Second harmonic Circular Dichroism in Achiral Nanostructures

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Abstract. We theoretically show that under circularly polarized plane wave or vortex beam illumination, the second-harmonic circular dichroism is possible even if the nanostructure is achiral. The interplay of nanostructure's and crystalline lattice's symmetries leads to specific conditions for observation of the circular dichroism, which can be expressed by a short formula. This can be particularly important for chiral sensing enhancement with nanostructure, where it is important to separate the signal from the nanostructure itself.

1 Introduction

Alongside linear chiral spectroscopy, nonlinear [1, 2] chiral spectroscopy is important for the complete characterization of chiral samples or biological molecules. Achiral nanostructures can be used for enhancement of chiral signal [3]. However, in the nonlinear regime, such nanostructures can have a strong chiral response, which was also shown experimentally [4]. The aim of our work is to provide a way to determine if a particular achiral structure possesses circular dichroism in second harmonic (SH-CD), or not. In our study, we define the nonlinear circular dichroism as

$$\text{SH-CD}_{m,\lambda} = \frac{(I^{2\omega}_{R_m} - I^{2\omega}_{L_m})}{(I^{2\omega}_{R_m} + I^{2\omega}_{L_m})}. \quad (1)$$

Here $I^{2\omega}_{R_m}$, $I^{2\omega}_{L_m}$ are total intensities of a second-harmonic generated by a nanostructure, integrated over a sphere in the far-field, under irradiation by a right- or left-handed vortex beam [5], with total angular momentum projection $+m$ or $-m$ respectively [6].

2 Results

We consider only the possibility of a CD signal, but not its amplitude, however, it was shown that it can be stronger in the vicinity of resonances [4]. The possibility of the SH-CD is solely determined by symmetries, (i) the nanoparticle's symmetry, (ii) the crystalline lattice symmetry, (iii) the relative rotation of the lattice to a nanoparticle's symmetry plane [4, 7, 8]. The nanoparticle's symmetry group is $C_{\infty v}$ or $D_{\infty h}$ (Schoenflies notation). The lattice's symmetry can be any without the inversion center to be able to produce the second-harmonic signal in the bulk material. However, one should take into account, that second harmonic generation is described via nonlinear susceptibility tensor $\hat{\chi}^{(2)} = \chi_{ijk}\mathbf{e}_i \otimes \mathbf{e}_j \otimes \mathbf{e}_k$ [9], which has higher symmetry than the lattice. It can be written in cylindrical coordinates (ρ, φ, z) [10] and the multipliers of the form $e^{im_\chi \varphi}$ can be extracted. There are always several different terms with different m_χ for each lattice, thus we denote all possible

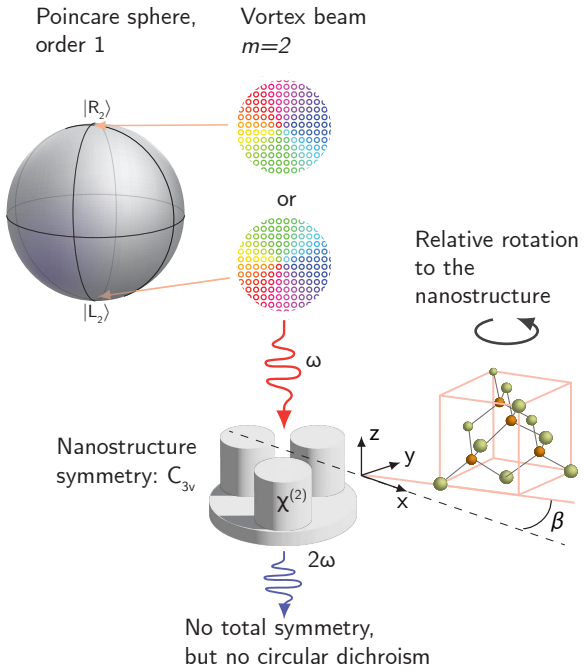

Figure 1. Schematic of the concept. The incoming vortex beams, with helicity $\lambda = \pm 1$, and angular momentum projection $m = \pm 2$ is illustrated with use of high-order Poincaré sphere [5]. Here, the example of dielectric nanostructure with the symmetry C_{3v} and GaAs (z||[001]) crystalline lattice is depicted. The crystal lattice is oriented at an angle β relative to the nanoparticle's symmetry axis and is characterized by Δm_χ as dictated by the $\hat{\chi}^{(2)}$ tensor. For this particular case, SH-CD is absent, despite the low total symmetry.

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differences between them as Δm_χ. We also denote the relative angle of rotation as β (see Fig. 1). After thoughtful analysis of modal excitations and their interplay, one can show that circular dichroism can only appear if the equality

$$\Delta m_\chi = \mathfrak{n}\nu, \quad \nu \in \mathbb{Z} \tag{2}$$

is possible for at least one Δm_χ. This reflects the interplay between lattice and nanoparticle symmetry, and allows eigenmodes of the same symmetry to be excited simultaneously in the second harmonic, but with different phases depending on the incident light polarization. However, to ensure this phase difference, the relative angle should be $\beta \neq \pi\nu/\Delta m_\chi$ at least for one Δm_χ. Let us thus apply this condition for a particular case of GaAs trimer, as depicted in Fig. 1. One can show, that for GaAs $m_\chi = \pm 2$, $\Delta m_\chi = 4$ and $\mathfrak{n} = 3$ for the trimer. Thus, $4 \neq 3\nu$, and SH-CD is not possible in this structure. We would like to emphasize that this happens not due to the total symmetry, and the nanostructure is large enough and should not be described as a "big molecule". However, it would be possible in a dimer ($4 = 2 \cdot 2$) or a symmetric quadrumer (or, equivalently, square prism, $4 = 4 \cdot 1$).

3 Conclusion

In our work, we derived a simple condition, to determine, if the second harmonic circular dichroism is possible in a particular nanostructure. We applied it to some particular cases, and showed, that despite the low total symmetry of the nanoparticle and lattice together, the dichroism does not appear in GaAs trimer (or, equivalently, triangular prism) structure, but possible in a dimer or square prism.

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