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NONLINEAR BLEACHING IN THE SELECTIVE REFLECTION OF NONABSORBING CHIRAL-NEMATIC LIQUID-CRYSTAL THIN FILMS

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Abstract High-intensity, circularly polarized light beams tuned to selective-reflection conditions in chiral-nematic liquid-crystal layers made possible the first observation of a light-induced drop in the chiral reflection coefficient of liquid-crystal layer. The dependence of the effect on intensity (and its corresponding absence at low intensities) permits one to connect it mechanistically with the chiral nematic's helix pitch dilation up to spiral untwisting. Up to now, this effect has been observed only in static and low-frequency electric and magnetic fields. Under the current, specifically chosen experimental irradiation conditions, nonlinear bleaching of the medium's reflectivity could be observed over time intervals that allow a distinction in the driving mechanism of nonlinear bleaching between optical-field-induced and thermal processes.

Athermal pitch dilation of the cholesteric spiral in chiral-nematic liquid crystals (CLC) with an abrupt change in the medium's reflection coefficient in the presence of a Bragg-reflection-tuned, circularly polarized light field was considered in Ref. 1. However, later attempts[2] at detecting this effect in laser fields remained unsuccessful, in spite of the fact that chiral untwisting under the influence of both *static* and *time-varying* electric and magnetic fields had been observed. For such chiral untwisting to occur, the electric field strength must exceed 10^4 V/cm (for a chiral pitch of about 0.6 μm) over time periods equivalent to the helix untwisting time constant of approximately several milliseconds. In trying to overcome this threshold condition, the authors in Ref. 3 employed high-intensity, but short laser pulses (15 ns) and were unable to observe the light-field-induced effect predicted in Ref. 1, even at intensities eight times larger than originally deemed needed.[1] In practically all experimental attempts toward clarifying the nonlinear changes in the optical charactersitics of liquid-crystal mirrors,[4–7] CLC's were used with absorbing trace impurities or dopants whose absorption and subsequent heating may have biased the evolution of selective-reflection changes. In only one series of reports[8–10] were cw-light-field-induced changes in curvature of *nonabsorbing* liquid-crystal mirrors demonstrated. These *phase-front changes* were claimed to be, first, of athermal nature, and second, the result of light-field-induced pitch dilation in the chiral-nematic helix. However, no indications were given in Refs. 8–10 that a liquid-crystal selective-reflection coefficient drop was observed in accordance with Ref. 1.

In the current work the sought-after effect of the weakening selective reflection is observed through irradiation of three separate, nonabsorbing liquid-crystal mirrors by *high-repetition-rate* laser pulses with interpulse spacing chosen such that the untwisting of the chiral-nematic helix was enabled by the interaction with the liquid crystal of more than a single laser pulse per untwisting time constant.

The liquid-crystal mirrors used in our experiments were prepared from photochemically stable mixtures of the nematic E7 and the chiral twisting agent CB15 (both from E. Merck),[8–10] with mix ratios adjusted for maximum reflection around 1064 nm. Materials were filtered, analyzed, and processed in a clean room and showed no measurable absorption at both the fundamental and second harmonic of the laser. Planar alignment of the cells was achieved by the following methods. The strong-anchoring side of each cell was polyimide covered and buffed. The weak-anchoring side was left unbuffed but covered with (1) polyimide—mirror I; (2) polyvinylcinnamate and subsequent photo-orientation—mirror II; and (3) polyvinylcinnamate and subsequent photo-orientation and additional shearing of the cell—mirror III. The liquid-crystal layer thicknesses in the hermetically sealed cells were 12.8, 13.8, and 6.5 μm, respectively.

Nonlinear irradiation of CLC mirrors was carried out by two approaches: CLC samples were either irradiated in the focus of freely propagating laser beams or *inside* resonators (for the experimental layout see Fig. 1) in which the CLC structure served as output coupler. A Nd:YAG laser (type: LTI-701) operated in two regimes: (1) in cw mode and (2) in an acousto-optically Q-switched mode, emitting high-repetition-rate (4.5 kHz) pulse trains, each pulse being 500 ns long. In either mode, the *average* power reaching the CLC samples was between 0.3 and 1 W. Beam cross sections before focus were monitored by a CID camera and video processing setup. From the recorded beam profiles, $1/e^2$ intensity beam diameters of between 50 and 200 μm were derived, corresponding to sample-plane peak-power densities (for 500-ns pulses) of 10^6–10^7 W/cm^2.

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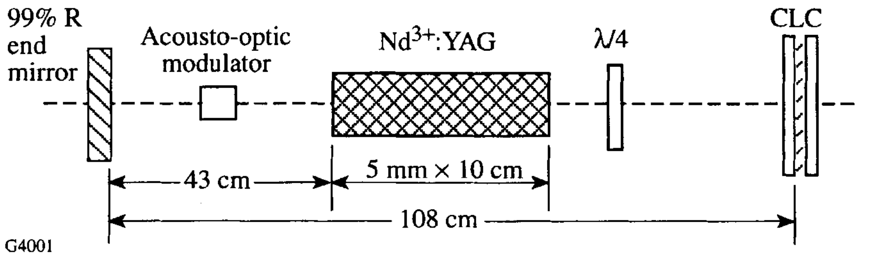

FIGURE 1 Schematic of the liquid-crystal-mirror-equipped laser oscillator: polarization-independent Q-switching by the acousto-optic modulator permits cw or 4.5-kHz pulsed-mode operation.

For the laser-resonator studies, a $\lambda/4$ plate was placed between the active element and the CLC output coupler for the purpose of controlling the power circulating inside the resonator and incident on the CLC device.

Upon focusing of free-space-propagating, 500-ns, 4.5-kHz-repetition-rate pulses into the CLC layer, we observed an increase in CLC-mirror *transmittance* of between 5% and 30%–80%. This reflectivity drop appeared within 1 to 10 min from the onset of irradiation (see Fig. 2), depending on the specific mirror. Interestingly, this effect *did not depend* on the magnitude of the average-power density. Under cw irradiation conditions, as reported in Refs. 8–10, we did not observe any effect even at power densities twice as large as the average-power densities in the pulsed mode. Furthermore, the

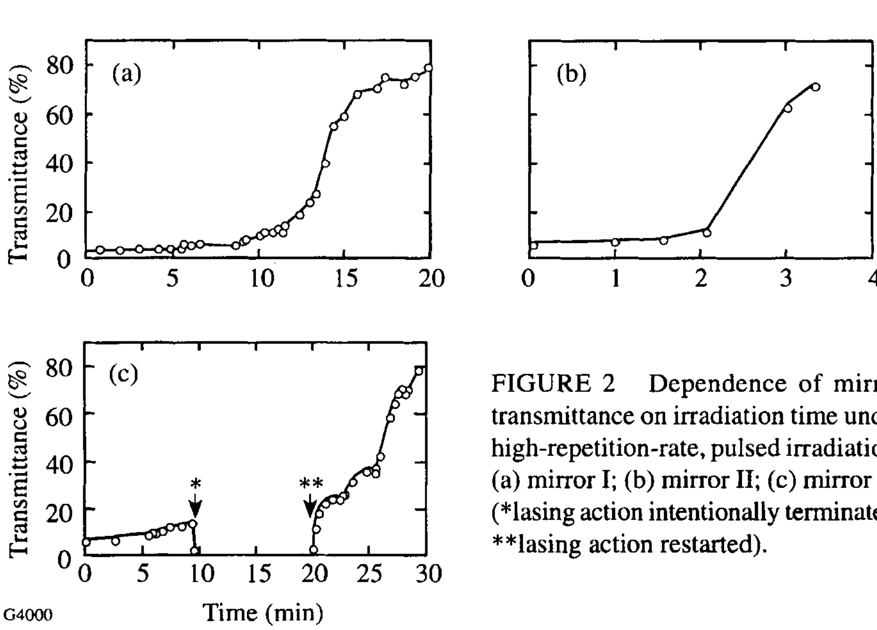

FIGURE 2 Dependence of mirror transmittance on irradiation time under high-repetition-rate, pulsed irradiation: (a) mirror I; (b) mirror II; (c) mirror III (*lasing action intentionally terminated; **lasing action restarted).

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effect appeared, even in the case of high-repetition-rate pulse irradiation, only under reflection of the incident light from the *strong-anchoring side* of the CLC cell. Under 180° reversal, i.e., with the weak-anchoring side facing the incident beam, we did not observe any effect. The temperature drift in selective reflection for our given liquid-crystal mixtures is estimated at ~+1 nm/C°, requiring a temperature excursion of more than 50°C to account for the ~70% change in CLC transmittance at 1064 nm. We did not observe such heating in the present CLC cells.

In the case of weak bleaching, a recovery of the mirror's reflectivity to its prior condition appeared after either terminating irradiation or switching it to the cw mode, which was easier to monitor with the mirror mounted in the resonator (Fig. 3). In this mode, cw generation was exceptionally stable. Under high-repetition-rate *Q*-switching the weakening of the CLC structure's reflection coefficient caused quenching of the lasing action that could be revived by switching to the cw regime (Fig. 3). This recovery was prompted by the chiral nematic's structural relaxation toward the inital-equilibrium, planar texture.

The observed bleaching of CLC mirrors may be interpreted within the frame of the chiral pitch-dilation model, in which helix deformation takes place under the influence of a high-intensity (1 to 10×10^6 W/cm^2) light field. In accordance with the derivations in Ref. 1, the critical field strength for helix untwisting in "thick" CLC layers of thickness $L > 2\lambda/(\pi\ \Delta n)^1$ takes on the form

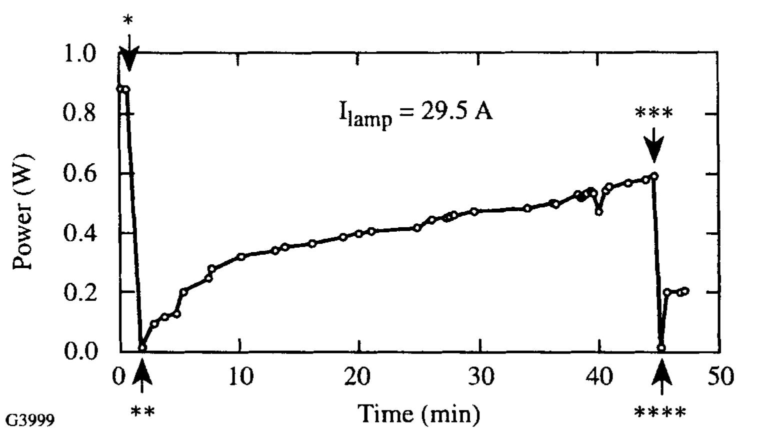

FIGURE 3 Time dependence of laser output power (*W*) for cavity with mirror III as output coupler under operating-mode switching: *switch from cw mode to pulsed; **return from pulsed to cw mode; ***switch from cw to pulsed; ****return from pulsed to cw mode.

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$$|E|^2_{cr} = 4\pi(\omega/c)^2(\varepsilon_a/\varepsilon)K_{22}, \quad (1)$$

where , $\varepsilon \sim n^2_{av}$, and the optical-regime dielectric anisotropy e_a is

$$\varepsilon_a = \varepsilon_e - \varepsilon_0 = n^2_e - n^2_0 \sim 2\ \Delta n\ n_{av}. \quad (2)$$

Substituting the approximation (2) into (1), yields

$$|E|^2_{cr} = \left(32\pi^3 K_{22}/\lambda^2\right)(\Delta n/n_{av}). \quad (3)$$

For $\lambda = 1064$ nm, $\Delta n = 0.174$, $n_{av} = 1.6$, and $K_{22} = 5 \times 10^{-7}$ dyne, the critical-field value becomes $E_{cr} = 2.1 \times 10^4$ V/cm, but $2\lambda/(\pi\,\Delta n) = 3.9\,\mu m$, i.e., considerably smaller than L for any of our three mirrors. Under our experimental conditions the field strength E at the beam center is ~ 2.2 to 7×10^4 V/cm, i.e., larger than E_{cr}.

Note that helix untwisting by a *light field* under selective reflection conditions differs from untwisting in a *low-frequency field* for which $E^2_{cr} \sim \varepsilon_a^{-1}$.[2] In the circularly polarized light field the electric-field vector changes direction throughout the CLC-layer thickness (while the low-frequency-field vector orientation remains fixed), suffering an exponential drop in magnitude over the critical length $L_c \sim \varepsilon_a^{-1}$ and, thus, remaining unable to "see" further into the CLC layer.[1] A comparison of our results with those on helix pitch dilation in a low-frequency field directed along the plane of a planar-alignment CLC layer[11,12] shows our optical field strength to considerably exceed the ~10^4 V/cm proven adequate[11,12] for helix untwisting at lower frequencies. In Ref. 12 the field-induced pitch changes relaxed in a *spatially non-monotonous, step-wise fashion*, owing to the particular field gradient chosen and the double-sided strong anchoring of the cell.

As shown in Figs. 2(a) and 2(b), our CLC mirrors' optical transmission-coefficient dependence on irradiation *time* is non-monotonous as well, although individual untwisting steps are in these graphs "blurred" by the experimentally necessitated spatial integration of the effect over the excitation beam's radial intensity distribution. However, by properly adjusting the driving pulses' pulse-pair separation relative to the CLCs' relaxation time constant, step-wise helix untwisting becomes possible: in the presence of each 500-ns pulse, orientation work is being carried out on the CLC helix dipoles, which subsequently dissipates again over the 200-μs "dark" time between pulses but only partially, as the helix's full initial order is recovered only within the ~1000-μs time constant. In this manner, the helix is being "ratcheted" into untwisting only over long periods ("pile-up effect" of the nonlinear change[13]).

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