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Super-twist generation and instabilities in photosensitive liquid crystal cells

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ABSTRACT

In planar nematic liquid crystals cells twist deformation was generated through photoalignment. By increasing the twist angle gradually, supertwisted cells were constructed in the range of 2π–3π twist angle. The supertwist relaxed through the formation of either π or 2π inversion loops, depending on the character of the photosensitive substrate. The difference in the relaxation process can be related to the zenithal anchoring strength on the photosensitive plate.

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1. Introduction

The phenomenon of photoalignment, i.e., the light-induced change of liquid crystal orientation on surfaces, was discovered in the early 1990-s by Yuriy Reznikov's group [1] and W. Gibbons [2,3]. For reversible photoalignment most often *trans-cis* isomerization of azo dyes [4] is exploited to control the surface director orientation by a polarized light beam. The azo dyes are either coated on the substrate as a molecular layer or embedded in a polymer film. When polarized irradiation is applied, light-induced conformational transitions from the *trans* to the *cis* state and back-relaxation to the *trans* conformer induces an orientational order of the azo-dyes. The order is transferred to the liquid crystal in contact with the photosensitive layer. With most azo-dyes the liquid crystal is aligned parallel to the substrate and perpendicularly to the light polarization.

In a typical experiment a liquid-crystal sandwich cell is constructed with one photosensitive substrate and one traditionally prepared "reference" plate, which ensures fixed planar orientation (e.g., a plate coated with polyimide and rubbed unidirectionally). Regarding the light irradiation, it can be either "direct", i.e., the light enters the cell through the photosensitive plate, or "reverse" when the light input is from the reference side [5].

Photoalignment provides the possibility to create twisted nematic liquid crystal layers, in which the director rotates from the reference substrate to the photosensitive one by an angle of Φ. Starting from a planar cell, it is straightforward to induce a twist angle in the range $-\pi/2 < \Phi < \pi/2$; the photosensitive plate should be irradiated with a light beam polarized by an angle between 0 and π with respect to the director alignment on the reference plate. Twist angles outside the range $-\pi/2 < \Phi < \pi/2$ can be also induced if the polarization direction is gradually increased, "winding" the nematic layer over the limit of $\pi/2$. The minima of the twist energy (Φ_m) always corresponds to a total rotation of the director across the cell between $-\pi/2$ and $\pi/2$, but the transition of the director configuration from $|\Phi| > \pi/2$ to the appropriate Φ_m cannot occur in a continuous way. In this manner metastable "supertwisted" structures can be created. The supertwist eventually relaxes back to the stable twist configuration through the formation of inversion walls.

The main subject of the present paper is the investigation of the light-induced supertwisted structures and in particular their relaxation process back to the stable twisted configuration. We compared two kinds of photosensitive substrates. In the first type a monolayer of an azo-dye was attached chemically to a glass plate. In the second type of photosensitive plate, the azo dye was incorporated in a polymer host as a side-chain. We found that in both types of cells the orientation followed almost exactly the direction perpendicular to the light polarization. However, the relaxation of the supertwist occurred through different types of inversion walls in the two cases, which indicates different mechanisms of relaxation. An essential difference was noticed also using the reverse geometry: in the case of azo monolayer light-induced instabilities were observed [5,6,7], which were not found in the case of the azo-functionalized polymer. In the paper we suggest that the difference in zenithal anchoring strength is responsible for

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the diverse behaviour observed in the presence of the two kinds of photoaligning layers.

2. Experimental details

2.1. Sample preparation

Two types of photosensitive plate were prepared. In the first one a silane derivative of the azo compound methyl-red was used (dMR, see Fig. 1a). The dye was chemically attached to a clean glass substrate and formed a monolayer [8]. For the second type, the polymer polymethyl-methacrilate was functionalized with the azo-dye Disperse Red 1 (pDR1, see Fig. 1b). The functionalized polymer was dissolved in toluene and spin-coated on the substrate. The basic photoorientation process for the two materials were described in [8,9], respectively.

The reference plates were rubbed polyimide coated slides from E.H.C. Co (Japan). The sample thickness were in the range 20–30 μm and were filled either with 4-cyano-4′-penthylbiphenyl (5CB) or with mixture E7 from Merck. Before filling the material, the cell was illuminated with white light, polarized perpendicularly to the rubbing direction. This procedure ensured a good quality planar initial alignment of the nematic liquid crystal.

2.2. Experimental setup

For qualitative observations a beam from a diode laser (wavelength 409 nm) was coupled into a polarizing microscope and irradiated the sample with fixed polarization. The stage was rotated slowly by hand while the sample was viewed in the microscope. For quantitative measurements the setup sketched in Fig. 2 was used. A beam from a DPSS laser (wavelength 457 nm) was converted to circular polarization and let through a slowly rotating polarizer (Fig. 2a). The light entered the cell from the photosensitive side (direct geometry). It was defocussed to a spot size around few mm. The rotation speed of the polarizer was chosen slow enough that the twist deformation in the cell could follow adiabatically the surface reorientation.

In order to detect the orientation on the photosensitive plate a polarized probe beam from a He—Ne laser (wavelength 633 nm) was sent through the cell, entering it at the reference plate (see Fig. 2b). The probe beam was positioned to the center of the pump beam. Behind the cell the probe beam crossed a polarizer (FRP), which was rotating with a frequency around 60–70 Hz. The intensity of the beam behind the polarizer was measured by the photodiode PHD, connected to a lock-in-amplifier (LA). In order to generate a reference signal for the lock-in amplifier, another signal, polarized in the same direction as the probe beam, was also sent through the FRP and its intensity was measured by the photodiode R_PHD. The lock-in amplifier provided the phase, θ, and the amplitude of the probe-beam signal A (Fig. 3).

In the Maguin limit of light propagation [10] $\Phi = \theta/2 + n\pi$ ($\theta = 0$ corresponds to a planar cell, n is an integer). The depolarization ratio, defined as $D = 1 - A/A_0$, where A_0 is the amplitude for the planar cell, indicates the deviation from the Maguin limit and can be used – in principle – to deduct the number of full turns of the director, n. Model calculations, based on de Vries theory [10] showed, however, that the depolarization ratio for a monochromatic light becomes an oscillating function of Φ for twist angles larger than about 2π. The depolarization is very sensitive to the sample thickness; changes of less than a micrometer can shift the maxima of the D vs. Φ curve considerably. This circumstance renders quite difficult to fit the measured curves to the theoretical ones. On the other hand, sharp changes in the detected depolarization ratio can be considered as a sign of a jump in the number of turns of the twisted structure. In the experiments we exploited this fact.

3. Results

In Fig. 4 an experimental result is shown with the dMR coated sample, filled with the liquid crystal E7. In the experiments the starting pump polarization was perpendicular to the director.

During the measurement the polarization direction of the irradiating light completed 5 full 360° turns; the rotation speed was 2°/s. If it is assumed that there are no discontinuities in the twist angle, one would find that $\Phi_{app} \approx \alpha$ during the whole period of rotation. Here α is the rotation angle of the polarizer, ranging from 0 to 1800° (see Fig. 4a). However, we observed sharp decreases in the depolarization ratio at certain times. These decreases occurred periodically; in the case of dMR it took place approximately at $\alpha \approx m \times 360^{\circ}$, with $m = 1, 2, 3\ldots$ We attribute the observed drops of the depolarization to the formation and expansion of inversion walls, which reduce the twist angle by 360° (Fig. 4b). The periodic change of the depolarization ratio is superimposed on a continuously increasing background. The background is probably due to the permanent formation of defects in the sample, which contribute to scattering and depolarization of the probe beam. We associate the reduction of the twist angle with the maxima of the depolarization ratio, in a somewhat arbitrary manner. The periodic repetition of the D curve, however, shows clearly the periodic reduction of the twist angle.

The result obtained for a sample with pDR1 layers is depicted in Fig. 5. As in the previous case, the „apparent" twist angle followed the rotation angle of the pump beam polarization. We observed similar periodic drops of the depolarization ratio. The first drop occured at $\alpha \approx 465^{\circ}$, a somewhat higher value than for dMR. The successive drops followed each other at every 180° rotation of the polarization, indicating the formation of π walls, in contrast with the 2π walls, observed with dMR layers. The minimal values of the depolarization ratio is 0.2, which indicates that the twist was reduced with a value of near 180°. The increase of the background value of D is much less pronounced than is the case of dMR.

An essential difference was found in the behaviour of the two types of cells after ending the irradiation. In the case of dMR-coated cell, the inversion walls moved to the edge of the laser spot, where they became stable for several months. The disclination loops surrounding the irradiated area were visible to the naked eye, due to scattering. On the other hand, in the pDR1 cells the inversion loops disappeared in few minutes after the irradiation finished. As revealed from observations in the

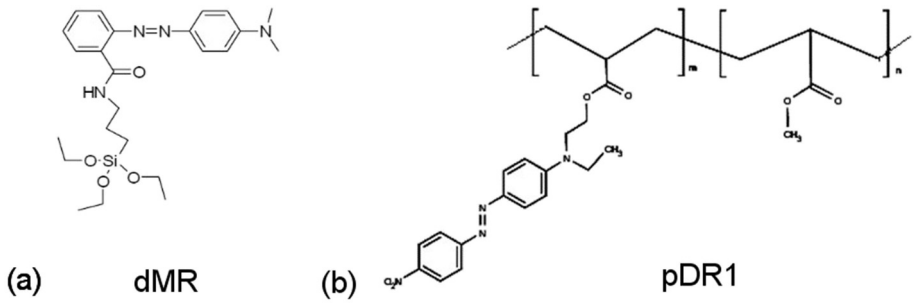

Fig. 1. (a) Structure of dMR (b) Structure of the azo-functionalized polymer, pDR1.

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