Second harmonic generation and spontaneous parametric down-conversion in Mie nanoresonators

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Abstract. The recent active research in the field of nonlinear Mie nanoresonators has led to significant increase of harmonic generation efficiency from subwavelength structures. The progress in the second order nonlinear processes became evident after switching to nanoresonators made of materials that possess a bulk nonlinearity such as semiconductor and perovskite-type materials. Despite of that, the theory of second-harmonic generation from spherical Mie nanoparticles with a bulk nonlinearity has not been developed yet. In this work, we present a theoretical approach based on Mie theory for describing the second order nonlinear processes in all-dielectric nanoresonators. We focus on the second harmonic generation (SHG) process as well as on the inverse one, the process of spontaneous parametric down-conversion (SPDC). We demonstrate the SHG-SPDC correspondence in terms of Mie resonances and identify the selection rules governing these second order nonlinear processes. We show how one can control the spatial correlations of photons in SPDC from a single Mie-nanoparticle by properly choosing the parameters of the system, in particular, allowing for Kerker-type unidirectional emission of photons.

1. Introduction

The progress of all-dielectric photonics has led to the development of a new class of nonlinear optical devices [1]. This became possible not in the last turn due to a variety of dielectric and semiconductor materials being used, which include those with a non-zero bulk second order dielectric susceptibility tensor $\chi^{(2)}$. This has resulted in the drastic enhancement of SHG from Mie-resonant nanostructures reported in the past two years. The experiments have immediately shown that SHG can be enhanced in dielectric structures with an intrinsic bulk nonlinear susceptibility up to the scale of $10^{-5} - 10^{-4}$ [2, 3, 4, 5, 6] comparing to $10^{-9} - 10^{-8}$ typical for plasmonics [7]. The observed values of the efficiency became achievable due to the high field of Mie modes concentrated inside the nanoresonators, allowing an overlap with bulk sources. However, to control the efficiency of SHG, it is vitally important to identify the mode coupling mechanism in a nonlinear process: eigenmodes of a resonator at the fundamental frequency couple to the modes at the second harmonic (SH) taking into account the specific form of the $\chi^{(2)}$ tensor. The symmetry of both fundamental modes and SH modes combined with the crystalline symmetry either allows or forbids the coupling channels.

Within the prospective of enhanced SHG efficiency in a single Mie resonant structure, it becomes accessible to observe the inverse process referred to as the spontaneous parametric
down-conversion (SPDC) of photons. This effect is well-known for its applications to the entangled photon pairs generation in bulk nonlinear crystals. The recent studies were focused on observing SPDC in one-dimensional on-chip microsystems [8] as well as on the photon pair generation from quantum dots [9], and now the first studies of the SPDC process in a single dielectric nanostructure have been reported [10].

In this work, we theoretically consider the problem of SHG-SPDC correspondence from the Mie theory point of view. We identify the selection rules, which show the mode coupling during the nonlinear emission of photons.

2. Results

Second harmonic generation. We describe the SHG process classically relying on the weak coupling approach, also known as the non-depleted pump approximation. The intensity of the second harmonic generation as of a coherent process is determined by the cross density of states [11]:

\[
I_{2\omega} = \frac{(2\omega)^3 \mu \mu_0}{2} \left[ \int \int dV' dV'' \mathbf{P}^* (r', 2\omega, \omega) \text{Im} \left( \hat{G} (r', r'', 2\omega) \right) \mathbf{P} (r, 2\omega, \omega) \right],
\]

(1)

where \( \hat{G} (r', r'', 2\omega) \) is the dyadic Green’s function, \( \mathbf{P} (r, 2\omega, \omega) = \chi^{(2)}_{ijk} E_j (r, \omega) E_k (r, \omega) \) is the induced polarization vector at the doubled frequency, \( E_j (r, \omega) \) is the component of the pump electric field inside the nanoparticle, and \( \chi^{(2)}_{ijk} \) is the second order nonlinear susceptibility tensor. From (1) one can derive that the intensity of SHG is given by the coefficients \( D_{\mathbf{q}, \mathbf{q}' \rightarrow \mathbf{q}''} \), defining the matrix elements of the transitions:

\[
D_{\mathbf{q}, \mathbf{q}' \rightarrow \mathbf{q}''} = \int \chi^{(2)}_{\alpha\beta\gamma} W^\alpha_{\mathbf{q}} (\omega, \mathbf{r}_0) W^\beta_{\mathbf{q}'} (\omega, \mathbf{r}_0) W^\gamma_{\mathbf{q}''} (2\omega, \mathbf{r}_0) d^3 \mathbf{r}_0.
\]

(2)

Here, the vector \( \mathbf{q} = (n, m, p, t) \) defines the particular resonant mode through four quantum numbers: \( t \) defines whether the mode is magnetic (M-harmonic) or electric (N-harmonic), \( p = \text{odd/even} \) defines the parity of the mode, \( n \) and \( m \) define the multipolar order and the
azimuthal number of the mode. The vectors $\mathbf{q}$ and $\mathbf{q'}$ describe the mode at the fundamental wavelength, while $\mathbf{q''}$ corresponds to the SH state. Here, the summation over different components $\alpha, \beta, \gamma$ is assumed. The intensity of the second harmonic is defined by these coefficients summed over all allowed transitions $\mathbf{q}, \mathbf{q'} \rightarrow \mathbf{q''}$: $I_{2\omega} \sim \sum_{\mathbf{q}, \mathbf{q'}, \mathbf{q''}} |D_{\mathbf{q}, \mathbf{q'} \rightarrow \mathbf{q''}}|^2$.

**Spontaneous parametric down-conversion.** The degenerate SPDC process considers the decay of a photon at the frequency $\omega$ into two photons, the signal and the idler, both having the frequency $\omega/2$, see figure 1a. Its consideration requires a quantum treatment that has been done in a general way in Ref.[12] and involves the two-photon amplitude

$$T_{is}(\mathbf{r}_i, \omega/2, \mathbf{d}_i; \mathbf{r}_s, \omega/2, \mathbf{d}_s) = \int_V \left| \langle \mathbf{r}_i, \mathbf{r}_s, \omega/2 | \hat{G}(\mathbf{r}_i, \mathbf{r}_0, \omega/2) \hat{\Gamma}(\mathbf{r}_0, \omega) \hat{G}(\mathbf{r}_0, \mathbf{r}_s, \omega/2) | \mathbf{d}_s^* \rangle \right|^2 d^3r_0, \quad (3)$$

where the integration is performed over the volume of a nonlinear nanoparticle, $\mathbf{d}_i, \mathbf{d}_s$ are dipole moments of the idler and signal detectors, and $\hat{\Gamma}$ is the generation matrix given by the expression $\hat{\Gamma}_{\alpha\beta}(\mathbf{r}_0, \omega) = \chi^{(2)}_{\alpha\beta\gamma} E_\gamma(\mathbf{r}_0, \omega)$. The magnitude of $|T_{is}|^2$ defines the rate of simultaneous detection of two photons at two given coordinates. The analysis of this expression shows that the detection probability is defined by the same coefficients $|T_{is}|^2 \sim |D_{\mathbf{q}, \mathbf{q'} \rightarrow \mathbf{q''}}|^2$, which represent the efficiency of a mode $\mathbf{q''}$ to decay into modes $\mathbf{q}$ and $\mathbf{q'}$. In the following, we consider a collinear detection when the signal and idler photons are detected at the same position $\mathbf{r}_i = \mathbf{r}_s$.

Considering a resonant pump at the magnetic dipole resonance (see figure 1b), one excites dominantly magnetic and electric dipoles. They can decay into two-photon states via different dipole channels shown in figure 1c. These are allowed transitions for SPDC and reciprocally for SHG. The correlations between different dipole modes can also be seen from figure 2a, where the simultaneous generation of down-converted modes is shown taking into account all modes excited at the pump frequency. The obtained mode coupling results in specific spatial correlations in the two-photon amplitude. The angular diagrams of the collinear two-photon detection rate are shown in figure 2b for GaAs and in figure 2c for BaTiO$_3$. One can see that in the case of GaAs, the simultaneous detection of two photons is most probable in the direction of $y$-axis. It is worth noting that in the case of a BaTiO$_3$ nanoparticle that has a perovskite type crystalline structure [3], one can observe a unidirectional emission of correlated photons in the backward direction. This can be considered as a Kerker-type nonlinear generation of correlated photons.

![Figure 2](image_url)

**Figure 2.** (a): The excitation of different resonant modes during the SPDC decay in a GaAs nanoparticle. (b,c): The directionality diagram of collinear two-photon detection rate $|T(\mathbf{r}_i, \mathbf{r}_s)|^2$ for GaAs (b) and BaTiO$_3$ (c) nanoparticles.
3. Conclusion

We have built a model of the second order nonlinear process in resonant Mie nanoparticles and identified the mechanisms of mode coupling and allowed resonant transitions. We applied this model to the SPDC process and found correlations between different resonant modes simultaneously generated during the decay. Our simulations show that one should expect high directionality of photons emitted during the SPDC process in subwavelength nanostructures. In particular cases, a unidirectional emission is possible. We believe that our results can be of high interest for experimentalists working in the field of nonlinear nanophotonics.

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