QWD5
Cumulative negative nonlinearity in planar nematics driven by nanosecond, 532-nm laser pulses with linear polarization parallel to the liquid crystal director
Svetlana G. Lukishova,* Liquid Crystal Institute, Kent State University, Kent, Ohio 44242 USA; E-mail: svetlana@cppkent.edu

For optical power limiting studies, we chose planar-aligned films of the well-known nematic liquid crystals alkyl-cyano-biphenyl 5CB (C_{11}H_{23}), and its mixture (1:4 by weight) with 7CB (C_{12}H_{25}). In some experiments, we added to this mixture 7.4 weight % of the two-photon absorptive chromophore bis(di-n-butylamino)stilbene.1

The liquid crystal (LC) molecules with their tendency to align with the direction of an applied optical electric field, do in general not respond when the LC molecular dipole is parallel to the linear polarization (nematic cell surface was perpendicular to the beam propagation direction) (Fig. 1a) of the incident pulse. However, during periodic illumination over timespans of 0.5–several seconds we observe planar-aligned nematic cells in such orientation to imprint onto a linearly polarized beam a far-field pattern of high-contrast, concentric, elliptical diffraction rings whose major axis orients itself perpendicular to the incident polarization direction (Fig. 2). Once formed, each set of rings may remain stable for up to several minutes. Upon continued irradiation over several minutes, the number of these rings varies systematically with laser intensity variation (sometimes between 1–2 and (up to) 20 rings) (Fig. 2).

This effect and its development exhibited threshold behavior depending on the incident peak intensity and the geometry of irradiation (I ~ 0.5 GW/cm², 7–10-ns pulse duration, 50-μm beam diameter), but not on the fluence or average power. Adding the chromophore did not change the character of the effect but lowered its threshold ~2–2.5 times. The effect existed at both 2-Hz and 10-Hz pulse repetition rate, however in the case of 2-Hz the threshold increased (cumulative effect).

It should be noted that the effect’s evolution depends on the irradiation geometry (focal beam diameter) and, for a given geometry, on the cell thickness, i.e., at 160-μm spot size, cells of thickness 30–50-μm did not permit evolution of the effect, while larger thickness (100–125-μm) cells did. For a 50-μm spot size, the effect was observed for all used cell-thicknesses (30, 50, 100, 125-μm).

The pattern evolves in time as shown in Fig. 3(a). The elliptical ring pattern develops from the original beam spot through the several "rays" of scattered light.

When the LC cell was rotated around the light-propagation direction, the elliptical rings' major and minor axes rotated in the same handedness, but the number of rings diminished [Fig. 3(b)] as the angle between LC director and incident linear polarization increased. However, when the LC director is oriented perpendicular to the light polarization, the effect vanishes entirely. It also disappears under CW laser irradiation of even larger average power than applied in the pulsed mode. This rules out ordinary thermal self-focusing as cause for the effect.

Instead, the strong polarization dependence of this effect may be explained by the two-photon-absorption dichroism in the oriented films of nematics.2 Our Z-scan, nonlinear-absorption measurements for LC director and incident polarization oriented parallel to one another show: (1) at lower intensities an anomalous value for the nonlinear absorption coefficient (~115 cm/GW at I = 0.36 GW/cm²), and (2) a change in sign for the nonlinear refraction from positive to negative.
value with the increasing incident intensity [Fig. 1(b) shows negative sign of refractive nonlinearity at $\lambda = 0.36 \text{GW/cm}^4$]. It is already a widely-held notion that for 532-nm, short-pulse laser irradiation two-photon absorption in LCs drives excited-state absorption. Decay of the excited states through radiationless-recombination channels causes the heating of the material. Strong local temperature gradient can give rise to a convection flow in the fluid, which then orients the molecules, in particular, also toward the direction of light propagation. Localized phase change of the nematic order into the isotropic state may also take place. Either process causes the realignment of the LC director with slow time constant.

The influence of the chromophore in lowering the nonlinearity threshold may be explained by the increasing of the two-photon absorption capability and/or the isomerization-induced intermolecular torque exerted by the dopant onto the LC molecules, facilitating realignment of the director toward the direction of the wave vector of the propagating light.

This work was supported by NSF under ALCOM grant DMR-890147 and California Institute of Technology/AFOSR No PC218970. P. Palfiy-Muhoray and T. Kosa are thanked for their support, and J. Perry for providing the chromophore.

*Also with Institute of Radioengineering and Electronics of the Russian Academy of Sciences, 11 Mokhovaya, 103907, Moscow, Russia; E-mail: svetlana@frontiernet.net


QWD6 Fig. 1. SHG spectrum (main panel) and the differential spectrum of the linear-optical absorption (inset) for the Gd-containing LB film.

QWD6 Fig. 2. The thickness dependence of the SHG magnetic contrast.

thin films. In the present paper, the magnetic properties of Gd-containing LB films are studied by means of the SHG spectroscopy and nonlinear magneto-optical Kerr effect (NOMOKE). The films studied are fabricated by LB vertical lift from the water solution of Gd acetate, as the Gd ions are adsorbed on the stearic acid LB monolayer created on the water subphase surface. The chemical analysis shows that the structural unit (period) of the LB films consists of the patterns reveal almost perfect periodic layered structure of LB films studied. For the spectroscopic MSHG studies the output of a femtosecond Ti-sapphire laser is used with the wavelength tuning range from 700 nm to 800 nm.

Figure 1 shows the SHG spectrum of Gd-containing LB film comprised of 40 structural units in the spectral range from 700 nm to 800 nm. The inset in Fig. 1 shows the linear absorption spectrum of the same film. The comparison of these spectra and direct comparison of the SHG intensity from Gd-containing the Gd-free LB films of stearic acid indicate that the nonlinear response of the LB films is attributed to the Gd ions. The NOMOKE is studied in the pre-resonant conditions at the SHG wavelength of 355 nm in the geometry of the longitudinal magnetization for the film thickness from 1 to 55 structural units (Gd monolayers). To exclude the optical interference effects the NOMOKE thickness dependence is compared with the thickness dependence of the nonmagnetic SHG response. Figure 2 shows the thickness dependence of the magnetic contrast $I_{\text{SHG}}(M_\perp) - I_{\text{SHG}}(M_\parallel)$, where $I_{\text{SHG}}(M_\perp)$ and $I_{\text{SHG}}(M_\parallel)$ are the SHG intensities for the opposite directions of the magnetic field. The dependence of the MSHG intensity on the applied magnetic field does not show hysteresis loop within the experimental accuracy for the magnetic field varied from $-2$ kOe to 2 kOe. The magnetization-induced rotation of the polarization of the SH wave for the opposite directions of the applied magnetic field of the amplitude 2.0 kOe is approximately 4 degrees.

QWD7 Second harmonic generation investigations in Zn$_x$Cd$_{1-x}$Se/ZnSe multiple asymmetric coupled quantum wells

Jianhua Xu, Xingde Lu, Wenjun Wang, Gongming Wang, Wencheng Wang, Xiqing Zhang, State Key Joint Lab. for Materials Modification by Laser, Ion and Electron Beams, Department of Physics, Fudan University, Shanghai 200433, China; E-mail: jhxu@fudan.edu.cn

From the point of view of device applications as well as basic physics, it is very important to study the nonlinear optical properties of dense excitonic systems in wide-band-gap II–VI compound semiconductors, especially in their quantum well structures. Interband second harmonic generation (SHG) including excitonic effects plays a crucial role in probing of the fundamental optical properties in near-band-gap regions. In this paper, we adopted an experimental setup (Fig. 1) for rotation-angle of sample and polarization angle of incident beam SHG measurements, and reported the reflected SHG at room temperature from Zn$_x$Cd$_{1-x}$Se/ZnSe multiple asymmetric coupled quantum wells grown on GaAs (100) substrates by molecular beam epitaxy with different well parameters. The incident beam of 50 ps pulsewidth, 10 Hz repetition rate, 1 mJ pulse$^{-1}$ energy at 1.064 µm from a mode-locked Nd:YAG laser was focused onto the samples with a spot of diameter 1 mm and a fixed incident angle 45°.

The in-plane anisotropy was clearly demonstrated in the form of a sinusoidal function of the azimuthal angle with a period of 180° in our observations.