Phase-Matched Third-Harmonic Generation in a Nematic Liquid Crystal Cell

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We show the generation of a strong phase-matched third harmonic in a simple parallel nematic liquid crystal cell. A single laser beam of femtosecond pulses is used to deform the internal structure of the molecular orientation distribution, and to generate the third harmonic from the twisted structure that is formed. Up to 10^{-5} of the laser power was converted to the third harmonic, in a volume as small as 5 μm^3. [S0031-9007(99)08859-6]

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Liquid crystals (LCs) are known for their strong nonlinear optical properties. A large contribution to the nonlinearity originates from molecular reorientation, an effect that has been used to demonstrate many interesting optical phenomena, such as [1] wave mixing, self-phase modulation, self-focusing, optical bistability, and optical Fréedericksz transitions. This nonlinear mechanism is effective even at relatively low optical powers, but is also relatively slow. Much faster is the electronic contribution to the nonlinearity of LCs, which leads to effects such as second and third harmonic generation. Second harmonic generation in most liquid crystals is only an interface effect because of symmetry requirements. Efficient third harmonic generation (THG) is difficult to achieve in the LC bulk because of phase mismatch. However, in cholesteric liquid crystals, phase matching can be achieved using the properties of the periodic medium [2].

In this paper, we present an observation of strong phase-matched THG from within a simple nematic LC cell which is one of the most widely used types of LCs. The nematic LC is composed of rodlike molecules with one molecular axis much longer than the other two. The long axis of the molecules tends to point along a common direction, the director, while they are free to move from one location to another. The phase-matched THG we present here resulted from the combination of two nonlinear processes of different origin. A tightly focused laser beam of femtosecond pulses is used, which slightly modifies the molecular orientation inside the LC cell, leading to the generation of phase-matched third harmonic from the focal point. The same laser beam that deforms the local molecular orientation is used to generate the third harmonic light.

The experimental setup used to generate third harmonic light from a nematic LC is similar to the setup used in THG microscopy [3]. The laser source is a synchronously pumped optical parametric oscillator (Spectra-Physics Tzunami-Opal system) which provides 130 fs pulses at a wavelength of 1.5 μm with a repetition rate of 80 MHz. The laser beam was focused onto a LC cell at normal incidence, using $\times 60$, numerical aperture $= 0.85$ microscope objective, to a spot size of $\sim 1\,\mu$m. The sample used for the experiment was a parallel-aligned E7 nematic LC with a thickness of 50 μm. The LC was held between two glass substrates. A rubbing material with a low pretilt angle was used (PI2555), to impose a high anchoring potential on the LC molecules at the cell surfaces. The depth of the focal point in the sample is controlled using a piezoelectric driven stage. The third-harmonic light at 0.5 μm is collected by a lens and measured by a photomultiplier after filtering out the fundamental wavelength. It was verified by spectral measurements that only third harmonic light was generated, while no second harmonics signal could be detected. The polarization of the input beam is controlled by a polarizer, $\lambda/4$ and $\lambda/2$ plates. The coordinate frame is chosen so that the top glass-LC interface is in the x-y plane while the director of the LC at this plane is pointed at the y axis. The electromagnetic field is therefore propagating along the positive z axis. Tightly focusing a Gaussian beam in a homogenous nonlinear medium usually results in no THG [4]. However, when the medium is not homogeneous or, more specifically, when there are variations of the nonlinear third-order susceptibility $\chi^{(3)}$ in the bulk, third harmonic light is generated [5].

Shown in Fig. 1 are images composed of sets of 91 line scans of the focal point along the depth of the sample (z axis) at a constant x-y point, depicting the polarization and depth dependence of the THG intensity. In the first and last scans in each set, the polarization is along the x axis. The angle θ between the optical field and the rubbing direction (y axis) increases by 2° from line to line, thereby spanning 180° rotation in each set.

In Fig. 1A the average laser intensity was 100 mW. As expected, the THG distribution is symmetric around $\theta = 0°$, and strong third-harmonic light is generated when the focal point is near the two LC-glass interfaces, at $z = 0$ and $z = 50$ μm. The THG intensity at these interfaces is proportional to the difference between the corresponding $\chi^{(3)}$s of the glass and the LC [3]. The THG from the

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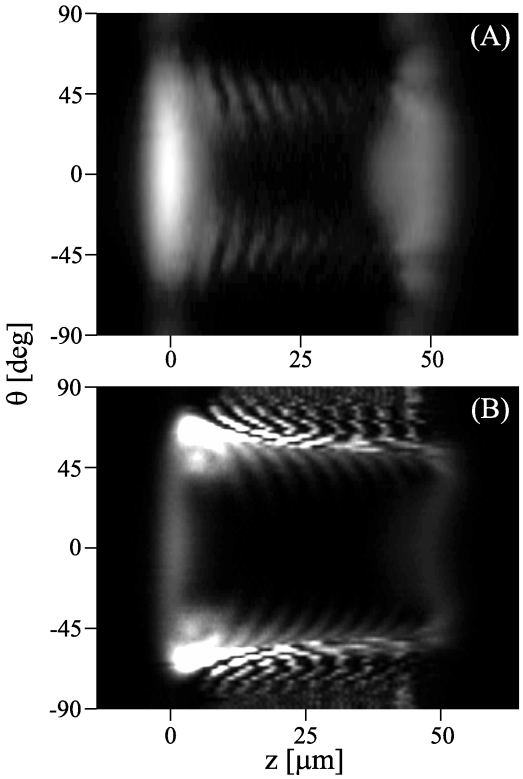

FIG. 1. THG distribution in a nematic LC cell for different linear polarization states with average laser intensity of (A). 100 mW, and (B) 200 mW. Bright gray levels represent high THG intensities. The THG from within the LC cell has an efficiency of up to 10^{-5}.

left interface ($z = 0$) is weak at $\theta = \pm 90°$, and becomes strong around $\theta = 0°$, where the direction of the field is parallel to the director of the LC. These measurements indicate that

$$\chi^{\mathrm{LC}}_{yyyy} \simeq 8\chi^{\mathrm{glass}}_{1111} \tag{1}$$

and

$$\chi^{\mathrm{LC}}_{xxxx} \simeq \chi^{\mathrm{glass}}_{1111}\,, \tag{2}$$

where $\chi^{\mathrm{LC}}_{xxxx}$ and $\chi^{\mathrm{LC}}_{yyyy}$ are the third order susceptibility tensor components of the LC along the x and the y directions, respectively, and $\chi^{\mathrm{glass}}_{1111}$ is the relevant tensor component of the glass substrate, where the index 1 may stand for any direction. The THG from the right interface ($z = 50$) is weaker and broader due to aberrations of the focal point as it is affected by the high refractive index of the LC, especially when the light is polarized parallel to the extraordinary molecular axis, around $\theta = 0°$.

In addition to the THG from the two interfaces, Fig. 1A shows that some third-harmonic light is generated within the LC volume. A structure of periodic THG peaks is visible near $\theta = \pm 45°$. When the laser intensity was increased to an average power of 200 mW, a more complex structure of THG distribution has been obtained, as shown in Fig. 1B. In addition to the relatively weak THG peaks of Fig. 1A, a series of intense THG peaks appear now at $\theta = \pm 60°$. The strongest peak at $z = 5\ \mu$m has an averaged intensity of about 2 μW, 2 orders of magnitude higher than the THG from the interfaces. The conversion efficiency in this case is 10^{-5}.

The appearance of the THG from within the cell volume indicates that the LC is no longer homogenous. Indeed, a structure of molecular orientation is expected to be formed within the cell due to the combined effect of the birefringence of the LC, and the force induced by the optical field. Reorientation of the molecules by an optical field results from the system's tendency to assume a configuration with the minimum free energy [6], which consists of the distortion energy and the dipolar interaction energy. Near the focal point, the optical field exerts a torque on the molecules:

$$M \propto \langle E^2_{\mathrm{op}} \rangle \sin(2\beta)\,, \tag{3}$$

where $\langle E^2_{\mathrm{op}} \rangle$ is the time averaged square of the optical field and β is the angle between the field and the director of the LC molecules. This torque is maximal for $\beta = 45°$.

To gain some insight into the processes leading to the results shown above, we have calculated the molecular structure induced by a focused optical field in a LC cell. The confocal beam parameter [7] used was 2.5 μm. The input field was chosen to be polarized at 45° to the x axis, which provides the maximal torque on the molecules. The polarization state of the optical field at the focal point, which determines the direction and strength of the torque, is changing as the focal point is scanned along the z axis, with a characteristic period of the beat length, given by $L_B = \lambda/\Delta n$. Here, λ is the fundamental wavelength and Δn is the birefringence. The reorientation angle is assumed to be small.

The resulting reorientation of the LC molecules for different locations of the focal point z_f is shown in Fig. 2. The figure demonstrates that the structure induced by the beam depends on the location of the focal point, and it is reproduced with the periodicity of the beat length. The length over which molecular deformation occurs depends on the focal depth, about 5 μm in our geometry. When the focal point is at a distance $z_f =$ 14, 18, and 22 μm from the left interface, the angle α between the director and the rubbing direction (y axis) varies linearly along z at the vicinity of the focal point. As a result, a twisted structure is formed around that point. The polarization state at the focal point is calculated to be elliptic with opposite handness to that of the molecular twist. It was already shown [2] that phase-matched THG can be achieved in a cholesteric LC when the pitch of the helical structure is compensating for the phase mismatch. Using the same arguments with the parameters of our experiment, we calculated that a helical structure with a pitch of 11 μm is required for optimal phase matching. The light-LC interaction generates, therefore, these twisted

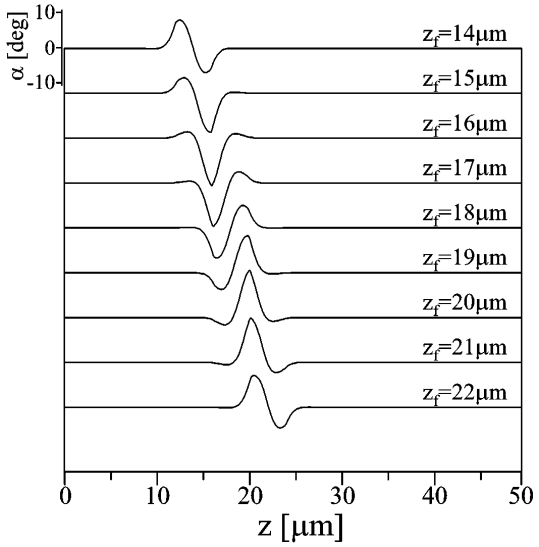

FIG. 2. The molecular reorientation induced by a focused laser beam in a LC cell. The director's angle distribution along the depth of the LC cell $\alpha(z)$ is plotted for different depths of the focal point z_f. A linear twisted structure is formed in a periodicity of 4 μm.

structures which lead to a series of THG peaks, with a periodicity of $L_B/2$ along the z axis. Between these peaks, at $z_f = 16$ and 20 μm, the structure of the molecular reorientation does not compensate for the phase mismatch, leading to zero THG. In our simulations, $L_B/2 = 4$ μm at room temperature, which is in a fair agreement with the experimental peak period of 4.2 μm, as in Fig. 1A.

The THG peaks in Fig. 1A are relatively weak compared to the THG from the interface, indicating that the phase matching is not fully achieved. The reason for this is that the induced pitch of the twisted structure is smaller than the optimal pitch. Larger values for the pitch can be achieved when the average laser power is increased. We believe that the efficient THG obtained in Fig. 1B is the result of a nearly optimal combination of twist and polarization state obtained near $\theta = \pm 60°$.

The typical characteristic time for molecular reorientation at room temperature for E7 LC material is about 50 ms. We have found that indeed the rise time of the THG from within the cell had the same characteristic time. Therefore, the rate in which the LC sample is scanned is very important. When scanning too fast the efficiency of the THG decreases, since there is no time for the molecules to reorient. Scanning too slow, on the other hand, with relatively high intensities ($I_{\mathrm{av}} > 100$ mW) leads to other nonlinear phenomena with longer characteristic time, such as optical Freédericksz transitions [1]. These transitions can support phase-matched THG, as can be noticed at $\theta = \pm 90°$ in Fig. 1B; however, this THG was found to be noisy and highly unstable.

In order to verify that the period of the THG peaks along the z axis is half the beat length, we heated the LC cell to a temperature above the critical temperature of the Isotropic-Nematic (I-N) phase transition, then let the sample cool down, across the phase transition temperature, back to room temperature. During the cooling process we scanned the sample with the focal point along the z axis, with one line scan per second at a constant linear polarization at $\theta = -60°$, and an averaged laser intensity of 50 mW. The resulting set of scans shown in Fig. 3 depicted the THG distribution inside the LC cell as the cell cools down. The phase transition I-N is clearly observed around the 10th scan, where THG first appears from within the LC cell. As expected, there is no THG from the isotropic phase at the first few scans. Right after the phase transition, the THG is most intense, since at high temperatures the elasticity is high, thus reorientation of the molecules by the optical field is large. We note that the number of THG peaks along the depth of the LC cell is continuously increasing as it cools down, indicating that the beat length is decreasing. The reason for this is the dependence of the birefringence on temperature [1]. As the temperature decreases across the critical temperature of the phase transition, birefringence is formed and increases up to $\Delta n \simeq 0.2$ at room temperature.

In summary, we have shown the generation of a strong phase-matched third harmonic from within a simple parallel nematic LC cell. A single laser beam of femtosecond pulses is used both for the deformation of the LC director distribution, and for the generation of the third harmonic from the twisted structure that is formed. Note that light-induced molecular reorientation is expected in cholesteric LCs as well. Hence, phase matching in cholesteric LCs [2] might actually be spoiled at high laser intensities. Generation of third harmonic in nematic LCs using tight focusing allows the use of high intensities per unit area,

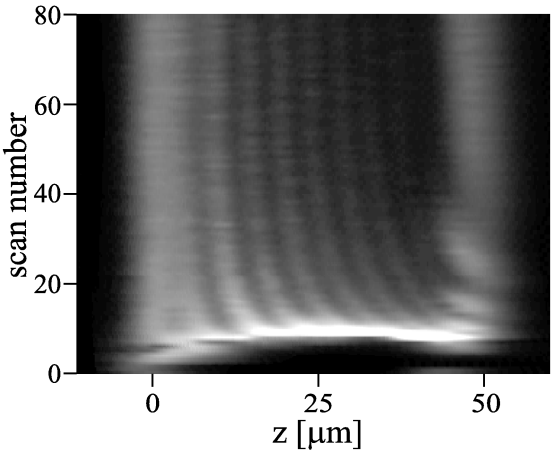

FIG. 3. THG from within a nematic LC cell during a cooling process of the cell across the isotropic-nematic transition point. The periodicity of the THG peaks along the depth of the cell is increased as the cell is cooled down, indicates the increasing birefringence.

and high conversion efficiency can thus be achieved. The attainable efficiency of the THG will be limited by damage of the LC material. At average laser intensities higher then 250 mW we observed damage to the samples, accompanied by continuum generation from the focal point. We expect that using a source of ultrashort pulses at a low repetition rate, that will allow high peak intensities with low average intensities, could significantly improve the process of phase-matched THG.

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[1] P. J. Collings and J. S. Patel, *Handbook of Liquid Crystal Research* (Oxford University Press, New York, 1997).
[2] J. W. Shelton and Y. R. Shen, Phys. Rev. A **5**, 1867 (1972).
[3] Y. Barad, H. Eisenberg, M. Horowitz, and Y. Silberberg, Appl. Phys. Lett. **70**, 922 (1997).
[4] R. Boyd, *Nonlinear Optics* (Academic, New York, 1992).
[5] T. Y. F. Tsang, Phys. Rev. A **52**, 4116 (1995).
[6] I. C. Khoo and S. T. Wu, *Optics and Nonlinear Optics of Liquid Crystals* (World Scientific, Singapore, 1993).
[7] A. Yariv, *Optical Electronics in Modern Communications* (Oxford, New York, 1997).