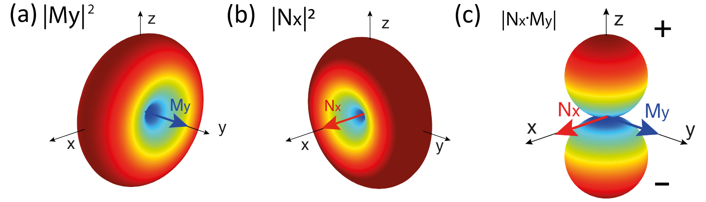


Fig. 3. Scalar products of vector spherical harmonics: (a,b) terms giving the same contribution in the Poynting vector in the forward and backward direction (c) term responsible for the directionality, is included with a different sign in the forward and backward direction.

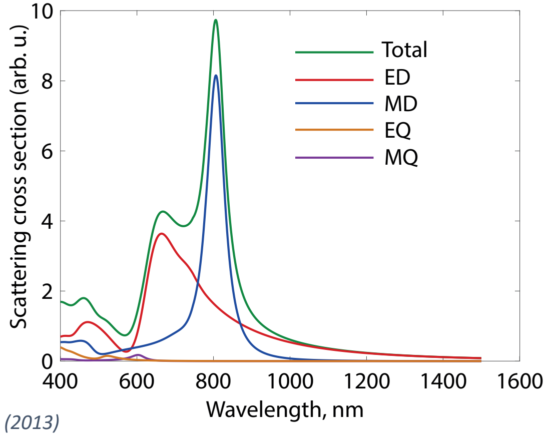


Fig. 2. Scattering cross-section included different multipoles (green - total, red - electric dipole ED, blue-magnetic dipole MD, orange - electric quadrupole EQ, purple - magnetic quadrupole MQ) depending on fundamental wavelength λ_p.

[1] C. F. Bohren and D. R. Huffman. "Absorption and scattering of light by small particles" (1983)

[2] Fu, Y. H. et al. Directional visible light scattering by silicon nanoparticles. Nat. Commun. 4:1527 (2013)

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Theoretical approach

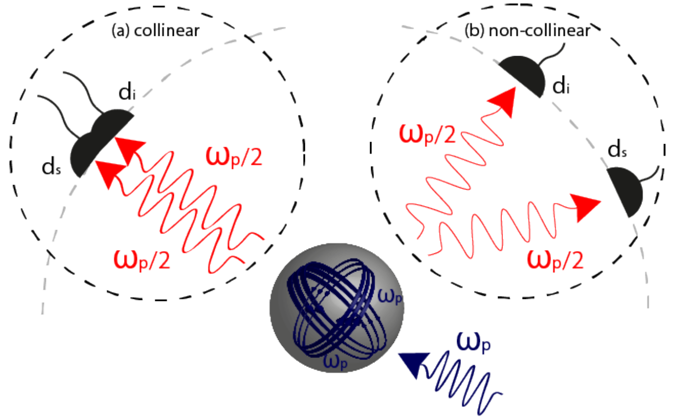

Fig. 4. Schematic SPDC process in collinear decay geometry (a), signal and idler photons are being detected at the same point in the far-field $\boldsymbol{r}_i = \boldsymbol{r}_i = \boldsymbol{r}$, and in non-collinear decay geometry (b)

Using the approach developed in the work [3], we describe correlations between photons of the produced pair via the two-photon amplitude

$$T_{is}(\boldsymbol{r}_i,\omega_i,d_i;\boldsymbol{r}_s,\omega_s,d_s) = \int\limits_V \langle d_i | \underbrace{\hat{\boldsymbol{G}}(\boldsymbol{r}_i,\boldsymbol{r}_0,\omega_i)}_{\text{idler photon}} \overbrace{\hat{\Gamma}(\boldsymbol{r}_0)}^{\text{Nonlinear generation}} \underbrace{\hat{G}(\boldsymbol{r}_0,\boldsymbol{r}_s,\omega_s)}_{\text{signal photon}} | d_s \rangle \, d^3 r_0$$

where d_i, d_s are dipole moments of the idler and the signal detectors

$\hat{G}(\boldsymbol{r},\boldsymbol{r}_0,\omega)$ is dyadic Green's function of the generating system

$\hat{\Gamma}_{\alpha\beta}(\boldsymbol{r}_0) = \chi^{(2)}_{\alpha\beta\gamma}\boldsymbol{E}^{\gamma}_p(\boldsymbol{r}_0)$ is the generation matrix

$\chi^{(2)}_{\alpha\beta\gamma}$ is the second-order nonlinear susceptibility tensor

$\boldsymbol{E}_p(\boldsymbol{r}_0)$ is the pump field which causes the nonlinear generation

Two-photon counting rate:

$$\boxed{W(\boldsymbol{r}_i,\boldsymbol{r}_s) = \frac{2\pi}{\hbar}\delta(\hbar\omega_i + \hbar\omega_s - \hbar\omega_p)|T_{is}(\boldsymbol{r}_i,\boldsymbol{r}_s)|^2}$$

*[3] A. N. Poddubny et al., Phys. Rev. Lett.**117**, 123901 (2016)*

SPDC-SHG correspondence

Let's consider collinear decay, when signal and idler photons emitted in the same direction. Two-photon amplitude can be expand in vector spherical harmonics

$$T_{is}(\mathbf{r}_i,\omega_i,\mathbf{d}_i;\mathbf{r}_s,\omega_s,\mathbf{d}_s) = \sum_{\mathbf{J}_p,\mathbf{J}_i,\mathbf{J}_s} \widetilde{T}_{\mathbf{J}_p\to\mathbf{J}_i,\mathbf{J}_s} \times D_{\mathbf{J}_p\to\mathbf{J}_i,\mathbf{J}_s}\left(\mathbf{d}_i^* \cdot \mathbf{W}^{(1)}_{\mathbf{J}_i}(k_{i1},\mathbf{r}_i)\right)\left(\mathbf{W}^{(1)}_{\mathbf{J}_s}(k_{s1},\mathbf{r}_s)\cdot\mathbf{d}_s^*\right)$$

where $\boldsymbol{W_J}$ is vector spherical harmonic (VSH) and $D_{J_p \to J_i J_s}$ is coefficient describing decaying channels, it contain VSH $\boldsymbol{W}_{J_p}$ from the pump field $\boldsymbol{E_p}(\boldsymbol{r_0})$ and harmonics from dyadic green functions $\widehat{\boldsymbol{G}}(\boldsymbol{r},\boldsymbol{r_0},\omega/2)$. The selection rules for the reverse process - second harmonic generation are determined by similar overlapping integrals [4]. Thus, we have a correspondence between spontaneous parametric downconversion (SPDC) and second harmonic generation (SHG) in the Mie configuration.

$$\boxed{D_{\mathbf{J}_p\to\mathbf{J}_i,\mathbf{J}_s} = \sum_{\alpha,\beta,\gamma}\chi_{\alpha\beta\gamma}\times\int\limits_V W_{\mathbf{J}_p,\gamma}(k_2,\mathbf{r_0})W_{\mathbf{J}_i,\alpha}(k_{i2},\mathbf{r_0})W_{\mathbf{J}_s,\beta}(k_{s2},\mathbf{r_0})d^3r_0}$$

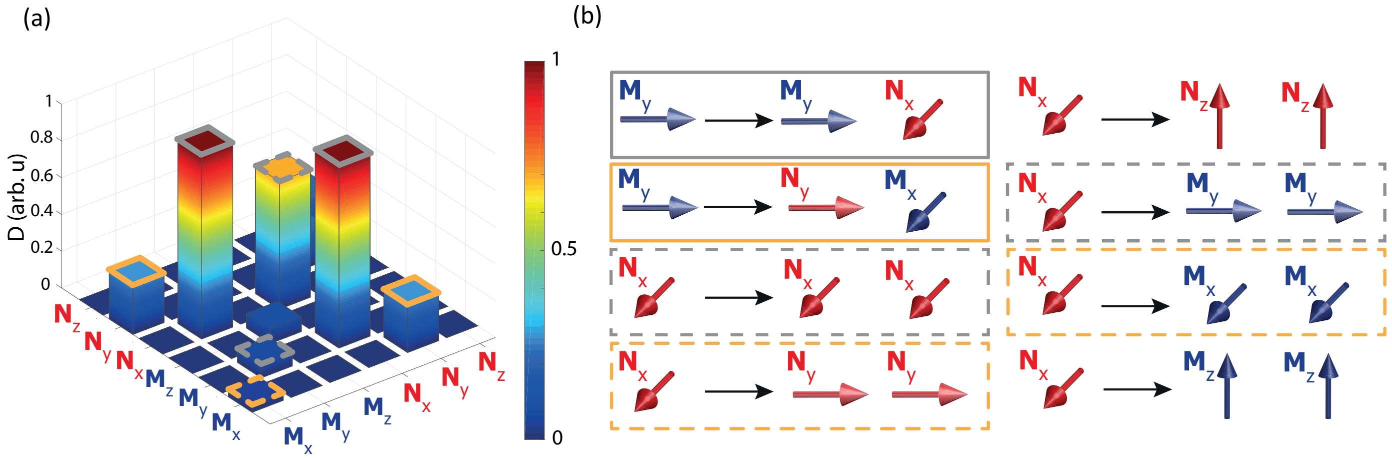

Fig. 5. (a) D-coefficients normalized to the maximum for all possible dipole decays at a wavelength λ_p=720 nm. (b) Possible decay channels in considering geometry, grey or yellow needed to directivity: solid - decay to the crossed dipoles, dashed - decay to the same dipoles.

[4] K. Frizyuk, I. Volkovskaya, D. Smirnova, A. Poddubny, and M. Petrov, Phys. Rev. B 99, 075425 (2019).

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Nonlinear Kerker effect

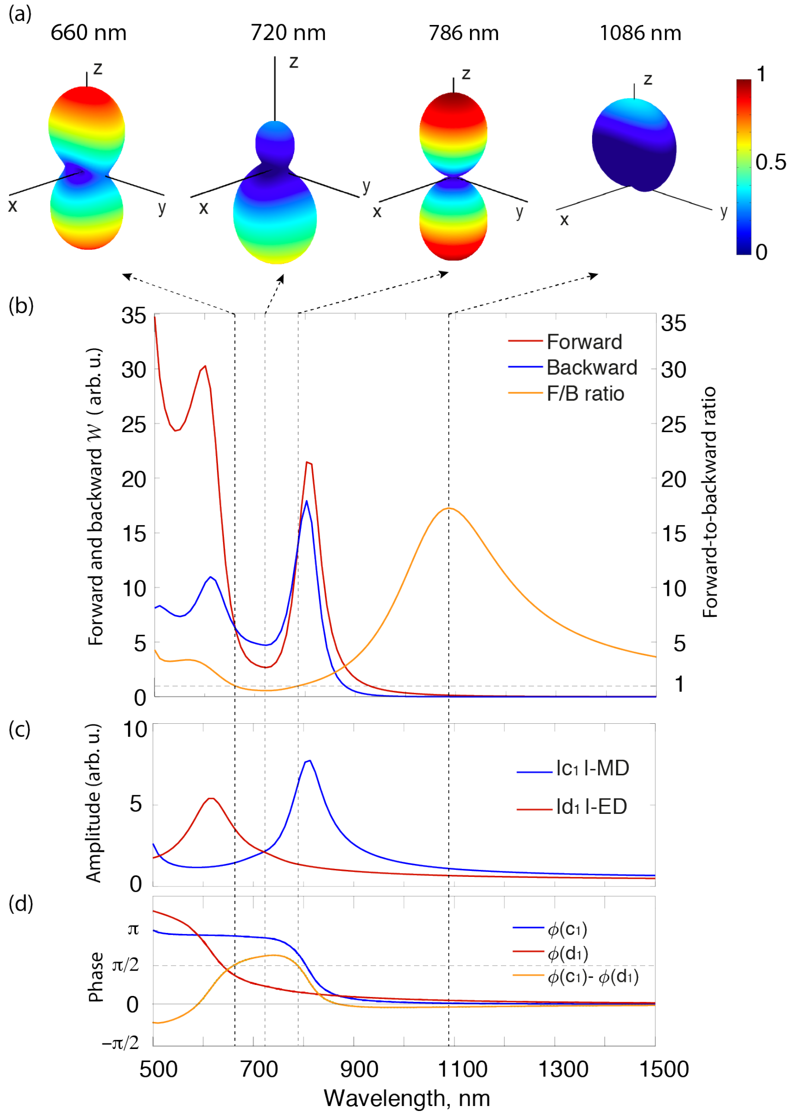


Fig. 6. The far-field patterns of collinear two-photon generation for different wavelengths λ_p=660 nm, 720 nm, 786 nm, 1086 nm. (b) Forward and backward counting rate and their ratio depending on pump wavelength λ_p. (c) Amplitudes of coefficients $|c_1|$ and $|d_1|$ in decomposition of pump field inside nanoparticle E_p. (d) Phases of this coefficients and their phase difference φ_{c_1}- φ_{d_1}.

We consider the difference of unpolarized counting rates in the forward and backward directions in collinear geometry and derive the condition for observing directivity. It can be assumed that the phase difference between the electric dipole $b_1^{(2)}$ and magnetic dipole $a_1^{(2)}$ coefficients in the decay is approximately zero $\varphi_{b_1^{(2)}}$- $\varphi_{a_1^{(2)}}$~0.

$$\boxed{\Delta w^{\text{unpol}} \sim |a_1^{(2)} b_1^{(2)} c_1 d_1| \cos(\Delta\varphi^{pump}) \left[\alpha |a_1^{(2)}|^2 + \beta |b_1^{(2)}|^2\right]}$$

$$\alpha = D_{N_x \to M_x, M_x} D_{M_y \to M_x, N_y} - D_{N_x \to M_y, M_y} D_{M_y \to M_y, N_x},$$
$$\beta = D_{N_x \to N_x, N_x} D_{M_y \to N_x, M_y} - D_{N_x \to N_y, N_y} D_{M_y \to N_y, M_x}.$$

Necessary decays to observe directivity in emission:

1) Crossed magnetic dipole MD and electric dipole ED modes:
$\to M_\alpha, N_\beta$ $(\alpha \neq \beta)$
2) Identical magnetic or electric dipole modes:
$\to M_\alpha, M_\alpha$ or $\to N_\beta, N_\beta$

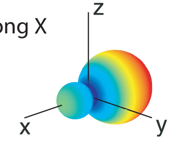
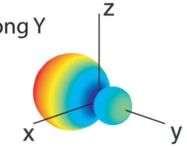
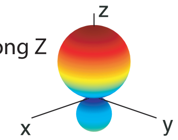
Directivity	Decay on crossed dipoles	Decay on co-aligned dipoles	Non-zero χ⁽²⁾ components	Groups of symmetry
along X 	M_y N_z N_y M_z	M_y M_y Or N_z N_y N_y Or M_z M_z	xxx xyy xzz xyz xxz xxy yxx yyy yzz yyz yxz yxy zxx zyy zzz zyz zxz zxy	C₁ C₁ₕ C₃
along Y 	M_x N_z N_x M_z	M_x M_x Or N_z N_z N_x N_x Or M_z M_z	xxx xyy xzz xyz xxz xxy yxx yyy yzz yyz yxz yxy zxx zyy zzz zyz zxz zxy	C₁ C₃ D₃
along Z 	M_y N_x N_y M_x	M_y M_y Or N_x N_x N_y N_y Or M_x M_x	xxx xyy xzz xyz xxz xxy yxx yyy yzz yyz yxz yxy zxx zyy zzz zyz zxz zxy	C₁ C₁ₕ C₃ D₃ C₃ₕ

Table 1. The first column - directionality along one of the x, y, z axes; the second - decay into crossed dipoles, magnetic and electric; the third is decay into identical dipoles; fourth – non-zero components of the second-order nonlinear susceptibility tensor $\chi^{(2)}$; last column- crystal symmetry groups required to observe directivity.

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Polarization correlations

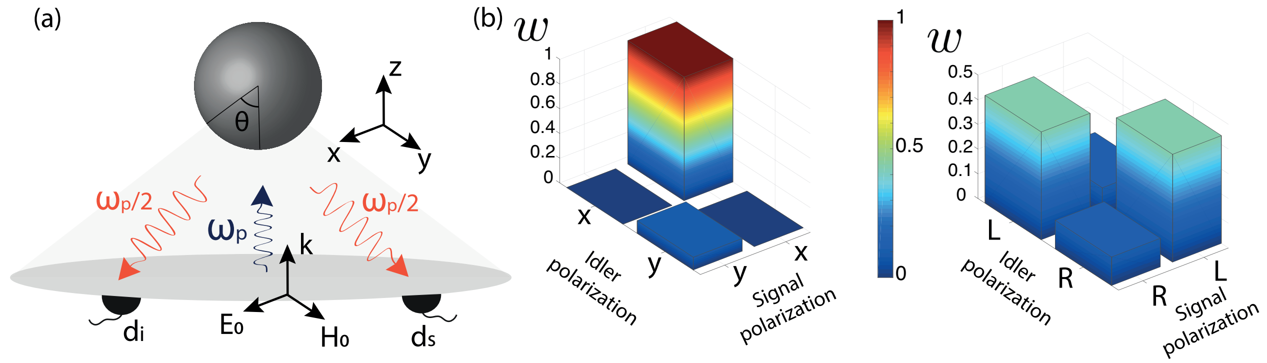

Fig. 7. (a) Configuration of detection in the area limited by the maximum angle $\theta = 60^{\circ}$relative to the backward direction, radius of the sphere and all other parameters are similar as at previous slides (b, c) polarization correlations at λ_p=720 nm in linear basis (b) and in circular basis (c), R- right circular polarization, L- left circular polarization.

You can see that the detected photons in non-collinear geometry have dominantly the same x-polarizations Fig. 7, which corresponds to the D-coefficients shown in the Fig. 5a, where it can be seen that the decays into two electric dipoles directed along x-axis and to the electric dipole along x- axis and magnetic dipole along y-axis are several times higher than all other possible decays.

Conclusion

We have proposed the theory describing the generation of correlated photons through spontaneous down-conversion process in sub-wavelength dielectric resonator supporting lower Mie resonant modes. Using the two-photon amplitude approach, we have identified the mechanism of spontaneous photons decay inn terms of electromagnetic multipoles. As a result we have shown that by proper designing the mode content for particular class crystalline materials one can achieve strongly directional emission of correlated photons. For the collinear geometry provided by idler and signal detectors positioned in the same place, we have formulated the conditions of strongly forward/backward photons generation which surprisingly appeared to be very similar to classical Kerker-effect conditions.