Temperature Dependence of Optical Harmonic Generation in Nematic Liquid Crystals

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Abstract: Temperature dependence of optical harmonic generation in nematic liquid crystals is investigated. Efficiency of the third harmonic generation spikes at the temperature of nematic-isotropic transition. Maintaining this temperature allows exploitation of the enhanced cubic nonlinearity.

We report on the strong temperature dependence and bistability of the nonlinear optical properties of nematic liquid crystals (NLCs) near the point of their nematic-isotropic phase transition.

In an experiment of femtosecond laser frequency multiplication by means of a fiber-coupled NLC [1, 2] we have found that efficiency of the third harmonic generation (THG) spikes at the temperature of nematic-isotropic transition (T_{NI}). The measured intensity of THG radiation rapidly increases by a factor of more than 20 since the temperature of a fiber-coupled NLC subjected to heating approaches T_{NI} (see Fig.1). NLC exhibits such a particular behavior of cubic nonlinearity within a temperature interval as narrow as few degrees Celsius. Further heating ends immediately in fall of THG intensity down to almost zero. At the same time the second harmonic generation (SHG), which was not sufficiently affected by prior heating, breaks off as well. Proceeding with overheating results in recovery of weak SHG and THG. When reversing the NLC temperature, efficiency of harmonic generation varies (according to the temperature) in a similar manner, but the spike of THG appears at a lower temperature (few degrees below T_{NI}). Thus, NLC exhibits bistabilty of its optical nonlinearity relevant to optical frequency conversion. This probably may be due to the effect of supercooling of NLC isotropic state [3].

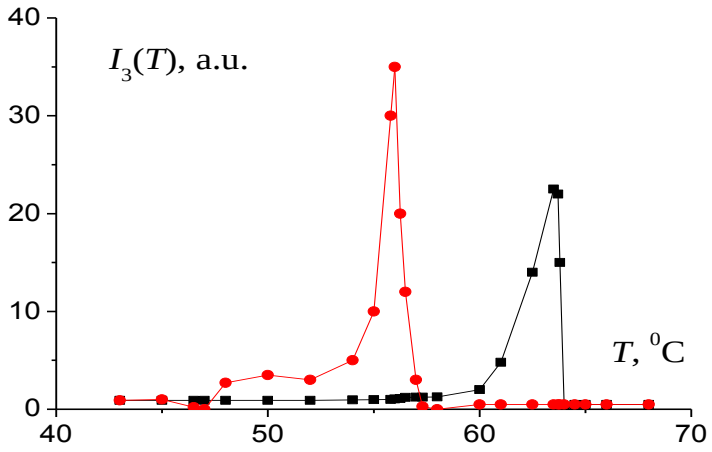

Fig.1. Dependence of the THG intensity on the NLC temperature: the black line (squares) corresponds to the heating; the red line (circlets) – to the cooling.

In the above experiment we made use of cyanobiphenyl liquid crystals with $T_{NI} \approx 58 \div 61$ °C. The used femtosecond laser source and the experimental arrangement for its fiber coupling with NLC are described in Ref. [2].

We have also demonstrated that thermostabilization of NLC at a proper temperature near its transition point enables maintenance of the improved THG efficiency for a long while. The improved THG efficiency can reach ~ 10% according to our estimation based on the comparison with data from Ref. [2]. At this rate, a laser frequency multiplier based on a fiber-coupled NLC becomes competitive with corresponding solid state nonlinear devices. As concerning the dimensions, controllability, and cost, the NLC features even superior properties.

Though the underlying physics of the revealed temperature-induced modification of effective cubic nonlinearity needs to be further clarified, NLCs can be already considered as prospective nonlinear optical media for the development of new practical devices for laser frequency conversion.

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