Electrically controlled surface diffraction gratings in nematic liquid crystals

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Photorefractive diffraction gratings were studied in cells of homeotropically aligned pentyl-cyanobiphenyl liquid crystal. These holographic gratings were induced by the simultaneous and nonsimultaneous application of dc and coherent optical electric fields. The observed behavior was consistent with a predominantly surface-mediated photorefractive effect. Beam coupling was observed in all cases and led to a model involving screened and unscreened interfacial trapped charges driving a modulation of the easy axis. Holographic gratings could be switched on and off by the application of a small voltage. © 2000 Optical Society of America

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Systems for information processing and storage require materials in which optical information can be recorded with high spatial resolution and high speed by use of low-power, inexpensive lasers.[1] Photorefractive materials are generally recognized to be the leading candidates for such applications.[2] Recently, observations of the photorefractive effect in nematic liquid crystals (LC's) were reported by several groups of researchers.[3–7] This effect is usually attributed to a light-induced modulation of electric charges in conjunction with an applied dc electric field, which produces a space charge in the bulk of the LC. This inhomogeneous bulk space-charge field was described as being due either to differential photocarrier diffusion in the LC's[5] or to a light-induced electrohydrodynamic instability.[7] In addition, Khoo[7] reported the recording of persistent photorefractive gratings. Khoo believed that these persistent gratings are due to a large perturbation of the surface director axis alignment by the current and nematic flows under the prolonged application of a dc field. This result implies that the anchoring properties of the aligning surface can be drastically changed during the grating recording and, in turn, that the aligning surfaces may play a crucial role in the photorefraction effect.

In this Letter we report two important results. First, new experiments indicate that the photorefractive effect in cells of homeotropically aligned LC's are mediated by surface charge rather than bulk currents. Here "surface" denotes the interface between the alignment layer and the LC material. We believe that this effect is due to a light-induced modulation of the easy axis caused by electric charges at the interface of the LC cell and the alignment layer. Second, the observed photorefractive effect is marked by the unusual property that the recorded grating can be hidden and then revealed by turning of a dc electric field off and then on. This procedure allows for low-voltage switching of the diffraction grating, which may be useful for diffractive beam steering. Cells of homeotropically aligned nematic materials have been well studied, yet it is remarkable that new chemical and physical effects at the interfaces have been revealed by an inhomogeneous optical probe. Thus holographic probes have revealed new phenomena and may become useful tools for studying surface and interfacial effects by inducing inhomogeneous surface properties that can be optically probed.

We studied the photorefractive effect in 12.5- and 25-μm-thick LC cells containing homeotropically aligned pentyl-cyanobiphenyl (5CB) from EM Industries. We used pure 5CB as well as 5CB doped with small amounts (0.01–0.05 wt.%) of fullerene (C_{60}). The results obtained for pure and doped 5CB were qualitatively the same, but doping made the cells more sensitive to the writing beams. Homeotropic alignment of the LC was provided by hexadecyl trimethyl ammonium bromide films that were deposited onto indium tin oxide–coated glass substrates. The cells were filled with LC in the isotropic phase and slowly cooled to room temperature. Homeotropic alignment was verified by examination in a polarizing microscope.

Phase gratings were recorded in the cells that were placed at the intersection of two Ar^+-laser beams. The total power of the light beams, P, was less then 30 mW. The diameter of the beams at the intersection was 1 mm. The period of the interference pattern, Λ, was in the range 20–75 μm. A dc or an ac field could be applied to the cell by application of a voltage $V_{\mathrm{dc,ac}}$. Raman–Nath diffraction was observed in all cases. The intensity of the transmitted beams, I_0, and the intensity of the first non-Bragg orders of self-diffraction, I_{-1}, were measured by photodiodes connected to a computer-controlled lock-in amplifier. We also measured the diffracted intensity I_p of a weak He–Ne laser probe beam from gratings recorded with an Ar^+ laser. Several experiments were carried out, as follows:

- Writing beams were turned on with no applied electric field. No self-diffraction or diffraction of the test beam was observed at any angle of incidence.
- A 2-V dc field was applied for 1 s, and then the dc circuit was opened. The writing beams were then turned on. Self-diffraction and beam coupling were observed.
- The writing beams were turned on for 1 s with no electric field applied and then were turned off. A 2-V dc field was then applied. Diffraction of the probe beam was observed.

The three results above suggest a surface-mediated photorefractive effect involving an unknown photo, electrical, and chemical process leading to charging at the interfaces. Current-induced bulk effects are ruled out.

The following experiment was then performed: A dc electric field was turned on, followed by the writing beams. At $V_{\mathrm{dc}} > 1.2$ V, self-diffraction of the writing beams was observed at oblique incidence only. The grating recording dynamics is shown in Fig. 1. When the dc field was then switched off, a sharp temporary increase in diffraction of both the probe and the recording beams followed and then disappeared, again as shown in Fig. 1. Despite this, a grating remained in the cell because, when a dc field was reapplied, the diffraction grating reappeared and then slowly disappeared. We could rejuvenate the grating diffraction by turning the dc field off for as long as several hours and then turning it back on. When a low-frequency electric field (0.1–0.5 Hz) was applied, the diffraction intensity appeared in phase with the maxima of the field.

No self-diffraction was observed at normal incidence of the writing beams, and the maximum I_{-1} was observed at an incident angle of ~45° to the cell normal. At the same time, the diffraction of the probe beam, I_p, was observed at any position of the cell, including normally incident recording beams (although it was considerably weaker).

Increasing V_{dc} increased the diffraction efficiency. In the vicinity of the electrohydrodynamic instability ($V_{\mathrm{dc}} \sim 3.5$ V) the intensity of the diffracted beams sharply increased, and strong scattering appeared.

To establish that these gratings are the same photorefractive gratings that were previously observed,[6,7] we studied two-beam coupling in the cell. The dynamics of the coupling of two beams of the same intensity is shown in Fig. 2. It can be seen that there is obvious asymmetric energy transfer from one beam to another, which indicates that the grating is photorefractive. The direction of the energy transfer was determined by the polarity of the dc field. The measured gain coefficient was approximately $\Gamma = 520\ \mathrm{cm}^{-1}$, which is typical for photorefraction in LC systems,[7] although we note that in the Raman–Nath regime such coefficients would not be descriptive as a figure of merit for applications.

To explain the experimental results we propose the following model: Irradiation of the cell with light intensity $I(x) = I_0(1 + \alpha \cos qx)$ causes a charge modulation at the aligning surface with the same spatial dependence:

$$\rho^{\pm}(x, z = 0) \sim (1 + \beta \cos qx),$$

where x is in the substrate plane and z is normal to it. The origins of this charge modulation could be, for example, photoinduced generation of charges followed by their spatial separation owing to different mobility of plus and minus ions[5] or photoinduced modulation of the conductivity of the LC and (or) aligning layer.

Suppose that the charges of one sign are trapped at the surface. Application of a dc field results in charge

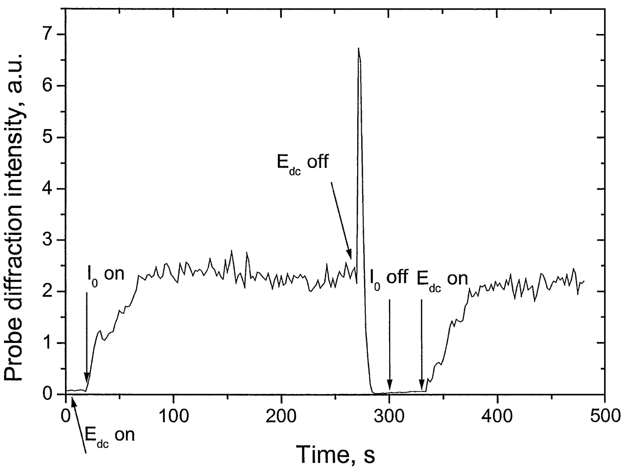

Fig. 1. Diffracted intensity (in arbitrary units) of a continuously incident He–Ne probe beam during a program of applying an electric field (E_{dc}) and a pair of Ar-laser writing beams (I_0) in the order indicated.

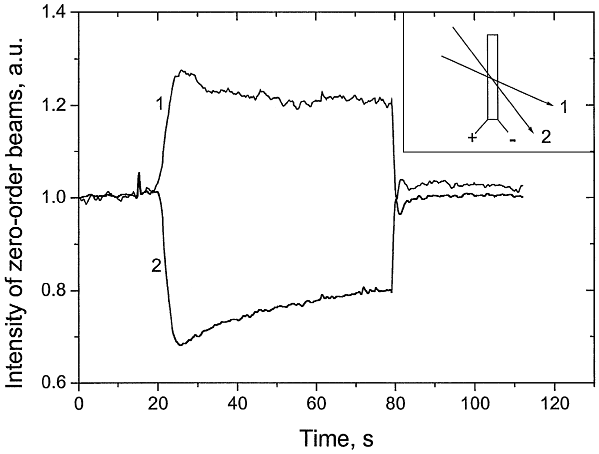

Fig. 2. Asymmetric two-beam coupling of the lowest diffraction order of Ar-laser writing beams in a homeotropically aligned LC cell. The experimental geometry is indicated in the inset.

separation and produces a surface-charge grating. This grating leads to the appearance of a modulated electric field:

$$E_x(x, z = 0) = E_\rho \sin qx ,$$

$$E_z(x, z = 0) = E_{\rho z} + E_\rho \cos qx .$$

The electric field may act on the LC in two ways. First, together with the dc field, it produces a driving force (torque) on the LC director in the cell bulk. The leading term of this torque is proportional to $E_{\mathrm{dc}} E_\rho \sin qx$.[8] Second, the electric field may cause a surface torque owing to modulation of the surface anchoring, and thereby a modulation of the easy axis. This modulation may appear as a result of a locally strong E_x component of the surface-charge field, $\theta(z = 0, x) \sim \sin qx$.

Both the volume and the surface torque result in a director deviation grating in the cell volume at any incident angle of the writing beams. The deviation of the director, $\theta \sim \sin qx$, is phase shifted from the interference pattern of the writing beams because it originates from a photoinduced charge modulation, as verified by the observation of two-beam coupling. When the dc field and the writing beams are switched off, the surface charges are compensated for by charges of the opposite sign accumulating either from the LC side or from the orienting layer side. This charge compensation hides the grating and results in the disappearance of diffraction. When the dc field was applied again, the hidden grating was revealed with a probe beam because the compensating charges drifted away from the interface. This result implies that the charge grating was fixed on the surface. The hidden grating was found to disappear naturally within a day.

Thus the experimental results can be qualitatively explained by either surface or bulk torque arising from charge modulation at the aligning surface. However, one can distinguish these two mechanisms by observing the characteristics of the grating decay when the dc field is switched off. The dc field acts in two different ways. First, it creates the driving force for the director grating, but it also prevents director deviation from the initial homeotropic orientation. If the volume torque were responsible for the observed effect, the director grating would disappear gradually because this torque is proportional to the product of dc and surface-charge fields, as described above. However, if surface torque or easy axis (anchoring) modulation were responsible for the effect, switching off the dc field would lead to a temporary enhancement of the grating because there is no stabilizing action of a dc field, whereas there is an easy axis grating at the beginning of the relaxation. Our data demonstrate this behavior (Fig. 1). Therefore we conclude that the second mechanism dominates in our experiments.

In summary, we have carried out experiments that indicate a surface-charge-mediated photorefractive effect in homeotropically aligned nematic LC cells. The surface charge creates a bulk grating by action of an anchoring modulation that causes the surface angular torque in the LC bulk. The recorded grating can be switched on and off by use of a dc field. Additional experiments are being carried out to further establish the nature of the surface mechanism, to elucidate the details of the photoelectrochemical process at the interface, and to stabilize the hidden grating. The observed low-voltage switching of the gratings may be useful in beam steering applications.

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