Circular dichroism in optical second harmonic generated in reflection from chiral G-shaped metamaterials

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Abstract. Influence of chirality on the optical second harmonic generated from planar array of G-shaped metamaterials is studied. Circular dichroism of these nanostructures manifests itself via different efficiency of left and right circularly polarized second harmonic that is observed for the samples of different handedness. This difference allows to distinguish between the two enantiomers.

1. Introduction
Metamaterials are artificial structures that possess unique properties not existing in natural materials [1], [2]. Optical planar metamaterials can demonstrate such intriguing effects as negative refraction [3] or asymmetric transmission [4]. In case of planar chiral metamaterials optical properties observed previously only for 3D chiral media can be observed, for example, circular dichroism [5] and circular conversion dichroism [6]. The shape of a metamaterial influences much the interaction of the electromagnetic field with the structure by causing specific field distribution in it. The symmetry of a metamaterial can bring about new features of this interaction and determines the existence of such effects as circular conversion dichroism and some other.

Second harmonic generation (SHG) is a perspective probe of 2D chiral metamaterials first of all because of an extremely high sensitivity of the SHG process to the symmetry of the structures [7]. It has been demonstrated that local field distribution strongly influences the parameters of the SHG [8], [9]. It is proven that SHG measurements can distinguish the chirality of the samples: namely, azimuthal dependencies of the SHG intensity have been demonstrated to be different for the two enantiomers composed of G- and mirror G-structures [10]. A strong field localization in such structures has been detected using confocal SHG measurements that turned out to be the main mechanism of the azimuthal dependence of the SHG intensity [11].

Linear optical effects such as changing the ellipticity of light and polarization plane rotation under the reflection from 2D chiral metamaterials have been studied in [12], while there has
been no similar studies of the nonlinear response of such metamaterials. Here we report the observation of circular dichroism in optical second harmonic generation from chiral G-shaped nanostructures under their excitation by linearly polarized fundamental beam.

2. Samples and experimental setup

Samples consist of periodic array of G-shaped nanostructures, made of 25 nm thick Au deposited onto SiO$_2$/Si substrate. Distance between two structures is 200 nm and linewidth of gold is also 200 nm, period of the structure is 1.2 µm. The whole sample consists of 3333x3333 structures. G-shaped structure was prepared by electron-beam lithography on an area of 1x1 µm$^2$ [13]. There were two types of samples: single- and double-periodic.

For the experiments we used the output of a Ti:Sapphire laser at 780 nm with the mean power of 150 mW, pulse width of 70 fs and the repetition rate of 80 MHz. The pump beam was focused onto a spot of 50 µm in diameter. The angle of incidence of the pump beam was 45°. Reflected SHG radiation was spectrally selected by BG39 Schott color filters, passed through a diaphragm with the angular aperture of 5° and was detected by a photomultiplier. SHG polarization was controlled by Babinet-Soleil compensator and a polarizer. Linearly polarized pump beam was used. SHG polarization for studied samples was elliptical. The azimuthal angle $\psi = 0^\circ$ corresponds to the case when the plane of incidence is parallel to one of the sides of the sample (for single-periodic structures the orientation of the plane of incidence is shown on the inset in Fig. 3).

3. Experimental results

3.1. Double-periodic samples

Figure 2 shows the dependencies of the intensity of the left and right circularly polarized SHG, $I_{\omega}$, reflected from G- and mirror-G-shaped structures on the azimuthal angle, $\psi$. The p-polarized fundamental radiation was used. All the graphs reveal the four-fold symmetry. It can be seen that the efficiency of right and left circularly polarized SHG, e.g. the sign of the third Stokes parameter $S_3 = I_{\text{right}} - I_{\text{left}}$ [14], is different for mirror structures. Here the subscripts correspond to the different circular polarizations. This result demonstrates that the SHG probe allows for distinguishing of the enantiomers with different handedness.

It is worth noting that there is no such difference in the linear optical response of G-shaped structures. Polarization of reflected from studied structures light at fundamental beam is also elliptical. Figure 3 shows the azimuthal angular dependencies of the intensity of the left and right circularly polarized light, $I_{\omega}$, reflected from G- and mirror-G-shaped structures for the p-polarized fundamental beam. The maxima of intensity of left circularly polarized light correspond to the minima of right circularly polarized light, while the sign of ellipticity (or direction of electric field vector rotation) of the radiation is the same for two mirror structures. This property may be caused by specific absorption of circularly polarized light by these structures because different ellipticity of the reflected light can be observed in two enantiomers of such symmetry for linearly polarized light [15].

![Figure 1. General view of single G-shaped structure and incidence plane location. Round arrow stands for direction of azimuthal rotation of the sample. Sample sizes are given in microns](image-url)
Figure 2. Circularly polarized SHG intensity dependence on azimuthal angle for double-periodic structures, a) is for mirror-G-shaped structures, b) is for G-shaped structure. Fundamental beam was p-polarized.

Figure 3. Intensity dependence of circularly polarized light reflected from the structure on azimuthal angle for double-periodic structures, a) is for mirror-G-shaped structures, b) is for G-shaped structure. Fundamental beam was p-polarized.

Figure 4. Circularly polarized SHG intensity dependence on azimuthal angle for single-periodic structures, a) is for mirror-G-shaped structures, b) is for G-shaped structure. Fundamental beam was s-polarized.
3.2. Single-periodic samples

The anisotropy of circularly polarized SHG from one-periodic metamaterials is shown in Fig. 4. The maximum SHG efficiency is observed as the fundamental electric field is parallel to the one of the sides of the \( G \) element, i.e. at the azimuthal angles \( \psi = 0^\circ, 90^\circ, 180^\circ, 270^\circ \). An interesting feature of the dependencies shown in Fig. 4 is the different efficiency of the left and right circularly polarized SHG. The highest maximum of the right circularly polarized SHG (open circles) for the case of \( G \)-shaped metamaterials (top panel) corresponds to \( \psi = 180^\circ \), while the highest maximum of the left circularly polarized SHG (filled circles) is attained at \( \psi = 0^\circ \). The opposite situation is observed in mirror-\( G \)-metamaterials (bottom panel): the highest efficiency of right circular SHG is observed at \( \psi = 0^\circ \) (open circles) and the highest left circular SHG at \( \psi = 180^\circ \) (filled circles). These differences are attributed evidently to the chirality of the samples when the handedness of metamaterials leads to different anisotropy of circularly polarized nonlinear-optical response.

4. Discussion

This effect can be explained in terms of local field amplification at the fundamental or SHG wavelength. It is well known [9] that the second harmonic intensity can be described as follows:

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I_{2\omega}(\psi) \propto (\chi^{(2)})^2 < L_{2\omega}^2(\psi) > I(\omega),
\]

where \( L_{\omega}, L_{2\omega} \) are the local field factors at the corresponding wavelengths, \( I(\omega) \) is the pump intensity and the brackets denote the statistical averaging over the laser spot area. In our resent paper [11] we showed that there is a strong field localization on the fundamental frequency in such \( G \)-shaped metamaterials. Thus there are SHG hotspots [13] which can be considered as the main SHG sources that determine the SHG azimuthal dependencies from the whole structure. Mutual arrangement of the SHG hotspots can influence the phase difference between p- and s-polarized SHG and thus give rize to predominantly right or left circularly polarized SHG.

For azimuthal angles \( \psi = 0^\circ \) and \( \psi = 180^\circ \) of single-periodic \( G \)-shaped structures (Fig. 3) the phase difference is maximal. For double-periodic structures the sign of the phase difference remains nearly constant due to the arrangement of all the \( G \)-elements, which influences much the field distribution[16] in the structure and consequently the SHG properties.

5. Conclusions

In conclusion, circular dichroism in SHG from \( G \)-shaped chiral nanostructures is observed for linearly polarized fundamental beam, while no such effect is detected in the linear reflectivity. SHG hotspots interaction is considered as possible mechanism of the observed nonlinear-optical effect.

6. References

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