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Letter

Harmonic generation in a nematic liquid crystal

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Abstract

The nonlinear optical properties of a nematic liquid crystal (NLC 1289) were experimentally studied according to the intensity and polarization of the pump radiation at a wavelength of 1.56 μm. It is shown that the maximum generation efficiencies of the second and third harmonics correspond to different polarization characteristics of the main radiance. In the investigated range ($10^6 \ldots 2 \cdot 10^7$ W cm^{-2}) the nonmonotonic dependence of the second harmonic intensity on the pump radiance power was demonstrated for the first time. In addition to this, explanation of this effect is also proposed.

Keywords: nematic liquid crystal, harmonic generation, nonlinear optical properties

(Some figures may appear in colour only in the online journal)

1. Introduction

After the 80 s of the last century, a rather large number of papers have been devoted to the issue of harmonic generation (HG) in liquid crystals (LC) (see, for example [1–5]). They can be divided into several directions: generation of even and odd harmonics, surface or bulk generation. The most controversial question formulated in [1] is the possibility of bulk generation of even harmonics in LCD media with central symmetry. In such media, the generation of the second HG (SHG) is impossible. In particular, an undeformed (homogeneous) nematic liquid crystals (NLC) in bulk does not have centers of inversion. However, LC is the so-called soft media. Their structure together with the symmetry in the bulk can easily change under the influence of external factors unlike to solid crystals. These factors also include influences that can be combined with one term—self-action, which was first demonstrated in [2]. In laser irradiation, self-actions [2, 6–9], which deform the NLC and change the symmetry in the bulk, may include the light-induced Fredericks transition [10, 11], thermo-mechanical [12], and thermo-orientation phenomena [13]. As a result, the generation of even harmonics seems to be divided into two consecutive stages: first, bulk deformation occurs, as a rule, leading to changes in the uniform direction of the optical axis (director) of the NLC in space and removing central symmetry, then nonlinear generation, as in solid crystals, occurs even harmonics. In particular, the generation thresholds are determined by the thresholds for the onset of deformation of the LC structure.

Nonlinear optical phenomena in liquid crystals which are associated with the bulk conversion of the radiance frequency (harmonic generation, sum-frequency and difference-frequency generation) are phenomena that may eventually be of great applied value [6, 7, 14]. The features of the frequency conversion in LC mostly include the high efficiency of the process, which can reach several percent [7–9] at thicknesses of 10–100 microns. Such efficiencies, combined with small sizes, allow to create fairly simple and small-sized converters or optical parametric generators [9] to expand the frequency range of laser systems with relatively small (~100 mW) average power.

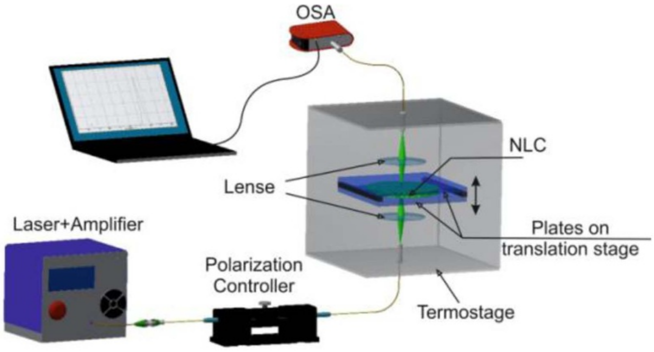

Figure 1. Scheme of experimental setup.

In the previous works [8, 9], a drop sample of a nematic LC (NLC) has been examined. It was applied directly to the end face of an fiber optic, so that it became possible to obtain the maximum power density of the pumped radiance for the laser systems. However, the simplicity of the installation did not allow changing some of the radiance parameters. Although these parameters were necessary to obtain additional, untypical characteristics of solid nonlinear crystals of the bulk generation of harmonics in the LC.

In the most of works [1, 3–5, 14] on the GH, it is mainly either about LC with broken central symmetry (smectics, external fields), or about surface effects where inversion centers may initially be present. In all these works, it is assumed that the optical axis of the LC is rigidly fixed, which is fundamentally different from our research [7–9].

The present work is devoted to a studying of the second and the third harmonics bulk generation in a nematic liquid crystal under the influence of picosecond pulsed laser radiation focused by the lens into the sample. The first systematic results are presented that determine the energy and polarization parameters of the generation of the second and third harmonics in an NLC with an initial centrally symmetric structure.

2. The experiment

We used a nematic liquid crystal (a nematic mixture of NLC 1289; NIOPIK, Russia) in our experiments. The temperature range of the existence of the mesophase: $-20 \ldots + 60$ ºC. To prepare a sample of the nematic in the isotropic phase, it was injected by a capillary method between two flat polished glass substrates, which were calibrated with respect to each other at a distance of 100 μm. The initial orientation of the director with a thorough cleaning of the glasses was homeotropic when the optical axis is normal to the surface. The uniformity of the NLC structure in the sample was monitored by a polarizing microscope. During the experiment, a thermal control system was used so that the temperature of the substrates was constantly kept ($T_c = 25$ °C). The experimental setup is shown in figure 1.

In our experiment, we used a master oscillator power amplifier (MOPA) laser system with a wavelength of $\lambda_p \sim 1560$ nm. The pulse repetition rate was 15.6 MHz, the duration was 50 ps, the average radiance power was about 400 mW, the peak power was about 480 W, and the pulse energy was about 25 nJ. A fiber polarization controller was located at the output of the laser system to control the polarization state of the radiance. In the installation a standard SMF-26e fiber with a core diameter $w_0 = 8.9$ μm was used.

The output end of the fiber of the polarization controller formed a diverging conical beam with a Gauss-shape profile. It should be noted that the generation of the second ($\lambda_p/2 = 780$ nm) harmonic due to the intrinsic nonlinearity of the pump laser was on average no more than 20–30 pW, while the third harmonic was not observed. The output radiation directed normal to the substrates with the sample was focused into the sample with a lens with a focal length of 25 mm. During the experiment, the focal waist size ($d = 55$ μm) was determined using the ‘sharp edge’ method [15]. The spectrometer QEPro Ocean Optics and the power meter HIOKI 3664 were as the apparatus used.

In previous papers [8, 9], an NLC droplet sample was located directly at the end of the fiber optics (the diameter of core is 8.9 μm). This made it possible to obtain higher power densities of the pump radiation to achieve high (∼several percent) conversion factors to harmonics. In the present work, while radiation was removed from the fiber and then focused, the power densities were ∼38 times lower, because the size of the focal waist was 6.2 times larger than the diameter of the fiber core. However, due to the assignment of the sample with NLC from the end of the fiber, this experimental scheme allowed to measure the state of polarization of the incident radiation without violating the NLC sample. The polarization state was measured by the Polarizing Beamsplitting Cubes. In order to get this done, a sample with an NLC was replaced by a polarizer. In this paper, we determine the main characteristics of the generation of the SHG and the third harmonic (THG), including the polarization of the pump radiation to achieve maximum generation efficiency.

3. Results and discussions

In figure 2 you can see the typical form of the spectrum obtained at the output of radiation from a sample with the initial homeotropic orientation of the NLC. As it clear from the graph (figure 2(a)), simultaneous generation of both harmonics is observed, while their amplitudes substantially depend on the polarization state ($\lambda_p \sim 1560$ nm) of the pump laser radiation. Different polarizations corresponded to the amplitudes of harmonics, which differed as much as possible from each other by an order of magnitude. Figure 2(b) shows the spectrum of the second harmonic generation by different pump power. The intensity of SGH increased with increasing of the pump power. Experientially, the intensity of the SGH was always higher than the third harmonic generation one according to the level of the maximum pump power used by the laser system (no more than 400 mW).

The divergence angle of the irradiation for both harmonics increased with the pump power, and reached ∼15° (at the level of 0.8) for the maximum value. The divergence is obligated to the self-focusing of irradiation in the NLC on a nonlinear

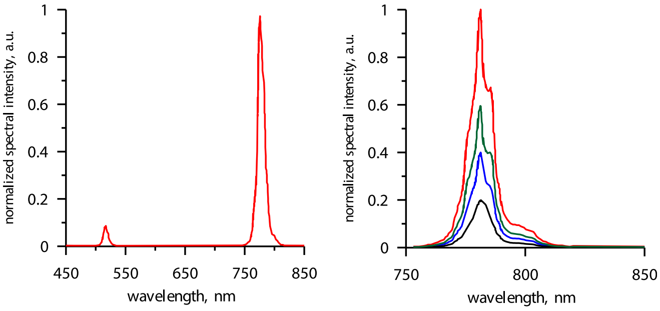

Figure 2. The typical form of the spectrum with simultaneous THG and SHG in an NLC 1289 sample 100 μm thick with a pump power of 270 mW (a); SHG spectrum (b): black curve with a pump power of 150 mW, blue is of180 mW, green is of 220 mW, red is of 370 mW.

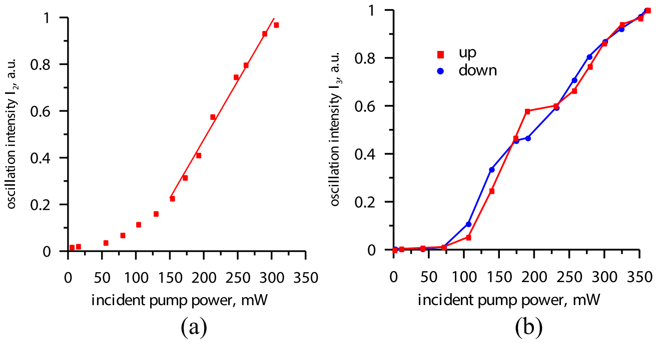

Figure 3. Dependence of the intensity of SHG (a) and THG (b) in an NLC on the pump radiation power. In figure (b), the red curve is obtained with increasing pump power, the blue one with decreasing.

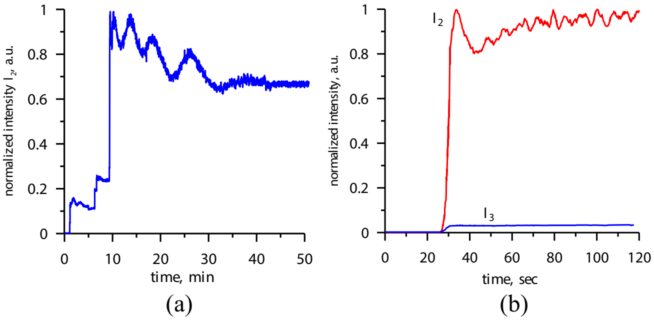

Figure 4. The dependence of the intensity of the second harmonic on time (a) after switching on the pump radiation. The power of 180 mW was included in a sequential increase in three stages (corresponding to three jumps on the graph): 125, 150 and 180 mW. The intensity of SHG (red curve) and GTG (blue curve) versus time (b) at a pump power of 220 mW.

lens, which arises due to orientation phenomena in the region of pump irradiation.

The dependence of the SHG intensity on the pump power is shown in figure 3(a). To obtain this result, the polarization state of the pump radiation was constantly tuned to the maximum intensity of the second harmonic. The SGH intensity value was determined by integration in the range of $\lambda = 740$–820 nm, see figure 2(b). The curve (figure 3(a)) does not have a threshold, however, such threshold must exist. The reason is in measuring SHG under other conditions [7, 9], when the threshold always existed and was at the level of 5–10 mW. Switching on or having a sharp change in the pump irradiation power, the harmonic intensity curve as a function of time may have a certain establishment mode (figure 4(a)) or behave non-stationary (figure 4(b)). The settling time was significant over a wide range from tens of seconds to tens of minutes. The SHG intensities for the graph (in figure 3(a)) were taken according to the average value. In the case of an unsteady mode of harmonic generation, the fluctuation times did not exceed ~5 s. Long-term (>5 min) measurements were carried out taking into account the possible instability of the laser pump system, which did not exceed 3% for an hour.

The observed temporal characteristics of the establishment and oscillations are explained by the instability of the NLC orientational structure in electric fields of elliptical polarization radiation. In this case ordinary and extraordinary waves can interact simultaneously and nonlinearly. They interact in dynamics [10], thermo-orientation [8, 13], and thermal processes that are inevitable for localization of radiation even when the sample is thermostated. The characteristic times of establishment and oscillations of harmonic intensities are close to magnitude to the accompanying orientational and thermal processes [8, 10, 13]. Non-stationary regimes SHG appeared at pump powers greater than 120 mW and depended on the pump polarization. In certain settings of the pump polarization, it was possible to remove fluctuations in power during the generation of the second harmonic, but its intensity became lower.

The dependence of the THG intensity on the pump power is shown in figure 3(b). To obtain it, the peak region of the third harmonic was integrated (figure 2(b)) in the wavelength range from 500 nm to 540 nm. As in the case of a similar SHG dependence (figure 3(a)), the polarization of the pump radiation was constantly adjusted to the maximum intensity of the third harmonic. The maximums of the SHG and THG amplitudes are reached for different polarization states of the pump radiation. Accordingly, the combination of the dependences shown in figures 3(a) and 3(b) on the same graph would not be completely correct. On average, the maxima of the harmonics amplitudes correlate no less than ~1: 10.

The temporal regimes of establishing THG were less than for SHG. They did not exceed 3–4 min, and no oscillations were observed. A record of the entire process of measuring the intensity of THG over time is shown in figure 5. In order to reveal the hysteresis properties of the generation, the pump power was sequentially stepwise increased at first and then decreased with the same stepwise. As is evident, the THG unlike the SHG has a noticeable hysteresis and a pronounced threshold, which under the experimental conditions was at the level of 70 mW and corresponded to a peak power in the sample region of $3.5 \cdot 10^6$ W cm^{-2}.

Measurements of the polarization state of the pumping radiation where the maximum generation is ensured, showed the following result. In both cases (maximum SHG and THG), the polarization was elliptical. For SHG, the average ellipse ratio was ~1: 5; for THG, this value was ~1: 4. The angle between

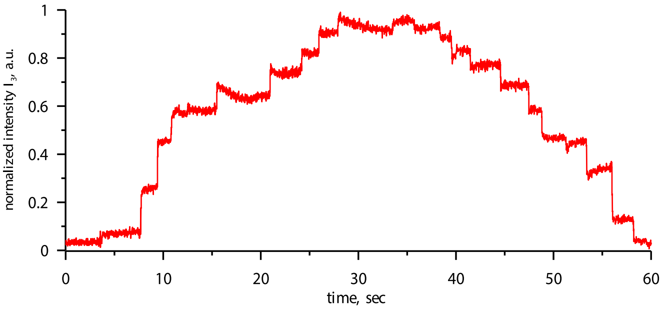

Figure 5. Time dependence of the THG intensity with a sequential stepwise increase and decrease in the pump power.

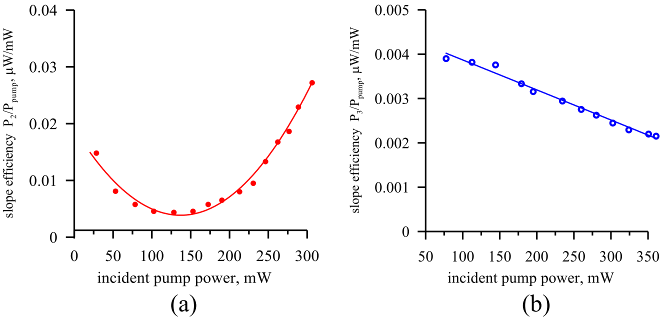

Figure 6. Dependence of the generation efficiency of the second (a) and third (b) harmonics on the pump radiation power.

the directions of the semimajor axes of the polarization ellipses for SHG and GTG was 40–50º. Deviations from the average ratio of the axes of the polarization ellipse and the direction of the major axis were of the order of ±10%. This depended on a point on the plane of the sample and (in a less degree) on the radiation power. In our opinion, the available scatter of the measured parameters was primarily determined by the initial heterogeneity of the orientation of the NLC director. The ellipticity of polarization, which ensures the maximum intensity of the harmonic generation, is determined by the competition between two types of deformation of the NLC structure. It arises when the electric field of the pumping wave and the heat flux are simultaneously absorbed. The electric field, due to the Fredericks light-induced effect [10, 11], causes the optical axis of the NLC (director) to rotate in the direction of polarization. The heat flux [13] does not depend on polarization and tends to form a radial (perpendicular to the direction of the pump wave) director distribution. As a result, non-linear polarization is optimal.

For absolute measurement of harmonic generation power, radiance was filtered and the corresponding harmonic was extracted. The absorption and scattering losses were measured tentatively in the used optical system and in the sample itself. Figure 6(a) shows the value of the SHG power related to the pump radiation power taking into account losses. As can be seen from the graph, this dependence passes through a minimum, while its value does not exceed ~$3 \cdot 10^{-5}$. This level is quite low and is a consequence of the conditions of the presented experiment. SHG efficiencies achieved in [8] is not possible here. The minimum on the SHG curve is explained by the influence of several competing factors in the formation of harmonic radiation. As far as the authors of the article know, the nonmonotonic dependence of SHG in the NLC (figure 6(a)) was discovered for the first time. A similar dependence for GTG is shown in figure 6(b).

Unlike SHG, the THG efficiency in the same range of the pump power density ($3.5 \cdot 10^6 \ldots 2 \cdot 10^7$ W cm^{-2}) monotonously decreases. Even at the maximum pump power under the experimental conditions does not reach its minimum, as in the case of second harmonic generation. However the THG magnitude is smaller than the corresponding dependence for the second harmonic (see figure 6(a)). The minimum and then the increase in the THG curve (figure 6(b)) should formally be realized due to the cubic growth of the third harmonic with increasing pump power density. But we cannot claim it with certainty because other factors may arise that have no analogues in solid crystals with a simple cubic dependence of nonlinear polarization on electric fields.

In our opinion, the SHG minimum is due to concomitant photoinduced orientational processes that locally change the direction of the NLC structure optical axis. These processes are explained below in the discussion. Let us remark here that the minimum was not found for similar dependences (figure 6(a)) in inorganic crystals [16] and, apparently, is a special property for optical nonlinearities in the so-called 'soft' media.

We have to add that a model for a complete description of nonlinear radiation generation in liquid crystals (the most studied 'soft' medium) at combined frequencies, which is a rather complicated combination of Maxwell's nonlinear 3D equations and material LC equations, has not yet been developed. The mechanism of harmonic generation in soft media and firstly in liquid crystals is more complicated than in solid crystalline materials. Accordingly, the character of generation can have its own characteristics. In addition to electronic nonlinearity, its formation can be affected by many processes that are inevitable due to the softness of the medium. First of all, NLC in a homogeneous orientation state is a centrally symmetric medium so it is not possible without the violation of the homogeneity of SHG. The source of an inhomogeneous violation of the structure or deformation with the subsequent effect on the generation itself can be the light-induced Frederiks reorientation [10, 11], thermo-mechanical [12] and the thermo-orientation mechanism [13], leading to the formation of disclination [9]. Additionally, due to self-focusing of the pumping radiation, stable radiation propagation channels [17, 18] and orientational structures forming optical vortexes [19] can be formed and also affect the generation of lasing. Many processes can be accompanied by hydrodynamic fluid movements [12, 20]. The same phenomena (except for symmetry breaking) inevitably affect the THG. Individually, all these processes can be described by the corresponding mathematical models, except for the process of birth-annihilation of disclinations and their structural transitions with a change in the force of disclination.

In the processes of harmonic generation, great attention is always paid to the phase-matching conditions, which are well developed for the nonlinearity of solid crystals. For NLCs, on the one hand, this is a rather complicated question, since in the liquid crystal medium the direction of the optical axis ‘floats’ depending on many factors and, first of all, on the intensity and polarization of the pumping radiation. The director tends to orient along the polarization of the incident radiation. Thermo-mechanical influences try to orient the director along the hydrodynamic flow. Thermo-orientation processes orient the director along the heat flux. All effects are of the same order and compete with elastic forces that tend to establish the initial state. In this work, an extremely simple geometry was chosen in order to maximally isolate the polarization features of harmonic generation in a soft LCD medium.

On the other hand, the dispersion of the refractive indices at the pump and generation frequencies is small. So the phases shift of the pump and harmonics radiation at the LC thicknesses (10–100 μm) are small and it would seem that the synchronism conditions in its classical sense can be ignored. Nevertheless, the observed dependence of the generation on the pump polarization is quite pronounced. In the selected simplest experimental design, the initial direction of the optical axis coincides with the direction of all waves propagation that have only an unusual appearance. Furthermore, the non-uniform deviation of the optical axis with exit from the plane of incidence will cause the appearance of ordinary waves in the pump itself (as well as in harmonics) which interact differently with the NLC [10]. The indices n_e and n_o significantly (0.05–0.1) differ from each other. Such a process has no analogues in solid crystals. This work is devoted to the study of its mechanism characteristic of NLC. Formally, the studied polarization phenomena can also be attributed to some more complicated concept of synchronism, if synchronism is defined as the possibility of coherent addition and obtaining the maximum of the generated waves.

According to our preliminary data [8], the quadrupole mechanism of nonlinearity formation plays an important role in the generation process in LCs, which is especially noticeable near phase transitions. Taking this into account, it is necessary to take into account not only the director of the NLC and the electric field of the wave, but also their gradients which further complicates the mechanism of bulk nonlinear generation.

In our experiments was not the goal to obtaining maximum energy characteristics. The main idea of this work is research aimed at revealing the features of SHG and THG in NLC, which have no analogues in solid crystals and are necessary for a deeper understanding of the characteristics of the conversion of the radiation frequency in soft media. To the best of our acknowledgments, such studies were conducted for the first time.

4. Conclusion

In this we presented experimental studies of the nonlinear optical properties of a nematic liquid crystal (NLC 1289) depending on the intensity and polarization of the pump radiation at a wavelength of 1.56 μm. In the experiments with NLCs, polarization states were revealed at the generation maxima of the second and third harmonics are observed. It has been established that these states are different for SHG and THG and have an elliptical structure that differs markedly from linear polarizations for similar conditions in solid crystals. Also in the studied range ($10^6 \ldots 2 \cdot 10^7$ W cm^{-2}) non-monotonic dependence of the intensities of the second harmonic on the pump radiation power in the NLC (possibly in other soft media) were demonstrated for the first time. The parameters of stationary and non-stationary modes of second and third harmonic generation are determined. With certain settings of the pump polarization, it was possible to completely eliminate power fluctuations in the generation of harmonics.

The results obtained in this paper are necessary for further study of the features of optical nonlinearities of liquid crystals, including the construction of a satisfactory mathematical model for the interaction of radiation with LCs and the development of nonlinear optical devices based on them.

Acknowledgments

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