Quantum Electronics **38** (4) 373 – 376 (2008)

NONLINEAR OPTICAL PHENOMENA

PACS numbers: 42.70.Df; 42.65.Ky
DOI: 10.1070/QE2008v038n04ABEH013719

Highly efficient lasing at difference frequencies in a nematic liquid crystal

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***Abstract*. Highly efficient lasing at difference frequencies is obtained in a nematic liquid crystal excited by several visible lines from a cw argon laser with a total power of 0.08 – 1.5 W. The maximum conversion efficiency was $\sim 1\,\%$ and the quadratic susceptibility was $\sim 2 \times 10^{-6}$ m V^{-1}. The field of application of the approximate mechanism of quadratic nonlinearity and frequency conversion considered in the paper requires specification. The nonlinear conversion of radiation in a nematic crystal has specific features compared to lasing at difference frequencies in solid crystals.**

Keywords: *lasing at difference frequencies, orientation nonlinearity, liquid crystal.*

The nonlinear optics of liquid crystals (LCs), as one of the directions in the optics of nonlinear phenomena, appeared comparatively recently (in the late 1970s – the early 1980s). A detailed story and bibliography on this subject are presented in review [1]. The physical mechanism responsible for specific nonlinear optical phenomena observed in LCs is analysed in this review and earlier papers. Unlike solid crystals, the optical axis directed along the long axis of molecules in LCs is not rigidly fixed and can change in time and space depending on the magnitude and direction of electric fields, in particular, light fields. To distinguish the effects at which the permittivity could be considered constant for monochromatic fields, the term 'orientation optical LC nonlinearity' was introduced. This nonlinearity is usually called the cubic or, more accurately, odd-order nonlinearity. The analogy (although incomplete) between photoinduced phenomena observed in LCs and the Fredericksz reorientation in static electric fields was pointed out already in the first papers [2]. The orientation self-focusing of limited beams appearing due to induced gradients of the refractive index (nonlinear lens) in the interaction region was simultaneously investigated. The angular divergence, which exceeded the initial divergence by two orders of magnitude, and the transverse intensity distribution of a light beam propagated through a lens self-induced in a nematic liquid crystal (NLC) were studied in paper [3], where this phenomenon was observed for the first time. Note that all the orientation Fredericksz processes related to the odd-order nonlinearity are rather slow, their characteristic times being 0.1 – 1 s [1, 3].

Phenomena related to the quadratic nonlinearity of LCs or the even-order nonlinearity proved to be more complicated, especially in the case of NLCs. In the unperturbed state (the optical axis – the director – has one direction in the entire volume), many LCs, in particular, NLCs have the central symmetry, and the existence of the even-order nonlinearity is forbidden. However, as was assumed earlier [4], due to the break of the unidirectional orientation of the director caused by the Fredericksz reorientation or some other interaction, the central symmetry can be violated and the direction of the optical axis of the LC will change in space and time. Thus, if the deformed state has the uncompensated macroscopic dipole moment (polarisability in NLCs appears due to the flexoelectric effect), it becomes possible to obtain lasing at difference and sum frequencies.

The orientation mechanism of nonlinear generation of radiation considered here does not allow one to separate the influence of the even and odd powers of the expansion of induction into a Taylor series in terms of the electric field strength of the wave. In fact, we have to deal with nonlinearity of the general type. Note also that classical crystallographic concepts applied to solids are not always can be used for LCs. In this case, the hydrodynamic model of Eriksen – Lesly [2] for the anisotropic (non-Newtonian) liquid can be applied. The fulfilment of the phase-matching condition, as the condition providing coherent accumulation of radiation in the nonlinear optics of solid crystals is achieved by selecting optimal geometrical parameters, which depend on crystallographic and spectral dispersion properties of a crystal. The direction of the optical axis in a LC depends on the electric fields of the waves propagating in the medium, and therefore the phase-matching condition in the LC should be more intricate than well-known geometrical relations in the nonlinear optics of solid crystals. This problem is not studied in our paper.

The generation of harmonics of electromagnetic radiation caused by the quadratic nonlinearity of LCs were studied in a number of papers mainly in the late 1970s and the early 1980s. A detailed bibliography on this subject is presented in book [5]. The second harmonic conversion efficiency in LCs was comparable with that observed in solid

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Received 13 August 2007; revision received 9 January 2008
Kvantovaya Elektronika **38** (4) 373 – 376 (2008)
Translated by M.N. Sapozhnikov

crystals (the susceptibility was $\chi \sim 10^{-13}$ m V^{-1}). The orientation mechanism of the appearance of quadratic nonlinearity was studied in theoretical paper [4]. The susceptibilities of NLCs obtained within the framework of assumptions used in [4] considerably differed from experimental values [6, 7]. The discrepancy was explained by the absence of reliable data for some material constants of NLCs in the range of radiation frequencies.

The possibility of lasing at the difference frequency in LCs in the submillimetre (terahertz) range was considered theoretically in paper [8]. The equations based on the orientation break of the symmetry of NLCs derived in [8] were analysed numerically. The study of generation of multiple harmonics has shown that approximations used in [4] have rather limited applications and the estimates presented in [4] cannot be used in practice. One of the results obtained in [8] was the prediction of the possibility of lasing at the difference frequency with a high efficiency exceeding the lasing efficiency observed for known LCs.

In this paper, we present preliminary experimental results demonstrating the efficient lasing at the difference frequency in the NLC. The scheme of the experiment is shown in Fig. 1. The nematic liquid crystal was excited by horizontally polarised radiation from cw argon laser (*1*). The laser generated several lines presented in Table 1, where the wavelengths corresponding to difference frequencies are also indicated. The total output power of the laser was changed from 0.08 to 1.5 W by varying the discharge current. The infrared radiation of the arc in the laser tube was rejected with water filter (*2*). Laser radiation was modulated by rotating half-disc chopper (*3*) at a frequency of 1 Hz or less, which was selected in accordance with the characteristic time of the photoinduced Fredericksz reorientation. Laser radiation was focused by lens (*4*) with a focal distance of 150 mm into the LC sample in cell (*5*).

Table 1. Wavelengths corresponding to the differences of frequencies of the argon laser lines.

λ_i/μm	λ_j/μm				
	0.5145	0.5017	0.4965	0.4880	0.4765
0.5145	∞	−20.1660	−14.1916	**−9.4746**	−6.4516
0.5017	20.1660	∞	−47.9027	−17.8708	−9.4865
0.4965	14.1916	47.9027	∞	−28.5049	−11.8291
0.4880	**9.4746**	17.8708	28.5049	∞	−20.2202
0.4765	6.4516	9.4865	11.8291	20.2202	∞

Note: The wavelengths of difference frequencies falling within the recorded spectral interval are underlined. The wavelengths corresponding to the difference frequencies of the most intense argon laser lines are presented in bold.

The cell containing the NLC 1289 mixture was made of a glass plate through which the NLC was irradiated and a ZnSe plate which was transparent for IR radiation. The distance between the plates was determined by the spacer thickness (100 μm). To produce the initial homotropic orientation of the NLC director (normal to the cell walls), the cell plates were treated with chromolan. After assembling, the cell was filled by the capillary method with the LC in the isotropic state. The homogeneity of the initial (unperturbed) orientation of the director was controlled with a polarisation microscope before and after experiments. The sample surface was oriented normally to the laser beam. At a distance of 30–40 mm from the sample, where laser radiation was defocused, either germanium window (*6*) with the AR coating for the 10-μm region was placed or narrowband dispersion filters were mounted (a set of 20 filters with transmission bands in the wavelength range from 5 to 15 μm) for approximate measurements of the spectral characteristics of generated radiation (*8*). The radiation intensity was measured with calibrated FSG-22-3A photoresistor (*9*) with an ac amplifier with the lower passband boundary 0.1 Hz. Signals were detected with digital oscilloscope (*10*).

We did not use a dc amplifier in our setup because filter (*6*) was heated by laser radiation during measurements and the increasing thermal radiation of the filter caused a slow increase in the detected signal. The detector was mounted on rotation-lifting mechanism (*7*) for studying the indicatrix of radiation emitted by the sample. The integrated power of the laser and generated radiation (exceeding 5 mW) was measured with an IMO-2 power meter. The stability of the laser radiation power was controlled with photodiode (*11*). The sample was in the LC phase, which was confirmed by the characteristic aberration picture of scattering of reflected laser radiation appeared due to orientation self-focusing. When self-focusing disappeared due the transition of the LC to the isotropic phase, the experiment was terminated. The transverse intensity distribution of reflected radiation observed in experiments is similar to that obtained in [9] and suggests that the homogeneous (initial) orientation of the optical axis of the LC is violated.

Figure 2 demonstrates the time dependence of the typical output signal of the detector for the modulation frequency 0.4 Hz and the output laser power 300 mW. The minima and maxima of the curve correspond to the instants (no more than 3 ms) of opening and closing the chopper. Unlike lasing at difference frequencies in solid crystals, this curve has regions of the slow rise and decay in time. By removing a cell with water and mounting the detector in the optical axis of the laser, we controlled the frequency characteristic of the measuring channel and detected IR radiation from the discharge in the laser tube modulated by the chopper. The signal had sharp fronts of duration determined by the shutoff time of the laser beam. The signal rise for $\sim$1 s in

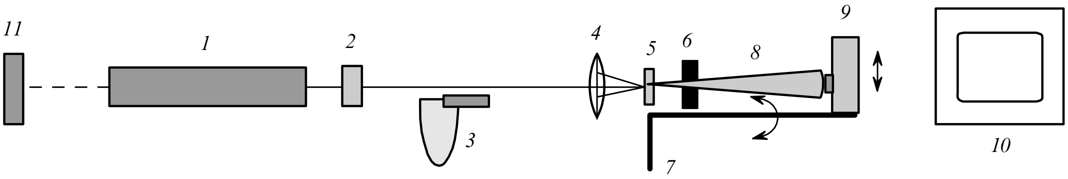

Figure 1. Scheme of the setup: (*1*) argon laser; (*2*) cell with water; (*3*) chopper; (*4*) focusing lens (f = 150 mm); (*5*) cell with the NLC; (*6*) germanium window or IR filters; (*7*) rotation-lifting mechanism; (*8*) generated radiation; (*9*) detector; (*10*) recording device (amplifier and oscilloscope); (*11*) laser power meter.

Fig. 2 is determined by the slow photoinduced Fredericksz reorientation [1, 3, 9], which breaks the central symmetry of the LC and leads to the appearance of a nonlinear lens. The presence of emission after the laser beam shutoff is not clear yet and requires further investigations. This emission can be related to some long-lived excited state existing in the medium. This assumption is based on the data obtained in paper [10] where the possibility of existing discrete quasi-stationary states of the orientation of the NLC director in alternate electric fields of complex configuration (at frequencies up to 5 MHz) was demonstrated.

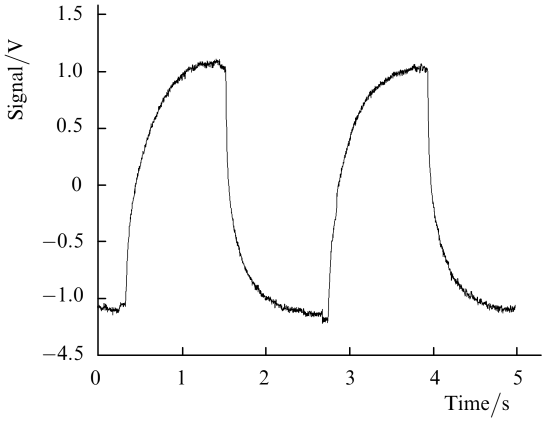

Figure 2. Time dependence of the generated radiation power for the output laser power 300 mW.

Figure 3 presents the angular intensity distribution of generated radiation. The radiation indicatrix is an ellipse with parameters depending on the laser radiation power. For the total incident radiation power $P_\Sigma = 0.3$ W, the horizontal and vertical angular dimensions of the indicatrix are 17 – 20° and 9 – 10°, respectively. The angular half-width of the indicatrix ellipse is 1 – 1.5°. The axis of the elliptic cone is displaced with respect to the laser beam axis by ~6° in the horizontal direction along the direction of the laser beam polarisation. The point of intersection of the laser beam with the figure plane (the coordinate origin in Fig. 3) coincides approximately with the left focus of the ellipse. The radiation intensity along the ellipse perimeter is not constant (changing approximately by 1.5 times) and is maximal in its lower part. Such a shape of the radiation indicatrix cannot be caused by thermal radiation or luminescence. The explanation of the angular radiation intensity distribution of the NLC sample and questions concerning phase matching are beyond the scope of our paper. The far-field intensity distribution depends at least on two factors: the presence of a nonlinear lens and diffraction of long-wavelength radiation from the transverse aperture of the interaction region in the focal waist (~50 μm) of the initial laser beam.

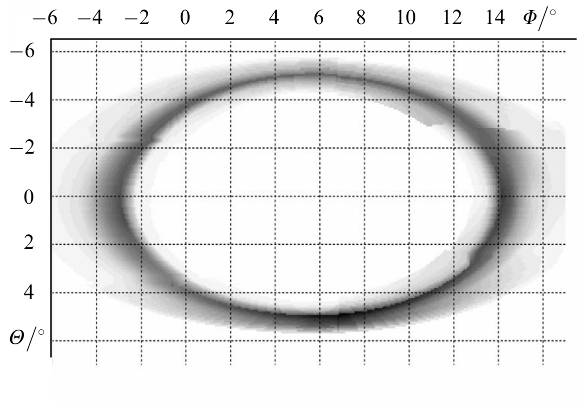

Figure 3. Angular intensity distribution of the generated radiation in coordinates Φ and Θ.

To obtain preliminary data on the spectral characteristics of generated radiation, dispersion filters were mounted in front of detector (*9*) instead of the germanium window (Fig. 1). Figure 4 shows the spectrum of generated radiation. The intensity maxima have comparable values although the intensities of the incident emission lines differ by an order of magnitude and more. This suggests that parametric amplification is possible during the formation of radiation at difference frequencies.

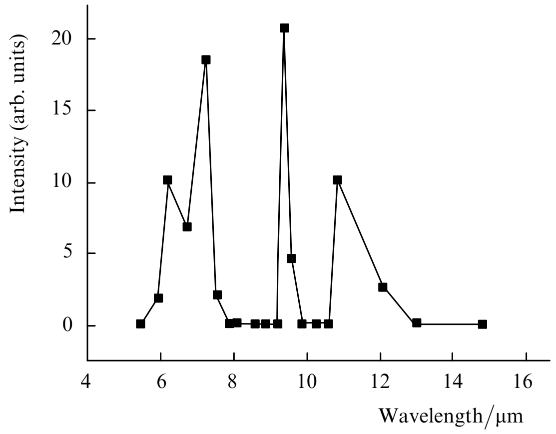

Figure 4. Spectrum of generated radiation.

Figure 5 presents the dependence of the integrated power P_Δ (over the forward solid angle and within the measured frequency interval) of generated radiation on the square P_Σ^2 of the total incident radiation power. This dependence saturates and for $P_\Sigma > 150$ mW is described with an accuracy of ~5 % by the exponential (the curve in Fig. 5). Because measurements were complicated in the power region near the coordinate origin due to a small signal-to-noise ratio, we failed to determine the threshold for lasing at

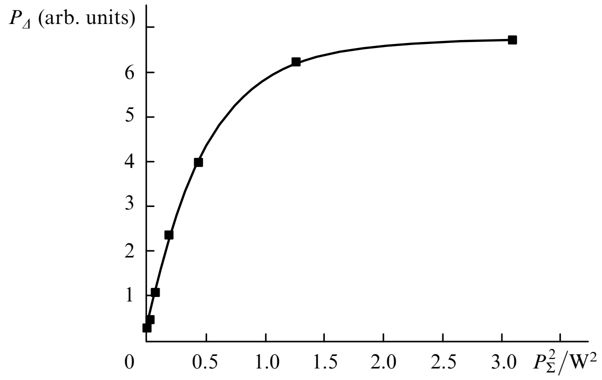

Figure 5. Dependence of the generated radiation power P_Δ on the square of the total incident radiation power P_Σ^2. Squares are experimental data, the curve is the interpolation dependence $P_\Delta = P_0[1 - \alpha\exp(-P_\Sigma^2/p_\omega^2)]$.

the difference frequency. If this threshold exists, then P_{th} does not exceed 80 mW, which does not contradict by the order of magnitude to data obtained in papers [3, 9] where the threshold characteristics of the photoinduced Fredericksz transition and a nonlinear lens were investigated. For the pump power exceeding 150 mW, the dependence in Fig. 5 is described by the relation

$$P_\Delta = P_0\left[1 - \alpha \exp\left(-\frac{P_\Sigma^2}{p_w^2}\right)\right].$$

Because of the presence of the noise component, the value of α cannot be accurately determined, while the other parameters are $P_0 \approx 10$ mW and $p_w^2 \approx 0.5$ W^2. By assuming that $\alpha \sim 1$ (with an accuracy of 10 % – 20 %), which corresponds to the formation of radiation without threshold, we can estimate by the linear part of the curve in the region of lower powers the quadratic susceptibility $\chi^{(2)} \approx 2 \times 10^{-6}$ m V^{-1}. This value is 5 – 6 orders of magnitude greater than the maximum quadratic susceptibility $\chi^{(2)} \sim 10^{-12}$ m V^{-1} measured for an NPP organic crystal [N-(4-nitropheny)-L-prolynol] [11]. The maximum efficiency of conversion of radiation 'forward' (along the incident beam) $\eta = P_\Delta/P_\Sigma$ is achieved for the incident power $P_\Sigma \approx p_w \approx 0.7$ W and is ~0.7 %.

The saturation of the radiation power at difference frequencies is most likely explained by the fact that the phase-matching condition changes at high incident radiation powers during the conversion process, and higher (than cubic) terms in the expansion of induction as a Taylor power series in the electric field strength of waves in the LC medium begin to play an important role or competing processes come to play.

We observed the nonlinear conversion of radiation in the NLC, which was accompanied by highly efficient lasing at the difference frequency. This process is more complicated than the manifestation of quadratic nonlinearity in solid crystals. The quadratic frequency conversion is considered as approximation whose applicability should be elucidated. The estimate of the effective nonlinearity taking into account only the quadratic mechanism gives the value greatly exceeding these values for solid crystals. This result has been obtained for the first time and has a number of specific features compared to lasing at difference frequencies in solid crystals. All this requires a careful analysis of the real mechanism of frequency conversion in LCs.

References

1. Zel'dovich B.Ya., Tabiryan N.V. *Usp. Fiz. Nauk*, **147**, 633 (1985).
2. Blinov L.M. *Elektro- i magnitooptika zhidkikh kristallov* (Electro- and Magnetooptics of Liquid Crystals) (Moscow: Nauka, 1978).
3. Zolot'ko A.S., Kitaeva V.F., Kroo N., Sobolev N.N., Csillag L. *Pis'ma Zh. Eksp. Teor. Fiz.*, **32**, 170 (1980).
4. Ou-Yang Zhong-can, Xie Yu-zhang. *Phyz. Rev. A*, **32** (2), 1189 (1985).
5. Arakelyan S.M., Chilingaryan Yu.S. *Nelinenaya optika zhidkikh kristallov* (Nonlinear Optics of Liquid Crystals) (Moscow: Nauka, 1984).
6. Arakelyan S.M., Grigoryan G.L., Nersisyan S.Ts., Chilingaryan Yu.S. *Zh. Ekst. Teor. Fiz.*, **80**, 1883 (1981).
7. Shtykov N.M., Blinov L.M., Dorozhkin A.M., Barnik M.I. *Pis'ma Zh. Eksp. Teor. Fiz.*, **35**, 142 (1982).
8. Trashkeyev S.I., Grachev G.N., Klementyev V.M., Statsenko P.A. *Modern Problems of Laser Physics. Techn. Digest* (Novosibirsk, Institute of Laser Physics SB RAS, 2004) p. 296.
9. Kitaeva V.F., Sobolev N.N., Zolot'ko A.S., Csillag L., Kroo N. *Mol. Cryst. Liq. Cryst.*, **91**, 137 (1983).
10. Trashkeev S.I., Klement'ev V.M., Statsenko P.A. *Liq. Crist.*, **33** (4), 417 (2006).
11. Zyss J., Chemla D.S. (Eds) *Nonlinear Optical Properties of Organic Molecules and Crystals* (Orlando: Academic Press, 1987; Moscow: Mir, 1989).