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Generation of harmonics and supercontinuum in nematic liquid crystals*

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Abstract. **Nonlinear optical properties of nematic liquid crystals (NLC) have been investigated. A technique for efficient laser frequency conversion in a microscopic NLC volume deposited on an optical fibre end face is experimentally demonstrated. An efficient design of a compact NLC-based IR frequency converter with a fibre input and achromatic collimator is proposed and implemented. Simultaneous generation of the second and third harmonics is obtained for the first time under pumping NLC by a 1.56-μm femtosecond fibre laser. The second-harmonic generation efficiency is measured to be about 1 %, while the efficiency of third-harmonic generation is several tenths of percent. A strong polarisation dependence of the third-harmonic generation efficiency is revealed. When pumping NLC by a cw laser, generation of spectral supercontinua (covering the visible and near-IR spectral ranges) is observed. The nonlinear effects revealed can be due to the light-induced change in the orientational order in liquid crystals, which breaks the initial symmetry and leads to formation of disclination structures. The NLC optical nonlinearity is believed to be of mixed orientational-electronic nature as a whole.**

Keywords: *optical nonlinearity, nematic liquid crystal, harmonic generation, spectral supercontinuum.*

1. Introduction

Nonlinear optical properties of liquid crystals (LCs) [1, 2] and their nanocomposites [3] are fairly complex and diverse. Study of these properties is of great interest for researchers specialising in photonics, nonlinear optics, physics of nanoparticles, laser physics, and information and telecommunication technologies. The reason is that LCs form a kind of the so-called soft matter, whose physical properties can be controlled at micro- and macroscopic levels using fairly weak external effects. In addition, the nonlinear susceptibility of nematic LCs (NLCs) may significantly exceed the susceptibility of solids, and the phase-matching conditions are not strictly related to the geometry of the medium, because they are determined in many respects by the parameters of light fields deforming the NLC orientational structure [3 – 5].

To date, many optical properties of LCs have been revealed, theoretically predicted, or experimentally studied in detail (the state of the art in this field is reviewed in [6]). Nevertheless, much attention is paid to the development of new practical methods implying the use of nonlinear optical properties of LCs, design of real devices on their basis (for example, miniature or microscopic laser frequency converters integrated with an optical fibre), and study of their limiting parameters. To date, experiments aimed at implementing nonlinear optical conversion processes in NLCs, related to both quadratic [4, 5, 7] and cubic [8 – 10] nonlinearities, have been performed using complicated and cumbersome laser systems having relatively low efficiency. New optical frequencies were generated using precisely aligned bulky focusing optics and cells with NLC samples. The large angular divergence and complex emission indicatrix in the systems of this kind hinder efficient radiation collection and collimation into beams fit for practical use.

In this paper, we experimentally demonstrate laser frequency conversion in a microscopic NLC volume (in particular, on the end face of an optical fibre delivering pump radiation) and report the results of preliminary study of this effect. In addition, we analyse the related structural (orientational) changes in the NLC and indicate possible mechanisms of the nonlinear phenomena observed. An efficient design of a simple compact NLC-based IR frequency converter with a fibre input and achromatic collimator is proposed and approved for the first time. Its efficiency for generating femtosecond radiation harmonics into a high-quality beam is demonstrated.

2. Experimental

Preliminary study of light-induced structural (orientational) changes in microscopic NLC volumes that are in optical contact with a fibre delivering laser radiation were performed using a planar NLC cell, formed by two plane-parallel glass plates (Fig. 1). The plates had a transparent conducting ITO coating, due to which an external electric field could be applied to the NLC. The distance between the plates (125 μm)

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was determined by the external diameter of the telecommunication optical fibre immersed in the NLC cell. The fibre end face was cleaved at an angle close to the right one. Near-IR cw laser radiation was fed to the NLC through the fibre. We used a semiconductor laser FOL1425RUZ (Fitel) having a single-mode fibre output and the following parameters: centre wavelength ~1480 nm, spectral width below 8 nm, and maximum optical power 400 mW. The diameter of the laser mode field in the fibre transverse cross section was ~10 μm. The orientational changes in the NLC near the fibre end face were investigated with a polarisation microscope. The radiation generated in the NLC was recorded beyond the cell by optical spectrum analysers using wide-aperture collecting optics.

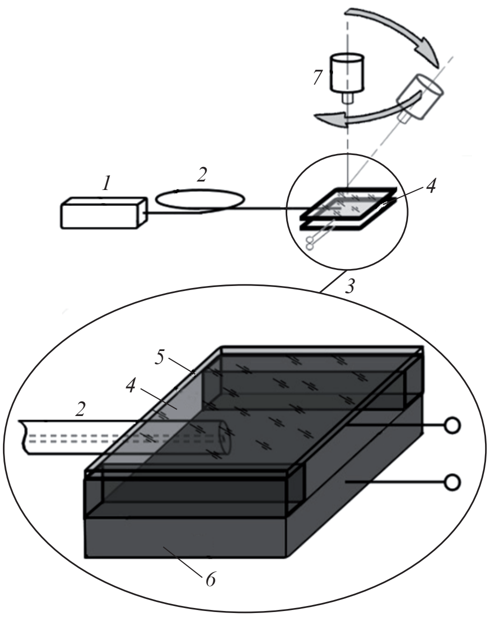
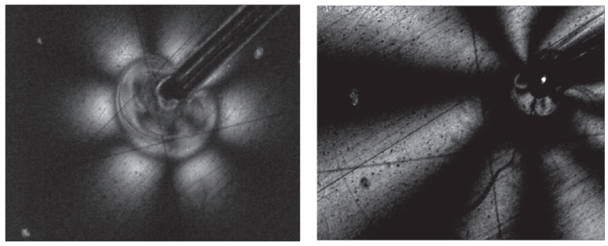


Figure 1. Schematic of the experiments with an NLC cell: (*1*) cw IR diode laser (λ = 1480 nm) with a fibre pigtail; (*2*) optical fibre supplying pump radiation to the NLC; (*3*) cell; (*4*) NLC; (*5, 6*) glass plate with ITO-coating; and (*7*) observation system (polarisation microscope, CCD camera, or spectrometer).

Preliminary experiments with this system demonstrated efficiency and reliability of direct optical contact between the single-mode optical fibre and NLC. Due to the small mode field area on the fibre end face, the power threshold for the occurrence of stationary light-induced changes in the NLC orientational structure (formation of disclinations) was extremely low (several tens of milliwatts). The threshold for nonlinear laser frequency conversion was also in the low-power range (100–200 mW) and had a hysteretic character. The disclination core and the generation of visible light due to nonlinear conversions are clearly localised in the microscopic NLC volume on the fibre end face (Fig. 2). In these experiments the NLC was not overheated and did not pass to the isotropic state even at the maximum pump power density (~500 kW cm^{-2}). Since the difference in the refractive indices at the glass–NLC interface is relatively small (the NLC refractive indices are 1.5–1.7 in the near-IR range), the backward reflections were much less intense than the Fresnel reflection from glass in air.

Based on the positive results of the aforementioned preliminary experiments [3, 11], it was concluded that an efficient



a b

Figure 2. Light-induced orientational NLC structures near the optical fibre end face, which correspond to disclinations of different strengths (a) at a pump power of ~100 mW and an ac electric field applied to the cell and (b) at a pump power of ~350 mW. The bright spot in panel b is due to the visible light generated in the microscopic NLC volume on the fibre end face.

laser frequency converter can be designed based on microscopic NLC volume, integrated with an optical fibre. The experimental optical scheme developed by us is shown in Fig. 3a. An NLC drop was deposited on a polished end face of telecommunication fibre connector (FC/PC) with a single-mode fibre. The drop volume was chosen so as to cover the central region on the connector (ferule) end face by a homogeneous thin NLC layer, with a thickness no more than ~200 μm. Due to the good adhesion and strong surface tension forces, the thus formed optical NLC layer was fairly

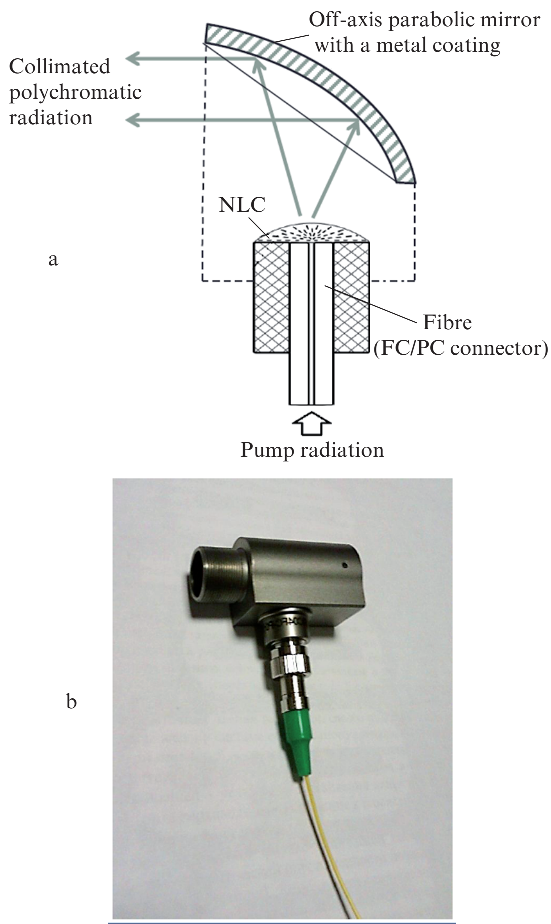


Figure 3. (a) Optical scheme and (b) the appearance of an NLC-based nonlinear optical converter integrated with optical fibre.

stable and reliably fixed on the connector end face, thus enabling free motion of the device in space. The connector operated in the vertical position (in this case, the NLC layer is not deformed).

A collimator (an off-axis wide-aperture parabolic aluminum mirror, which directed the collimated beam at an angle of 90° to the optical fibre axis) was installed above the connector. An advantage of this scheme is its universality: the achromatic wide-aperture collimator we used makes it possible to efficiently collect generated radiation with large angular divergence and an arbitrary indicatrix in a wide spectral range (from long-wavelength UV to far IR). The collimated output beam lies in the horizontal plane. The design of the laser frequency converter based on this optical scheme is extremely simple, compact, and does not require alignment. The collimating mirror is placed in a Γ-shaped metal housing with a standard optical socket FC, to which a fibre connector with the NLC is fastened through a threaded connection. The design appearance is shown in Fig. 3b. The converter outlet (output aperture ~5 mm) was not equipped with a glass window but was closed by a hermetic lid when the converter was not in use. This procedure made it possible to avoid fast drying and contamination of NLC, whose nonlinear optical properties did not degrade even after several months. In particular, the results obtained with the same NLC sample were reproduced well during three months. In addition, this design allows one to change easily NLC samples in the converter. Experiments were performed with commercial NLC samples based on cyanobiphenyls with a mesophase temperature range from –30 to +90 °C (NLC-1289, NLC-7814A). All samples had similar nonlinear optical properties.

The nonlinear optical frequency converter was pumped by a femtosecond erbium fibre laser developed by us. The laser had a linear-ring cavity, almost the same as that reported in [12]. However, laser mode locking was performed in our case using a saturating semiconductor SESAM mirror, mounted in the linear part of the cavity instead of the conventional mirror. The laser was supplemented with a fibre optical power amplifier, which was developed at the Institute of Laser Physics (Russian Academy of Sciences, Siberian Branch). The amplifier was designed according to the classical EDFA scheme [13], based on erbium-doped fibre (nLight Liekki Er 80-8/125), which was pumped in the opposite direction at a wavelength of 1480 nm. Fibres that do not maintain polarisation were used in both laser and amplifier designs. The femtosecond radiation parameters at the output of the laser system were as follows: centre wavelength ~1560 nm, optical spectrum width ~5.5 nm, pulse width ~600 fs, pulse repetition rate ~17 MHz, and maximum average power ~60 mW.

3. Results of the experiment on generation of harmonics

The output radiation of the nonlinear optical converter was investigated using a QE65000 optical spectrum analyser (Ocean optics) with a resolution of ~0.7 nm. Simultaneous second- and third-harmonic generation (SHG and THG, respectively) was observed at wavelengths of ~780 and ~520 nm (Fig. 4).

The threshold pump power at which SHG was recorded amounted to ~3 mW. When the pump power increased, the second-harmonic intensity grew almost linearly, and the optical spectrum width remained almost constant and equal to ~2.5 nm (Fig. 5). The difference of the shape and width of the

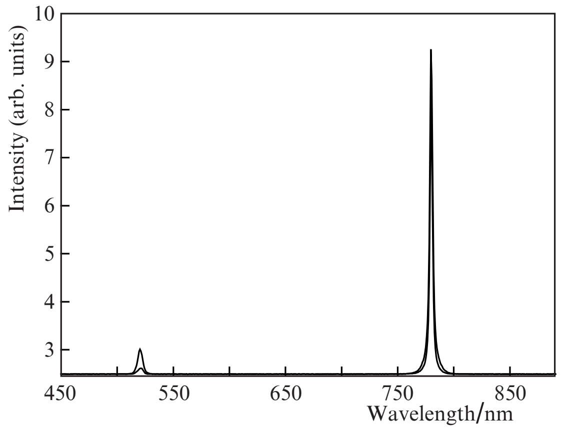


Figure 4. Optical spectrum of the output radiation of nonlinear optical converter pumped by 1560-nm femtosecond laser radiation with the uncontrolled polarisation state.

harmonic spectrum from the fundamental radiation spectrum is typical of frequency conversion for ultrashort laser pulses in nonlinear optical media. The second-harmonic spectrum width can be either larger or smaller than the fundamental-radiation spectrum width; their ratio is determined by the optical-medium dispersion characteristics, pulse width, and the profile of the fundamental-radiation spectrum [14].

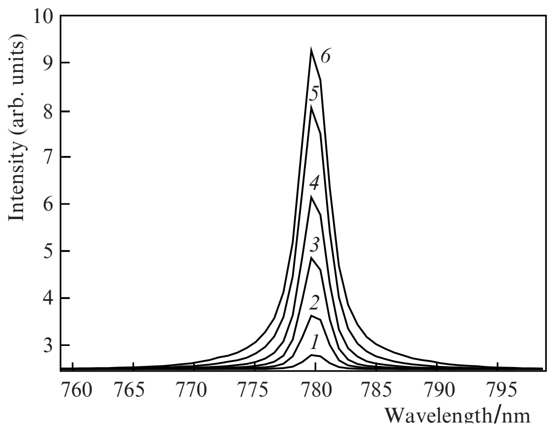


Figure 5. Optical second-harmonic spectra at pump powers of (*1*) 10, (*2*) 20, (*3*) 30, (*4*) 40, (*5*) 50, and (*6*) 60 mW.

The threshold pump power at which THG was recorded turned out to be ~17 mW. With an increase in the pump power (from threshold to 50 mW) the third-harmonic intensity, as well as in the case of SHG, increased almost linearly. However, with a further increase in the pump power (from 50 to 60 mW), the third-harmonic intensity grew at a higher rate. The optical-spectrum width remained almost constant: approximately 5 nm (Fig. 6). Estimation of the SHG/THG power ratio after integration over the spectral profiles of harmonics showed that, even at the maximum pump power, the second-harmonic power exceeds the third-harmonic power by a factor of more than 10 on average. The pump polarisation was not monitored in this experiment.

In this experiment we observed time fluctuations of harmonic intensities and spectral profiles, caused by thermal and orientational perturbations in the NLC, as well as by the

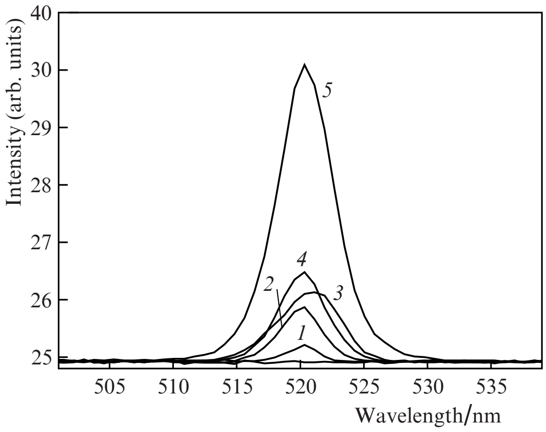


Figure 6. Optical third-harmonic spectra at pump powers of (*1*) 20, (*2*) 30, (*3*) 40, (*4*) 50, and (*5*) 60 mW.

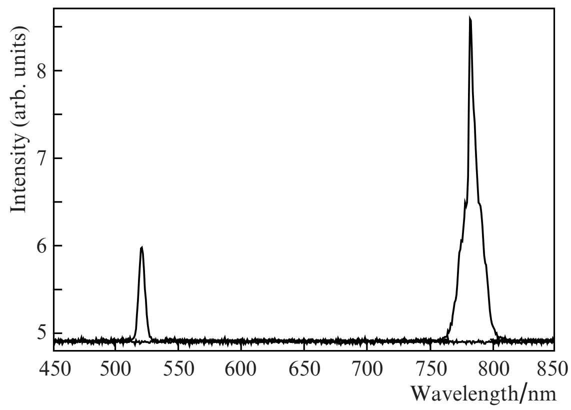


Figure 7. Optical spectrum of the output radiation of nonlinear optical converter pumped by 1560-nm femtosecond laser radiation with the optimal (linear) polarisation state (optimised THG).

polarisation instability of the fibre laser system used for pumping. They were most pronounced in the third-harmonic signal, whose intensity could spontaneously change in a fairly wide range. Nevertheless, the SHG stability and reproducibility are comparable with those for nonlinear conversion in purely solid-state laser systems. According to our estimates, fluctuations of the second-harmonic signal intensity did not exceed ~5% at pump intensity instability less than 1%. The reason is as follows: when multifrequency laser radiation (femtosecond laser modes) is used, second-harmonic intensity fluctuations are mainly related to the phase (rather than amplitude) fluctuations of the fundamental radiation. It is impossible to obtain ideal phase locking of all longitudinal modes of femtosecond laser in practice; therefore, harmonic intensity fluctuations are also observed for stabilised amplitudes of laser modes (see [14] for details).

To study the effect of pump polarisation on the harmonic generation efficiency, we installed a polarisation controller between the nonlinear optical converter and femtosecond fibre laser system. This controller was a set of zero-order phase plates with fibre outputs. The radiation at the converter output was analysed using a film polariser. The third-harmonic intensity was found to depend strongly on the pump polarisation. The maximum and minimum THG efficiencies were observed for, respectively, linearly and circularly polarised pump radiation. The polarisation dependence was studied only qualitatively. Figure 7 shows the optical spectrum recorded for optimal pump polarisation: the third-harmonic intensity multiply increases, and its total power is less than the second-harmonic power by a factor of no more than three. At the same time, the pump polarisation only slightly affects the second-harmonic intensity.

The strong polarisation sensitivity of the third harmonic, as well as its nonlinear dependence on the pump power, can be explained by the more complicated (in comparison with SHG) conversion mechanism and phase-matching conditions. The third optical harmonic in a nonlinear medium without inversion centre may arise both as a result of nonlinear mixing of three pump waves with the same frequency ω (due to the cubic susceptibility) and in the cascade (stepwise) way during second-harmonic generation and nonlinear mixing of two waves with frequencies 2ω and ω [15]. This combined process with a susceptibility $\chi^{(3)}_{\rm casc}$ can be described in terms of effective cubic nonlinearity as a relation

$$\chi^{(3)}_{\rm casc}(3\omega,\omega,\omega,\omega) \sim \chi^{(2)}(2\omega,\omega,\omega)\chi^{(2)}(3\omega,2\omega,\omega),$$

where $\chi^{(n)}$ are nth order nonlinear susceptibilities.

The nonlinear optical converter and femtosecond laser system were designed using conventional telecommunication fibres that do not maintain polarisation. Therefore, to maintain the optimal polarisation state of pumping and, correspondingly, the maximum third-harmonic signal power, it was necessary to perform regular tuning of the polarisation controller and protect the system from external (acoustic and vibrational) perturbations.

The energy efficiency of the observed nonlinear optical conversions was determined by spectral selection of the second-harmonic signal using a glass prism, measuring its average power, and correlating the value obtained with the pump power, which was measured directly at the output of the fibre connector before depositing the NLC on it. The maximum measured value of the second-harmonic power was ~0.3 mW (at a pump power of ~60 mW).

Thus, the conversion efficiency of the entire optical scheme (including the nonlinear optical converter, prism, and additional IR filter in the meter power) for SHG was no less than 0.5%. Taking into account the optical loss for second-harmonic radiation in auxiliary optical elements of the scheme (~20% in the collimating mirror, ~8% in the prism, and ~8% in the IR filter), one can estimate the NLC efficiency. In the case of frequency doubling it is ~0.78%.

The THG efficiency was estimated without direct measurements of the third-harmonic signal power. To this end, we used the ratio of the total harmonic powers corresponding to the optimal pump polarisation state (it was found to be approximately 1 : 3 by analysing the spectra). In addition, we took into account the wavelength dependence of the optical loss introduced by the collimating mirror (~20% for the second harmonic and ~14% for the third harmonic). Thus, the NLC efficiency for THG was ~0.24%, which is two orders of magnitude larger than the maximum efficiency obtained previously [8]. In total, more than 1% of pump power was converted into harmonics in the microscopic NLC volume, which was confirmed by measuring the unabsorbed pump radiation at the output of the nonlinear optical converter.

Except for SHG and THG, no other nonlinear optical conversions were found under NLC pumping by femtosecond laser radiation. Study of the optical spectrum of femtosecond pump radiation after its passage through the NLC did not reveal its significant broadening (the measurement accuracy was limited by the analyser resolution). This fact indicates the absence of significant phase self-modulation of femtosecond pulses in the NLC, which could be caused by fast (with a femtosecond time response) cubic nonlinearity. However, this pattern can be explained by the small interaction length of radiation with the nonlinear medium occupying a microscopic volume.

4. Results and discussion

Currently, there is no sufficiently complete description of nonlinear optical properties of LCs in the form of a complete physico-mathematical model. The development of a correct model for LCs is impeded by the mixed electronic-orientational nature of optical nonlinearity, strong nonlocality of nonlinear response, giant nonlinear orientational susceptibility, and the related LC optical instability. This circumstance hinders analytical description and formalisation of the effects of laser radiation self-action that arise in NLCs (in particular, during frequency conversion) in terms and approximations of classical solid-state nonlinear optics. Therefore, we did not perform detailed analysis of the problem of phase matching during harmonic generation in NLCs. Below we report only some considerations and suggestions concerning this issue.

The direction of optical axis in NLCs is related to the director, which is defined as the vector of preferred orientation of the long axes of NLC molecules and depends on many external and internal factors. The orientation of NLC molecules is determined by applied electrostatic and light fields (the Freedericksz effect [2]), heat flows (thermal orientation effects [16]), the shape of the volume occupied by the NLC and the boundary conditions on the surface [6], and the internal disclinations [3]. All these factors may lead to complex NLC anisotropy, inhomogeneous in space and time. Due to the nonlinear orientational susceptibility, spatial optical solitons (the so-called nematicons [17]) and filaments [3, 17] can be formed in NLCs under laser irradiation; as a result, the general pattern of nonlinear optical conversions in NLCs is complicated even more.

The possibility of phase matching of a particular type at frequency multiplication in NLC depends on the specific experimental conditions and parameters. For example, it was shown in [4] that, when pulsed laser radiation is used, spatially periodic orientational deformation may arise in the NLC bulk, which not only leads to local removal of inversion center but also provides (at a proper spatial period) synchronous SHG of the oo–o interaction type. The spatially periodic orientational deformation of the NLC by femtosecond laser radiation also allows for synchronous THG [8]. Due to the localisation of femtosecond pump radiation in spatial solitons, I-type phase matching can be implemented for THG in the NLC bulk [10]. The possibility of synchronous THG in NLC (provided that the II-type phase-matching condition is satisfied) was demonstrated in [9]. Note that in all cases NLC samples were located in planar glass cells. This configuration makes it possible to control the angle between the NLC director and the Poynting vector of the pump radiation, and, therefore, perform tuning to synchronism of a particular type. At the same time, this design is poor for practice because it does not allow one to efficiently collect and collimate generated radiation. As a result, the efficiency of the demonstrated synchronous conversions did not exceed 10^{-5}.

On the whole, it is a fairly difficult problem to find out the parameters determining the phase-matching conditions in the NLC volume. To solve this problem, one must take into account the strong deforming effect of the pump radiation, external perturbations, and internal defects on the NLC orientational structure. Nevertheless, we can suggest that, when harmonics are generated in microscopic NLC volumes (i.e., in configurations similar to our case), the problem of tuning to phase matching may be not so urgent as in the case of harmonic generation in solid crystals. For example, the radiation frequency can be doubled in a thin layer of a nonlinear medium, the thickness of which does not exceed the coherence length l_{coh} related to accumulation of the generated field energy (nonphase-matched SHG) [14, 15]. If the NLC layer thickness is odd multiple of l_{coh}, due to the extremely high dipole quadratic susceptibility (which is characteristic of NLC with removed inversion centre [4]), the nonphase-matched SHG efficiency can be relatively high, despite the small coherence length ($l_{coh} \leqslant 10$ μm for NLC). The absence of explicit dependence of the second-harmonic intensity on the pump polarisation state can be considered as an indirect confirmation of the dominance of SHG nonphase-matched regime in the case under consideration. However, one can also suggest that the conditions for noncollinear phase matching can be locally fulfilled for some indicatrices within the divergent pump beam in the NLC drop.

In our case one cannot also exclude the possibility of the so-called self-phase-matched THG. Direct self-phase-matched THG may occur in nonlinear media with anomalous dispersion when using a Bessel pump beam [18, 19]. A Bessel beam can also provide self-phase-matched (with noncritical longitudinal phase matching) THG via four-wave mixing ($\omega_3 = 2\omega_2 - \omega_1$) in nonlinear media with normal dispersion [20]. The transverse field distribution in a single-mode optical fibre is known to be described by the zero-order Bessel function J_0. In addition, the NLC drop on the fibre end face, which has a certain radial orientational structure [21], can play a role of a microscopic axicon, which transforms the pump beam emerging from the fibre into a ring conical Bessel-like beam. This possibility was indirectly confirmed by observing the shape of the spot of unabsorbed pump radia-

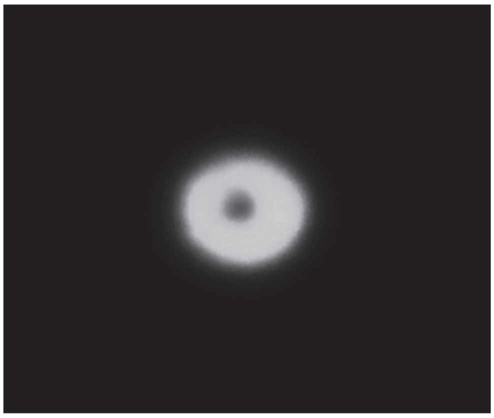


Figure 8. Spot of unabsorbed pump radiation, observed with the aid of a luminescence IR visualiser at the converter output.

tion in the far-field zone (Fig. 8). The dependence of the third-harmonic intensity on the pump polarisation state, which was found by us, can also be considered as an indirect evidence of the dominantly phase-matched character of THG.

To determine exactly the character of nonlinear interactions during harmonic generation by the technique proposed by us, it is primarily necessary to reconstruct the structure of induced orientational anisotropy in a microscopic NLC volume, which is a fairly difficult problem. Nevertheless, even putting it aside, the unique design of our converter provided a much higher harmonic generation efficiency in the NLC in comparison with earlier studies.

5. Generation of a supercontinuum

Along with generation of optical harmonics, we demonstrated generation of a spectral supercontinuum upon pumping a nonlinear optical converter by a cw laser. Previously a similar effect was observed and investigated by us when pumping microscopic NLC volumes by cw diode lasers with fibre pigtails, operating in the multifrequency regime at wavelengths of ~980 and 1480 nm [3, 11]. In this study pumping was performed by a cw single-frequency SFL1550P diode laser (Thorlabs) with a fibre pigtail (λ = 1550 nm). The laser was supplemented with a fibre optical amplifier, which provided a maximum output power of ~110 mW. The amplifier was similar to that used in the femtosecond laser system. Figure 9 shows the optical emission spectra at the converter output, which were recorded at different pump powers. The threshold pump power at which spectral supercontinuum arose, was ~40 mW. The integral power of supercontinuum increased quadratically with the pump power and its spectral width (at a level of 0.1) exceeded an octave. The spectrum envelope and amplitude were unstable (fluctuating with time); this instability was more pronounced at higher pump powers. The characteristic times of supercontinuum fluctuations (0.1 – 1 s) coincided with the those of hydrodynamic and orientational perturbations of the microscopic NLC volume, which are observed at high (above 100 mW) cw pump powers.

Detailed analysis of the characteristics and explanation of the physical mechanisms of generation of such supercontinua is beyond the scope of this paper; we plan to perform it in subsequent studies. We should only note that a similar phenomenon was observed previously by Enikeeva et al. [7], who pumped an NLC cell by a solid-state femtosecond laser with a wavelength of ~800 nm. They observed a wide spectral background (luminescence) in the range from 350 to 500 nm when studying the second-harmonic generation ($\lambda \approx$ 400 nm) and suggested a multiphoton mechanism of luminescence excitation. Indeed, this spectral background is in good agreement with the fluorescence spectrum of 5CB cyanobiphenyl (NLC base), which arises when pumping by UV laser radiation [22]. However, spectral supercontinua were observed in our studies in the case of NLC pumping by cw IR lasers, while pumping by a femtosecond laser gave rise to only harmonics. A possible reason for this difference is that a higher power and, simultaneously, shorter wavelength femtosecond laser was used in [7]; under these conditions multiphoton excitation is more likely to occur. In addition, the spectral supercontinuum obtained by us is much wider and barely overlaps the known fluorescence spectra of cyanobiphenyl-based NLCs, because it lies in the longer wavelength region (from 500 to 1100 nm). Therefore, along with photoluminescence, another possible mechanism of spectral supercontinuum formation in our case is the complex combination of different nonlinear optical conversions of cw laser radiation in the NLC. An example is a similar (in outward appearance) phenomenon of forming spectral supercontinua (which also covers the visible wavelength range) when near-IR cw laser radiation propagates through highly nonlinear optical fibres. The mechanism of formation of these supercontinua is based on the combination of modulation instability effects with the interactions between solitons and dispersion waves [23]. The possibility of this manifestation of temporal modulation instability in NLCs, which leads to pump radiation conversion into a sequence of ultrashort pulses and occurrence of new frequencies in the optical emission spectrum, was predicted theoretically in [24].

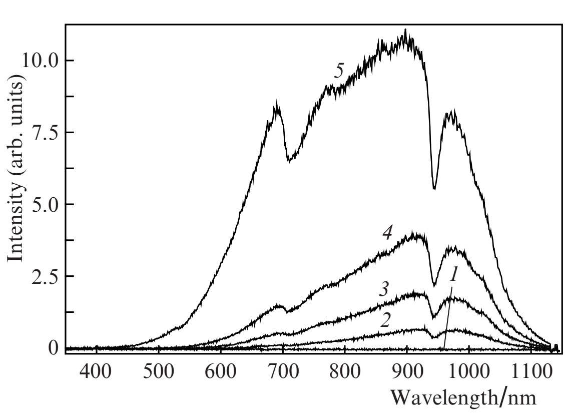


Figure 9. Spectra of supercontinua generated in the nonlinear optical converter at cw pump powers (λ = 1550 nm) of (*1*) 0, (*2*) 55, (*3*) 67, (*4*) 80, and (*5*) 93 mW.

6. Conclusions

We experimentally investigated a new method of laser frequency conversion in a microscopic NLC volume that is in optical contact with a fibre through which laser radiation is delivered. This technique allowed us to design an efficient and compact NLC-based nonlinear optical converter for the IR range. It was shown that, pumping this converter by a femtosecond fibre laser, one can obtain high-efficiency generation of the second and third optical harmonics. Along with the high nonlinear conversion efficiency, the proposed optical scheme provides high-quality achromatic collimation of generated radiation and good reproducibility of its parameters. At the same time, it was shown that, to ensure maximum THG efficiency and stability, one must maintain the pump polarisation in the linear state; therefore, polarisation-maintaining fibres are preferred. The harmonic generation has a threshold character, which is related to the threshold effects of deformation of the orientational structure and violation of NLC symmetry when the light field power density reaches a certain level in the NLC. On the whole, the NLC optical nonlinearity has in reality a mixed orientational – electronic nature, which is to be completely understood. Along with the generation of femtosecond optical harmonics, the possibility of generating a spectral supercontinuum in the visible and near-IR spectral ranges upon pumping a microscopic NLC volume by cw IR lasers was experimentally demonstrated. The spectral envelope of these supercontinua is characterised by complex (stochastic) dynamics. Their formation mechanism calls for further study.

The results obtained are of great practical interest, because they may provoke further investigations and refinement of the fundamental physical properties of LCs and the development of nonlinear optics, laser physics, and telecommunications.

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