Second harmonic generation driven by magnetic dipole moment in dielectric nanoparticles of different shapes.

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Abstract. In this work we suggest a technique helping to identify the selection rules in second harmonic generation from dielectric nanoparticles pumped at their magnetic dipole resonance. We present the identified mode content of the second harmonic field for nanostructures of cylindrical, cone, and prism shapes made of material with $T_d$ and $C_{4v}$ symmetries. This method is applicable for finding the selection rules for arbitrary particle shapes and crystalline structures and can be expanded to other nonlinear processes.

1. Introduction

The generation of nonlinear optical signals is a versatile approach to the generation of the optical signals of different wavelengths. The generation of photons in bulk crystalline media obeys the general principles of energy, momentum, and polarization conservation imposed by the optical properties of nonlinear crystals [1] forming a set of ”selection rules” in bulk media. However, the last two decades the research in nonlinear optics has been focused on efficient subwavelength scale sources. Among other, the metallic nanostructures, which provide surface plasmon resonance, attracted a lot of interest of the researchers as they can enhance the light-matter interaction at the extremely small scales [2–4]. The nonlinear signal generation in subwavelength structures has a pronounced difference in the selection rules, which start to inherit not only the properties of the crystalline structure but also the shape of the nanostructure. In this paper, we restrict ourselves to considering the effect of second-harmonic generation (SHG). The selection rules for SHG from plasmonic systems are quite well studied up-to-date [5]. Recently, the studies of the dielectric nanostructures made of non-centrosymmetric materials have been reported identifying the selection rules in spherical nanoparticles[6]. But still, the selection rules of SHG in dielectric nanostructures of arbitrary shape have not been discussed before and are the central topic of this work.

2. Theoretical approach

We identify the selection rules in dielectric nanostructures in terms of the vector spherical harmonics (VSHs). Being convenient basis functions for expansion of Helmholtz equation
solution in isotropic homogeneous media [7]. VSHs $M_{p,r,mn}$ and $N_{p,r,mn}$ possess special symmetry properties under a point group transformations [8, 9]. They behave in the similar way as tesseral spherical harmonics under SO(3) group rotations, being transformed via Wigner D-matrixes [10] and have well defined behavior under inversion and reflections: for $N_{p,r,mn}$ it is similar as for scalar functions behavior and for $M_{p,r,mn}$ it is opposite. They also correspond to the electric field of photon with angular momentum $n$ and projection $m$, $N$ and $M$ stand for different polarizations [11]. Index $p_r$ exists only for tesseral harmonics, and $p_r = 1$ if function is even under $\phi \rightarrow -\phi$ transformation (or reflection in the $y=0$ plane) and $p_r = -1$ if it is odd.

We will denote the VSH functions with $W_{p_i p_r,mn}$ for both types of vector spherical harmonics, where $p_i = (-1)^n$ or $p_i = (-1)^{n-1}$ stands for $N_{p,r,mn}$ or $M_{p,r,mn}$ respectively. Symmetry properties of vector and scalar spherical harmonics allow us to determine the selection rules for nonlinear processes in nanostructures, e.g. spontaneous parametric down conversion [12, 13], harmonic generation [14], sum frequency generation [15].

$$E^{2\omega}(r) = \sum_{n=1}^{\infty} \sum_{m=0}^{n} \sum_{p_i,p_r} E_0 |D_{p_i p_r,mn}(2\omega)|.$$  

(1)

Here $D_{p_i p_r,mn}$ is proportional to the coupling integral between the incident field and the multipole mode in second harmonic field over the nanoparticle’s volume:

$$D_{p_i p_r,mn} \propto \int_V [W_{p_i p_r,mn}(2\omega) \cdot \tilde{\chi}^{(2)} \cdot E^{\omega}(r) \cdot E^{\omega}(r)] dV .$$

(2)

The dielectric nanostructures provide an artificial magnetic response when the magnetic dipole modes are excited, which is of strong interest for the optical signal manipulation [16]. Thus, we consider the case of the single-mode excitation [17, 18] by $z$-oriented magnetic dipole and rewrite the integral as a coupling integral between three multipoles cartesian projections and the nonlinearity tensor:

$$I_{W',W''\rightarrow W} \propto \chi_{\alpha \beta \gamma}^{(2)} \int_V [N_{\alpha} \cdot W_{p_i p_r,mn}(2\omega)] [N_{\beta} \cdot M_{1-101}(\omega)] [N_{\gamma} \cdot M_{1-101}(\omega)] ,$$

(3)

where $N_{\alpha}$ is the unit vector of the cartesian coordinate system, proportional to $N_{p,m1}(\omega = 0)$. The only non-zero integrals are with $x$- and $y$- projections of the fundamental magnetic dipole as $[N_z \cdot M_{1-101}(\omega)] = [M_{1-101}(\omega)]_z = 0$. 

![Figure 1. Two possible types of nanoparticles shapes with noncentrosymmetric crystalline lattices. Fundamental $z$-oriented magnetic dipole far-field.](image-url)
3. Results and conclusion

In Fig. 2 two cases of lattice symmetries are shown. In the case of $T_d$ lattice symmetry, the components second-order polarizability tensor, which are involved in the process of the single-mode SHG are $\chi_{xyz} = \chi_{yxz}$, all other integrals in (3) are equal to zero. In the case of $C_{4v}$ lattice (actually, tensor has a conical symmetry), the components are $\chi_{xxz} = \chi_{yyz}$.

In order to identify the generated multipole modes, we apply the selection rules, listed in [6, 8], which come from the fact that the integrand of (3) should be invariant under all symmetry transformations of the structure. In Fig. 2 we illustrate this, showing several possible allowed processes is nanostructures of different symmetries. We provide the far-field radiation pattern of the generated harmonic $W_{p_m p_n m n}(2\omega)$, its projection on the $z$-axis (other projections are not involved into the SHG process in this case), and the total angular distribution of the integrand, which contains invariant for considered structure's symmetry.

![Figure 2. Selection rules for a single-mode excitation by magnetic dipole for different particle and lattice symmetries ($T_d$ in top and middle of the table and $C_{4v}$ at the bottom). Some allowed generation cases are illustrated. All cases, where $z$- projections of generated harmonic have similar symmetry with respect to the nanoparticle shape, are also allowed.](image_url)

It is not obvious from the symmetry of three interacting multipoles, which of them are coupled, but taking into the account the lattice symmetry, we obtain that product of six functions under the integral Eq. (3) must contain invariant under particle’s symmetry transformations. For the structures with $T_d$ lattice with the cylindrical ($D_{\infty h}$) shape, the only possible generated multipoles are $M_{-122}$, $N_{-132}$, $M_{-124}$, etc. while the integrand in Eq. (3) contains invariant functions $\psi_{10(2l)}$. For $C_{\infty v}$ shape (cone-like structures), the magnetic dipole at the fundamental wavelength additionally generates the electric quadrupole $N_{-122}$ and so on, as its VSF provides the invariant scalars $\psi_{10(2z-1)}$ in the integral Eq. (3). We obtain that lowering the symmetry provides additional VSHs which become allowed in the second harmonic.
In addition, we can obtain specific multipoles generated by tailoring particle’s symmetry or orientation with respect to the incident wave. For the structures with $C_4$ lattice with the cylindrical (D$\infty$h) shape, the only allowed multipoles are $N_{101}, N_{103}, \ldots$ which are totally different from the ones generated for the $T_d$ cylinder.

The case of cylindrical AlGaAs nanoparticle was also considered experimentally in [18, 19]. Our theoretical results are in good coincidence with experimental results. Since imperfect particle shape can significantly affect the generated fields, the results of this work can also be used to explain the possible discrepancies.

Concluding, in this work we explained the nature of the second-harmonic signal generated by a magnetic dipole in dielectric nanoparticles of non-centrosymmetric material and presented a method for finding the selection rules. We applied it for identifying the mode content of the second harmonic field generated by exciting a magnetic dipole mode at the fundamental wavelength in nanostructures of different shapes. This method is applicable for all possible particle shapes and crystalline structures and can be expanded to other nonlinear processes allowing for shaping the directivity of the nonlinear emission by tailoring the nanoparticle’s symmetry.

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