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Nonlinear circular dichroism in dielectric nanoparticle dimers and trimers

A. D. Nikitina [1]✉, A. A. Nikolaeva [1], M. I. Petrov [1], K. S. Frizyuk [1]

[1] ITMO University, St. Petersburg , Russia

✉ anastasia.nikitina@metalab.ifmo.ru

Abstract. We performed the theoretical study of circular dichroism (CD) in the second harmonic (SH) signal generated in nanostructures of different symmetries. In particular, we explored nonlinear response of dielectric AlGaAs nanoparticles and showed that even in the case of symmetric achiral shape circular dichroism is possible only for some specific shapes and crystalline lattice orientations. Using the apparatus of group theory, we compared dimer and C3v symmetric trimer structures, and explained the appearance of SH-CD in dimer, as well as the absence of dichroism in AlGaAs trimers. In summary, we proved, that knowing the general symmetry of the nanostructure with crystalline lattice is not enough, and detailed analysis of the eigenmodes and nonlinear polarization symmetry is required.

Keywords: second harmonic generation, nonlinear circular dichroism, AlGaAs, dielectric nanoparticle, Mie resonance

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Нелинейный циркулярный дихроизм в димерах и тримерах диэлектрических наночастиц

А. Д. Никитина [1]✉, А. А. Николаева [1], М. И. Петров[1], К. С. Фризюк[1]

[1] Университет ИТМО, Санкт-Петербург, Россия

✉ anastasia.nikitina@metalab.ifmo.ru

Аннотация. В работе проведено теоретическое исследование кругового дихроизма (КД) в сигнале второй гармоники (ВГ), генерируемом в наноструктурах различной симметрии. В частности, был изучен нелинейный отклик диэлектрических наночастиц AlGaAs симметричной ахиральной формы и показано, что даже в этом случае нелинейный круговой дихроизм возможен только для некоторых определенных форм и ориентаций кристаллической решетки. С использованием аппарата теории групп были проанализированы димерные и тримерные структуры и объяснено появление ВГ-КД в димерных структурах, а также отсутствие дихроизма в тримерах AlGaAs. Как итог, было

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продемонстрировано, что знания общей симметрии наноструктуры с кристаллической решеткой недостаточно, требуется проводить детальный анализ собственных мод и симметрии нелинейной поляризации.

Ключевые слова: генерация второй гармоники, нелинейный круговой дихроизм, AlGaAs, диэлектрическая наночастица, резонанс Ми

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Introduction

Circular dichroism is a relevant phenomenon in nanophotonic for exploring the properties of media and molecules. Meanwhile, the nonlinear circular dichroism such as dichroism of the second harmonic (SH-CD) opens new prospective for various application of nonlinear photonics [1–5].

One of the recent works on CD in the SH signal considered a structure that consists of two identical AlGaAs cylinders irradiated by circularly polarized light [5]. The crystalline lattice has a specific orientation [100]‖x, [001]‖z, and the structure can be rotated around the z-axis. The study revealed that CD exists only if the dimer's axis is rotated with the respect to the [100] crystalline axis. An erroneous conclusion can be drawn, that this is due to the low total symmetry of the structure with lattice.

In this work, we demonstrated that the existence of CD in the SHG signal depends on the symmetry in a tricky way, and one should always consider the symmetry of the induced nonlinear polarization and the nanostructure's eigenmodes. To show that the total symmetry does not play a major role, we compared the nonlinear response of dimers and C3v symmetric trimers and obtained and explained via the group theory the total absence of CD in the trimer. All our results were also verified using numerical modeling in COMSOL Multiphysics™.

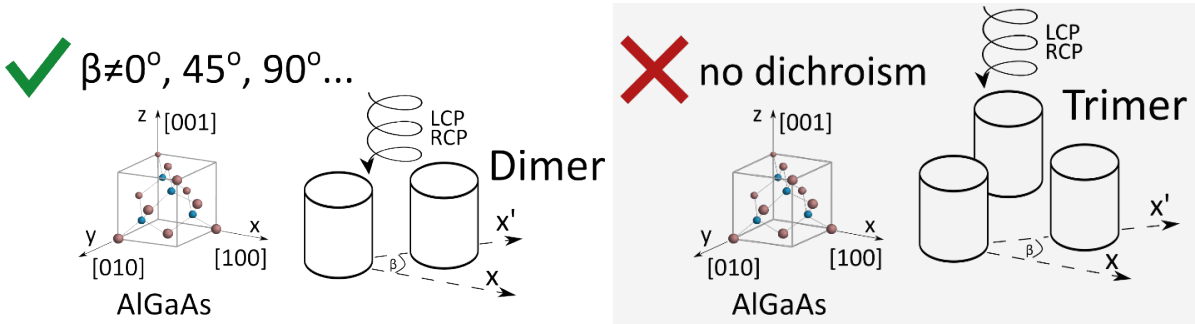

Fig. 1. Illustration of main result of the study of CD in SH signal in AlGaAs nanoparticles. On the left side of the figure, a dimer structure shows a nonvanishing nonlinear CD. It exists only if $\beta \neq Z\pi/4$, where β is the angle between the crystalline x-axis and the sample's x'-axis and Z is an integer number. On the right side of the figure, a trimer structure is shown. SH-CD never appears here for any lattice orientation.

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Materials and Methods

We study dielectric AlGaAs nanostructures with particular symmetry. One can achieve an efficient second-harmonic generation signal because of the lack of inversion symmetry of the crystalline lattice. In this case, nonlinear circular dichroism can appear. The size of the nanostructure is about 1 μm, and the incident wavelength is in the infrared range, while the SH wavelength is in visible range. The size of the AlGaAs nanostructure should be large enough to allow obtaining hexadecapole ($n = 4$) Mie resonances, as it allows us to observe a significant nonlinear circular dichroism in the vicinity of these resonances, which will be explained later. The particular shape of the nanostructure does not play a significant role, only the symmetry group is important. Thus, our considerations are the same for a dimer and rectangular prism, or for a trimer and triangular prism, or any other shapes with these symmetries. For both dimer and trimer structures, we consider the same AlGaAs lattice orientation, $[001]\|z$, $[100]\|x$. The structures are irradiated by normally incident circularly polarized plane wave and can be rotated around the z-axis by arbitrary angle β as it is shown in Fig. 1.

Results and Discussion

Nonlinear circular dichroism manifests itself in the different nonlinear responses of structure for left and right polarization (LCP and RCP). It is described by the formula

$$\text{SH-CD} = 2\frac{(I^{2\omega}_{\text{RCP}} - I^{2\omega}_{\text{LCP}})}{(I^{2\omega}_{\text{RCP}} + I^{2\omega}_{\text{LCP}})}.$$

Considering the dimer, i.e., two identical AlGaAs cylinders, we obtain the nonvanishing nonlinear CD for majority of angles. To provide the theory of the appearance of the nonlinear CD, we should analyse SH polarization $\mathbf{P}^{2\omega}(\mathbf{r})$, defined by the formula

$$\mathbf{P}^{2\omega}(\mathbf{r}) = \varepsilon_0 \hat{\chi}^{(2)} \mathbf{E}^{\text{inc}}(\mathbf{r}) \mathbf{E}^{\text{inc}}(\mathbf{r}),$$

where $\hat{\chi}^{(2)}$ is the nonlinear second-order susceptibility of the crystalline lattice, and $\mathbf{E}^{\text{inc}}(\mathbf{r})$ is the fundamental field inside the nanoparticle. We use the dyadic Green's function formalism to describe the second harmonic field [6, 7]

$$\mathbf{E}^{2\omega}(\mathbf{r}) = (2\omega)^2 \mu \int_V \mathrm{d}V' \widehat{\mathbf{G}}(\mathbf{r}, \mathbf{r}', k) \mathbf{P}^{2\omega}(\mathbf{r}') =$$

$$= (2\omega)^2 \mu \int_V \mathrm{d}V' \sum_n \frac{\mathbf{E}_n(\mathbf{r}) \otimes \mathbf{E}_n(\mathbf{r}')}{2k(k - k_n)} \mathbf{P}^{2\omega}(\mathbf{r}'),$$

where the contribution of each eigenmode $\mathbf{E}_n(\mathbf{r})$ to the SH is described by an overlap integral:

$$D_n = \int_V \mathrm{d}V' \mathbf{E}_n(\mathbf{r}') \mathbf{P}^{2\omega}(\mathbf{r}').$$

We use the approximation for the field on the fundamental frequency inside the nanostructure, assuming that it is the same as in cylindrically symmetric nanoparticle. This approximation is valid, while it is possible to show that taking the exact form of this field does not change the considerations. Thus, field inside the nanoparticle in cylindrical coordinate system is written as follows

$$\mathbf{E}^{inc} = (E_r(r,z)\mathbf{e}_r + E_z(r,z)\mathbf{e}_z \pm \mathrm{i}E_\varphi(r,z)\mathbf{e}_\varphi)\mathrm{e}^{\pm \mathrm{i}\varphi},$$

where φ is an angle in cylindrical coordinates, and ± sign stands for two different circular polarizations. Rewriting the susceptibility tensor in cylindrical coordinates, we obtain that the SH polarization for AlGaAs contains two terms:

$$\mathbf{P}^{2\omega}(r,z,\varphi) \propto \mathbf{P}^{2\omega}_0(r,z) + \mathbf{P}^{2\omega}_4(r,z)\mathrm{e}^{\pm 4\mathrm{i}(\varphi-\beta)},$$

where β is the angle between the crystalline x-axis and sample's x'-axis. The particular form of SH polarization does not matter, we should know only how it depends on φ and β. Let us note that the first term does not transform under rotations around the z-axis, while the second is proportional to exponential term and transforms as the functions with $m = \pm 4$.

Then we should find out which modes are excited in second harmonic, which is defined by the overlap integral. If the integral with a mode is non-zero, it will be excited. The mode content for dimer and trimer is shown in Fig. 2. $\mathbf{W}_{\pm 1 m \ell}$ denote vector spherical harmonics in the far-field of the mode with angular momentum l and projection m. Index ±1 refers to the parity under reflection in $y = 0$ plane [8]. The symmetry group of the dimer is C2v [8]. It has four irreducible representations; hence, it has four types of modes. However, considering the selection rules [7] and overlap integral, we obtain, that only two of them are excited in second harmonic, A1 and A2. We will only consider A1 mode, while for the A2 mode all considerations are the same. According to the group theory, multipoles with indexes $m = 0$ and $m = 4$ both correspond to the A1 irrep (see the multipolar content in Fig. 2). In this case both terms of nonlinear polarization $\mathbf{P}^{2\omega}(\mathbf{r})$ give nonzero overlap integral D_n with the eigenmode A1. Comparing accurately the complex overlap integral for RCP and LCP polarizations [5] and different β, one can obtain that nonlinear circular dichroism appears for proper orientation between the lattice and pattern axis. In contrast, symmetry group of the trimer is C3v. Its eigenmodes with $m = 0$ and $m = 4$ belong to different irreps, A1 and E (see Fig. 2). So, the first term of nonlinear polarization will excite the A1 mode, and the second term the E mode. These two modes are excited independently, and due to this fact, the coefficients of their excitation are equal for both polarizations. As a result, in trimers, nonlinear CD cannot be observed, despite the low symmetry of a timer.

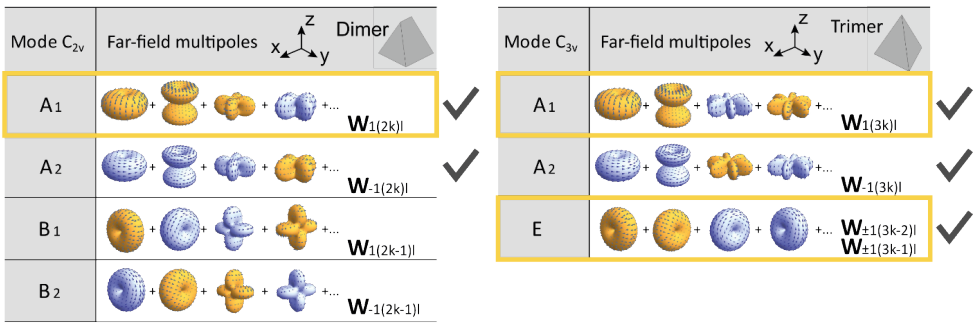

Fig. 2. Comparison of multipolar content of the modes of a dimer and a trimer. $\mathbf{W}_{\pm 1 m \ell}$ denote vector spherical harmonics in the far-field of the mode with angular momentum l and projection m. Index ±1 refers to the parity under reflection in y = 0 plane. The modes excited in SH are marked by a tick. In case if A1 mode is excited in dimer by both terms of the polarization, in trimer A1 mode is excited by the first term, and E by the second. This leads to independent contribution of these two modes in the intensity.

Conclusion

In summary, we provide the theoretical description of SH-CD. We describe, how the symmetry of the crystalline lattice and the structure together affect nonlinear response. We show that the symmetry behaviour of nonlinear polarization, as well as the mode content in the SH plays a crucial role. For considered case, the nonlinear polarization contains two terms with a different symmetry behaviour, each of them excites modes of the same symmetry in dimer case, and of different symmetries in case of a trimer. This leads to the presence of circular dichroism in case of a dimer structure and the absence in case of a trimer.

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THE AUTHORS

NIKITINA Anastasia
anastasia.nikitina@metalab.ifmo.ru
ORCID: 0000-0002-0396-2761

NIKOLAEVA Anna
anna.nikolaeva@metalab.ifmo.ru
ORCID: 0000-0003-1692-9298

PETROV Mihail
m.petrov@metalab.ifmo.ru
ORCID: 0000-0001-8155-9778

FRIZYUK Kristina
k.frizyuk@metalab.ifmo.ru
ORCID: 0000-0002-0506-464X

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