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Pillai, R.S.; Oh-e, M.; Yokoyama, H.; Brakenhoff, G.J.; Müller, M.

Published in:
Optics Express

DOI:
10.1364/OE.14.012976

Citation for published version (APA):
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Rajesh S. Pillai
Swammerdam Institute for Life Sciences, Universiteit van Amsterdam, P.O. Box 94062, 1090 GB Amsterdam, The Netherlands
sreedhar@science.uva.nl

Masahito Oh-e, and Hiroshi Yokoyama
Liquid Crystal Nano-system Project, ERATO/SORST, Japan Science & Technology Agency, 5-9-9 Tokodai, Tsukuba, Ibaraki, 300-2635, Japan
oh-e@nanolc.jst.go.jp, yokoyama-hiroshi@aist.go.jp

G. J. Brakenhoff and Michiel Müller
Swammerdam Institute for Life Sciences, Universiteit van Amsterdam, P.O. Box 94062, 1090 GB Amsterdam, The Netherlands
muller@science.uva.nl, Brakenhoff@science.uva.nl

Abstract: The nature of the third-harmonic generation (THG) process in a nematic liquid crystal is investigated for the case of tightly focused, low intensity, laser beams. Colloidal particle induced topological defects in a liquid crystal are visualized in three-dimensions using the dependence of the THG signal on both changes in non-linear susceptibility and the orientation of the liquid crystal director relative to the incident laser polarization state. We have found that the interpretation of THG images in a liquid crystal is complicated not only by the change in polarisation of the electric field as it propagates through the medium but also by anisotropic refractive index mismatch induced aberrations.

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OCIS codes: (190.4160) Multiharmonic generation; (190.4720) Optical nonlinearities of condensed matter; (190.4410) Nonlinear optics, parametric processes; (180.6900) Three-dimensional microscopy; (160.3710) Liquid crystals

References and links


1. Introduction

Due to their orientational ordering, liquid crystals (LCs) exhibit specific optical properties which have found many important technological applications. For instance, controlling the birefringence of LCs with an external electric field has found widespread applications in display technology [1], as well as in spatial light modulators used to control amplitude and phase of electromagnetic fields [2]. Recently, colloidal particles dispersed in a nematic LC host have played an important role in LC nano-technology. Current research on LC/colloid systems focuses on explaining how particles induce topological defects and self-assembled ordered structures [3]. Suspended small spherical particles induce elastic distortions in a LC host, and hence give rise to long range changes in the LC molecular orientational order near the particles. Depending on the type of interactions between the particles and the LC molecules, a number of topological defects have been reported [4-6]. Here, we investigate such orientational changes in the vicinity of particles by using three-dimensional third-harmonic generation (THG) microscopy. In addition, LCs are known for their strong non-linear optical properties, which, combined with the inherent birefringence of the LC, can result in efficient non-linear optical conversions [7-9]. A detailed understanding of the THG process in LCs is required for proper interpretation of the THG images of such a system.

Due to the usually small third-order non-linear optical susceptibility (\(\chi^{(3)}\)), THG typically is a weak process. In addition, for focused laser beams THG is not observed, due the Gouy phase shift [10, 11], within the bulk of a homogeneous medium with normal dispersion (i.e. with \(\Delta k = 3k_f - k_s = 6\pi(n_f - n_s)/\lambda < 0\)). Here \(n_f\) and \(n_s\) denote the refractive index at the fundamental (\(\lambda_s\)) and third-harmonic wavelength, respectively. Indeed, an interface in either refractive index and/or \(\chi^{(3)}\) is required for significant THG [12, 13]. This property of THG is used in label free imaging in both biology and the material sciences [14-18]. Due to the non-linear nature of the interaction, THG has inherent optical sectioning properties and thus true three-dimensional imaging capability.

For a medium with positive dispersion (\(\Delta k > 0\)), however, as in the case of a strongly birefringent medium [10, 19], efficient THG is possible also in the absence of an interface. Two different schemes have been reported in the literature for enhancing the detected THG signal from the bulk of a nematic LC. First, a high power tightly focused polarized beam orients LC molecules and creates a helical structure within the focal volume [8]. Such laser-
induced molecular orientations start to appear when the intensity at the focal plane is of the order of 1 MW/cm² [7]. By adjusting the pitch of this helical structure, through changing the average input power, a phase matched THG from the bulk of the LC can be obtained [8]. Second, when the LC is kept slightly away from the beam waist, the focused beam and the polarization of the incident beam can be adjusted to realize a type II phase matching [9]. Changing the temperature of the LC, and thus the dispersion, can be used to further enhance the signal generation in both approaches.

In this report, we demonstrate the use of THG microscopy to image - in three dimensions - colloidal particle induced topological defects in a nematic LC for the first time.

2. Materials and methods

The THG microscopy experimental set-up is described in detail elsewhere [20]. Briefly, the collimated, 1062 nm output of a 113 fs, 72 MHz repetition rate laser (High Q Gmbh, Austria) is focused down to a diffraction limited spot using a high numerical aperture (NA) objective (63x/1.25 oil). The sample configuration and the coordinates are illustrated in Fig. 1. The polarization of the input fundamental beam and the detected output THG signal is controlled using wave plates and Glan Taylor prisms. The sample consists of a nematic LC medium, which is obtained by sandwiching the liquid crystal (type 5CB) molecules mixed with polystyrene beads of approximately 5 μm diameter, between two cover glasses coated with polyimide. The polyimide coated sides of the cover glass face each other and the coating is rubbed in the y direction, so that the LC molecules orient along the y-axis. Consequently, the LC director is also parallel to the y-axis. Typically the distance between the two cover glasses, i.e. between the glass-LC and LC-glass interfaces, is 30 μm. The THG signal is detected using a spectrometer equipped with a cooled CCD camera. Typical acquisition times are 100 ms per pixel. The sample is piezo scanned in three dimensions (PZT P-611.3S, Physik Instrumente).

3. Results and discussion

In our measurements a tightly focused laser beam is used within the LC to observe topological defects induced by the presence of particles. So we need to make sure that the laser beam does not change the orientational structure of the LC [8]. Figure 2(a) shows THG z-responses (i.e. the THG intensity as a function of axial position) as a function of power. The z-responses are taken far away from the particles excluding topological defects in the sample. The THG...
signals are normalized to unity at the glass/LC interface \((z = 0 \, \mu m)\). The (linear) input polarization is set at an angle of \(\phi = \pi/4\) relative to the rubbing direction. This is the angle at which a maximum torque is applied to the LC molecules [8]. Consistent with earlier work [8], an oscillatory behavior is observed in the THG signal from the bulk of the LC for a laser intensity at the focal plane of 0.75 MW/cm\(^2\). However, reducing the intensity to 0.37 MW/cm\(^2\) diminishes the laser induced oscillatory features, which eventually disappear at an intensity level at the focal plane of 0.25 MW/cm\(^2\). Figure 2(b) shows a z-response for the case of an input circular polarization with an intensity of 0.25 MW/cm\(^2\). Note that for these polarization conditions a clear non-zero THG signal is observed from the bulk of the LC medium at all laser intensities. Unless indicated otherwise, an excitation intensity level of 0.25 MW/cm\(^2\) has been used in all experiments presented below.

In order to understand the nature of the signal generation process, we measure the THG signal from within the LC (at a depth of approximately 5 \(\mu m\) from the interface and far away from any sphere). Figure 3(a) shows the THG signal as a function of the angle between the input polarization and the LC director. Maxima in the THG signal are observed at \(\phi = 0.22\pi\) and 0.83\(\pi\). Figure 3(b) shows the THG signal as a function of the angle between the LC director and analyzer. In this case the input polarization used corresponds to either linear polarization (red circles) at an input polarization angle \(\phi = 0.22\pi\) [i.e. the maximum in Fig. 3(a)] or circular polarization (blue squares). Also in the case of circular polarization there is no indication of a laser induced molecular reorientation at an intensity level of 0.25 MW/cm\(^2\).

### 3.1 Type II phase matching

While for a medium with normal dispersion \((\Delta k < 0)\) THG requires an inhomogeneity, such as an interface in refractive index and/or \(\chi^{(3)}\), the third-harmonic signal from the bulk of a medium with positive \(\Delta k\) is given by [10]:

\[
I_{\text{THG}} \propto b^4 \Delta k^2 \exp(-b\Delta k) \quad (1)
\]

In general, this situation of positive phase mismatch can be achieved for the case of a medium with strong birefringence such as crystalline materials [19].

For the, birefringent, nematic LC 5CB, the measurements in Fig. 3(b) show that the emitted signal is predominantly polarized along the x-axis. The refractive indices of the LC are \(n_e(\omega)=1.6744\), \(n_o(\omega)=1.5126\), \(n_b(3\omega)=1.5959\) [9], with \(e\) and \(o\) denoting the extraordinary and ordinary axes, respectively. Since the LC molecules align along the rubbing direction, the y-direction is the extraordinary (or slow) axis and the x-axis is the ordinary or fast axis. Since...
both the fundamental and the third-harmonic are far from molecular resonances, Kleinmann’s symmetry can be used and there are only a few non-zero \( \chi^{(3)} \)-tensor elements. While a type I process (eee→o) is not possible since \( \chi_{oxx}^{(3)} = 0 \) [9], a type II process (eeo→o, or in our notation yyy→x) can result from the non-zero tensor elements \( \chi_{xyz}^{(3)} = \chi_{xzy}^{(3)} = \chi_{xyy}^{(3)} = \chi_{yyx}^{(3)} = \chi_{yxy}^{(3)} = \chi_{xyy}^{(3)}. \) Since for type II phase matching there is a positive phase mismatch in 5CB of \( \Delta k = 0.436 \text{ rad/\mu m} \), the observed THG signal from the bulk of the LC medium is attributed to a type II process, consistent with earlier work [9]. The fitted curve in Fig. 3 shows that the angle dependent data is indeed consistent with a type II phase matching process. Here the signal generation is given by the product of projections of the laser polarisation onto the e, e and o axis, i.e. a \( \cos^2(\phi)\cos^2(\phi)\sin^2(\phi) \) dependence.

### 3.2 THG imaging of topological defects

Now that the nature of the THG process in the LC is understood, we turn our attention to THG imaging of the LC in the vicinity of the spheres. Figure 4 shows two THG optical sections, for two input polarization conditions \( \phi = 0 \) [Fig. 4(a)] and \( \phi = \pi/2 \) [Fig. 4(b)]. These images are obtained from xy scans in the bulk of the LC near the equatorial plane of the polystyrene sphere. The imaged sphere is attached to the first cover glass along the beam path, and the images are thus taken at a depth of \( \sim 3 \mu \text{m} \) in to the LC medium. By imaging other beads attached to the second cover glass and from transmission images at the fundamental wavelength, we verified that refraction of the fundamental beam does not significantly affect the result of the measurements [see e.g. Fig. 5(b)]. One of the most striking features of the images in Fig. 4 is the THG signal from the four regions around the sphere with quadrant symmetry. For the polarization conditions used here, no THG signal from an undistorted LC is expected, as indeed observed far away from the sphere. Thus, the sphere does induce a local reorientation of the LC molecules that shows up in the THG image.

A number of topological defects in nematic LCs can result from different types of anchoring of the LC molecules on to the surface of spherical particles [4]. The topological defects result from the interplay of LC-sphere anchoring forces and the elastic interactions

![Fig. 3. THG intensity from within the LC medium as a function of (a) the angle between the input polarization and the LC director and (b) the angle between the analyzer and the LC director. In (b) two different excitation polarization states have been used: linear (red) with \( \phi = 0.22\pi \) and circular (blue). The error bars correspond to the standard deviation of five intensity measurements. The solid lines are least squares fits of the experimental data to theoretical expressions for (a) the signal generation in a type II phase matched THG process; and (b) a detected polarized signal through an analyzer.](image-url)
between the LC molecules. For the planar anchoring that is favored by the LC molecules at the surface of polystyrene spheres, the main type of topological defect is known as 'Boojum' [4].

The non-zero THG signals from the bulk of the LC near the sphere observed in Fig. 4 can be understood by considering a 'Boojum' topological defect. Within the quadrant, the planar anchoring of the LC molecules to the sphere result in a local reorientation away from the y-axis. This results in a THG signal in two different ways. First, because of the local angle between the LC director and the laser polarization, a quasi phase matched THG signal is obtained as explained in Fig. 3(a). Second, the local ordering yields a local inhomogeneity, i.e., a local change in effective $\chi^{(3)}$ and refractive index. The drop in intensity at the "north" and "south" pole of the sphere is consistent with the dominant alignment of the molecules along the y-axis. The fact these polar nodes are slightly tilted with respect to the y-axis indicates a small off-set between the experimental polarization and LC coordinate systems. This also explains the difference in intensities that is observed among the different quadrants and the fact that this difference between the different quadrant reverses when the polarisation is rotated by $\pi/2$ [see Figs. 4(a) and 4(b)].

![THG intensity images](image1.png)

**Fig. 4.** THG optical sections of a polystyrene bead in a nematic LC medium near the equatorial plane. The input polarization is $\phi = 0$ for (a) and $\phi = \pi/2$ for (b).

The THG signal generation process in the LC becomes even more complex when considering the propagation of the electric fields through the LC. An example of this is shown in Fig. 5. Figure 5(a) shows a yz THG image (at a fixed x coordinate) of the LC in the vicinity of a sphere. The input polarization is perpendicular to the rubbing direction ($\phi = \pi/2$). For this orientation a THG signal is expected only in the vicinity of the equatorial plane of the sphere as a result of the planar anchoring of the LC molecules. However, apart from the signal from the glass/LC and sphere/LC interfaces, there is also significant THG in areas of the LC bulk extending well beyond the equatorial plane. This does not, however, necessarily imply that the defects are present also at those remote locations. Indeed, a local defect in the LC orientation close to the bead will induce a change in the polarization of the input beam when it propagates through the defect. Once the input polarization has been turned away from the y-axis, again a THG signal from the bulk is observed [cf. Fig. 3(a)].

Figures 5(b) and 5(c) demonstrate that the topological defect is indeed localized to the sphere only. Figure 5(b) is an xy image in a plane coinciding with the equatorial plane of a bead attached to the second (LC/glass) interface. With the input polarization at $\phi = \pi/2$ this...
image is essentially identical to that of Fig. 4(b). Figure 5(c) is an xy image of a plane only 1 μm above the equatorial plane of Fig. 5(b). The signal has almost completely disappeared demonstrating the confinement of the topological defect to the bead only.

An additional complication of the THG image formation in a LC is the fact that the refractive indices are different between the ordinary and extraordinary axes of the LC. While the magnitude of $n_o(\omega)$ is relatively close to the refractive index of the cover glass, that of $n_e(\omega)$ is significantly larger. This gives rise to substantial aberrations along the y-direction as a result of the refractive index mismatch between the immersion medium (oil) and the LC [20]. When the beam propagates deeper into the sample, its focus is astigmatically distorted. That is why Fig. 5(b) is taken with $\phi=\pi/2$ input polarization, since the - much larger - aberrations for $\phi=0$ significantly distort the image.

Fig. 5. (a). yz-image of a polystyrene sphere attached to the first cover glass of a nematic LC cell, (b) and (c) xy images in planes respectively coinciding with and 1 μm above the equatorial plane of a sphere attached to the second cover glass. For all the three cases the input polarization is $\phi=\pi/2$.

Fig. 6. yz-image of a polystyrene sphere attached to the first cover glass of a nematic LC cell. The input polarization is $\phi=0$. The intensity at the focal plane is above the threshold level as pointed out in Fig. 2.
Figure 6 provides additional credit to the explanation of THG imaging of the topological defects in the LC as outlined above. It represents a yz image acquired at an intensity above the threshold level as discussed in connection with Fig. 2. The input polarization is set at $\phi=0$. For this polarization setting there is no torque on the LC molecules and therefore no laser induced reorientation which would result in the oscillatory behavior shown in Fig. 2(a). However, the topological defect at the equatorial plane of the sphere consists of reoriented LC molecules, which induce a polarization change of the input laser field. This results in an angle - and therefore torque - between the laser polarization and the LC director further inside the LC, where there is no influence of the sphere. Consequently, laser induced oscillations in the THG image are observed below the topological defect induced by the sphere.

4. Conclusion

In summary, we have explored the nature of THG from a nematic LC using a low intensity, tightly focused beam. We have determined the threshold for laser-induced reorientation of the LC molecules and found that below this threshold THG from the bulk of the LC medium results from a type II phase matching. Using three-dimensional THG microscopy a ‘Boojum’ topological defect induced by a colloidal particle in a nematic LC has been visualised. THG imaging can provide us with detailed information about these topological defects in LCs without using any labeling. In contrast to conventional polarization and phase contrast microscopy, THG provides inherent optical sectioning which permits detailed localization of the topological defect. At the same time, we also note that the interpretation of THG images from these ordered systems is complicated by both laser propagation through the medium, which can influence the polarization state, and by anisotropic refractive index mismatch induced aberrations.

Acknowledgments

This work was financially supported by the Stichting Technische Wetenschappen (STW), The Netherlands, under grant no. ABI. 4929. M. Oh-e would like to acknowledge the support by Hitachi Displays, Ltd.