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Optically induced periodic structures in smectic-*C* liquid crystals

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We explore periodic structures of smectic-*C* (Sm*C*) liquid crystals, induced optically by a polarization grating. The studied cells contain a passive surface of rubbed polyimide and an active photosensitive substrate of azo-dye doped polyimide. In a nematic phase the director field can be periodic independent of the angle between the grating vector and the rubbing direction. In the Sm*A* phase periodic structure can be induced only by layer undulations. The Sm*C* behaves similarly to the nematic phase, but the director can rotate only on a cone, which results in a more complex geometry. The periodic pattern is superimposed with four different director and layer structures. In spite of the coexistence of the nonuniform structures the diffraction efficiency is better in the Sm*C*, than in the nematic phase.

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INTRODUCTION

Basically there are two fundamentally different mechanisms of inducing easy axis for liquid crystals by optical means. One way is to create direction selective cross-linking or selectively breaking up photosensitive polymer chains [1]. Another approach is to incorporate azo dyes into a polymer matrix [2] and to induce an anisotropic orientational distribution of the dye molecules by polarized light. Research has mainly focused on nematic liquid crystals, but recently photoalignment of smectic-*A* phase was studied too. It was shown that a periodic structure could be written into the smectic-*A* phase, provided that the polarization grating is perpendicular to the rubbing direction [3].

In this paper we extended these studies to the smectic-*C* (Sm*C*) phases, where the director is tilted with respect to the layer normal. In Sm*C* liquid crystals the director can move on a cone given by the tilt angle θ, therefore it is expected that this extra degree of freedom can facilitate the formation of a periodic structure. We studied 4-*n*-hexyloxyphenyl-4′-*n*-decyloxybenzoate having the phase sequence

$$Cr\,60\,^{\circ}\mathrm{C}(44\,^{\circ}\mathrm{C}\,\mathrm{Sm}B)\mathrm{Sm}C\,78\,^{\circ}\mathrm{C}\,\mathrm{Sm}A\,82\,^{\circ}\mathrm{C}\,N\,90.6\,^{\circ}\mathrm{C}\,I,$$

and compared the structures formed in the different phases under different geometries.

EXPERIMENTAL ASPECTS

We made cells containing a passive surface of rubbed polyimide and an active photosensitive substrate of azo-dye (Disperse Orange 3) doped polyamide. For irradiation, the 488 nm line of an argon laser was used. The pump beam was split into two parts with equal power. The beams were orthogonally polarized and brought together at the sample under an angle of 2°, the illumination was carried out at 94 °C in the isotropic phase of the liquid crystal. In this way a polarization grating was formed in which the polarization direction of the beam varied between 45 and −45 degrees with respect to the rubbing direction. The total power was around 200 mW and no focusing lens was used. The setup is shown in Fig. 1. The resulting periodic structures were observed under a polarizing microscope and probed with a He-Ne laser by measuring the intensity of the diffracted light peaks.

The textures, formed in the liquid-crystal phases after the optical alignment, depend on the angle α between the grating vector and the rubbing direction. We studied the two extreme conditions, i.e, when $\alpha=0$ and $\alpha=90°$. The textures and the corresponding director structures are summarized in Fig. 2 and Fig. 3 for $\alpha=0$ and $\alpha=90°$, respectively.

In all phases and in both geometries the periodicity (25.6 μm) is determined by the irradiation conditions. Textures of the nematic and smectic-*A* phases are similar to those observed previously [3]. In the nematic phase, periodic patterns can be written in both geometries, but the quality of the patterns is generally better for $\alpha=0$. In the active surface the extinction direction alternates by $\delta_N=\pm 15°$ with respect to the rubbing direction. In the smectic-*A* phase a periodic pattern is possible only when the grating vector is perpendicular to the rubbing direction ($\alpha=90°$) [3]. In this case the layers become undulated resulting in a modulation of the optical axis with respect to the rubbing direction by $\delta_{SA}=\pm 5°$.

In the smectic-*C* phase fairly regular stripe patterns appear in both geometrical arrangements. The temperature dependence of the modulation angle $\delta_{\mathrm{SC}}(T)$ is shown in Fig. 4. The behavior strongly resembles the typical temperature dependence of the director tilt angle θ in the vicinity of the Sm-*A*-Sm-*C* transition. The temperature dependence of the modulation angle is basically the same in both geometries, but the measurement error is much larger for the $\alpha=90°$ case. In addition to the stripes, zigzag defects appear, too (see Fig. 2). Under uncrossed polarizer-analyzer conditions greenish and reddish domains coexist. They are typically not separated by the zigzag defects. The colors of the domains

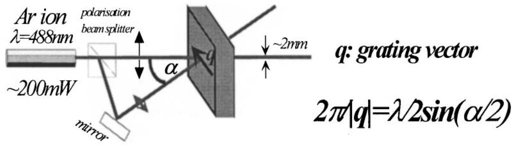

FIG. 1. Experimental setup to make polarization grating.

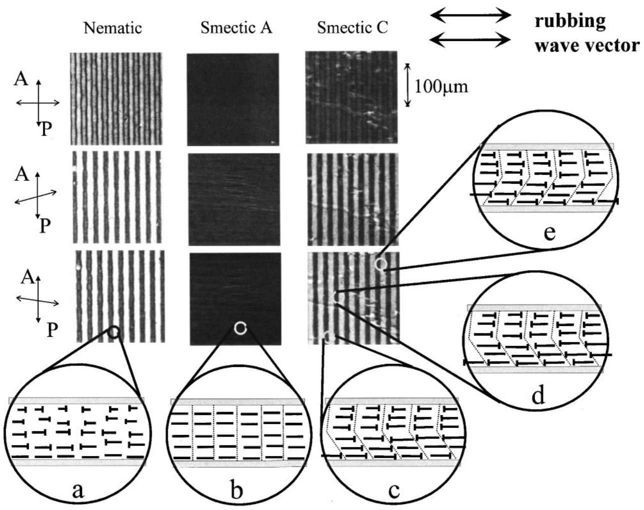

FIG. 2. Typical textures for $\alpha=0$ in the *N*, Sm*A*, and Sm*C* phases under different polarizer-analyzer configurations. Sketches within the circles are the proposed director structures. They represent front views of the areas of the small circles. Nails indicate directors tilted out of the plane of the cross section: the head of the nails are closer to the observer, shorter nails correspond to larger tilt angles.

interchange by reversing the uncrossing direction, and the difference disappears exactly when the contrast between the stripes vanishes. Another complication occurs after repeatedly heating the material to the isotropic phase and cooling back to the liquid-crystal states. We observed for $\alpha=0$ that the contrast between the subsequent stripes does not disappear exactly when the stripes are parallel to the crossed polarizers, but when they are rotated by a small angle ξ. After 4–5 heating-cooling cycles ξ increases up to about 5°, then remains constant. No periodic pattern survives upon cooling the material to the Sm*B* or to the crystal phases. The temperature dependence of the light intensity of the first diffraction spot is shown in Figs. 5(a) and 5(b) for $\alpha=0$ and $\alpha=90°$, respectively.

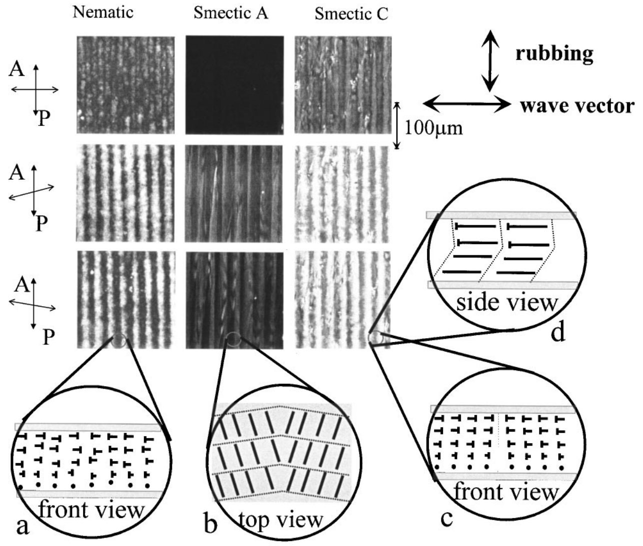

FIG. 3. Typical textures for $\alpha=90°$ in the *N*, Sm*A*, and Sm*C* phases under different polarizer-analyzer configurations. Sketches within the circles show the model structures deduced from polarizing microscopy. They represent the front or the top views of the areas of the small circles. Nails indicate directors tilted out of the plane of the cross section: the head of the nails are closer to the observer, shorter nails correspond to larger tilt angles.

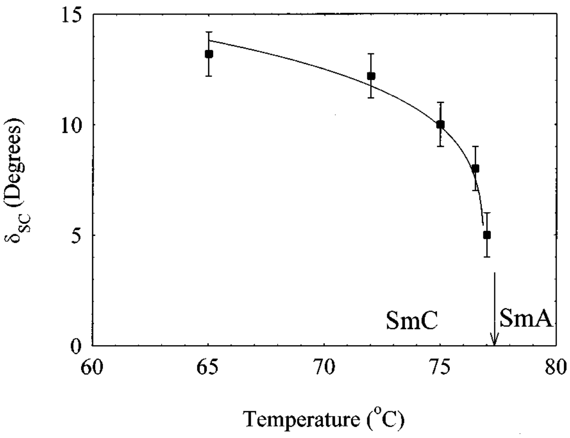

FIG. 4. Temperature dependence of the modulation angle in the Sm*C* phase as measured for $\alpha=0$.

These behaviors confirm the microscopic observations that there is no periodic pattern in the Sm*A* for $\alpha=0$, but there is one in the Sm*C* phase. The diffraction is more effective in the Sm*C* when cooled from the uniform smectic-*A* structure, than when it forms from undulated Sm*A* layers. Interestingly, for $\alpha=0$ the diffraction efficiency of the Sm*C* is even higher, i.e., the periodic structure is better than in the nematics. Another important feature is the behavior in heating and cooling. In the smectic phases the diffraction efficiency is basically reversible, whereas there is a large hysteresis in the nematic. Keeping the material in the fluid isotropic or nematic phase the original diffraction efficiency recovers slowly (in an hour or so).

DISCUSSION

Most of the observations are in agreement with the previous results concerning the nematic and smectic-*A* phases, and the same arguments [3] can be used for explanation. *In the nematic phase* periodic patterns can be written in both geometries because the director twist, splay, and bend are all allowed. Twist and bend are involved when $\alpha=0$, whereas twist and splay are needed when $\alpha=90°$. The quality of the patterns is generally better for $\alpha=0$, indicating that it is easier to induce a bend than a splay.

In the smectic-A phase, where splay is allowed, but bend and twist are forbidden, a periodic pattern is possible only when the grating vector is perpendicular to the rubbing direction. For $\alpha=90°$ the grating involves director splay and layer bend deformation [3], which is allowed, but for $\alpha=0$ a periodic pattern would lead a director bend and layer splay, which however is incompatible with the constant layer thickness requirement of the phase. The results concerning the *smectic-C phase* confirm the expectation that the director

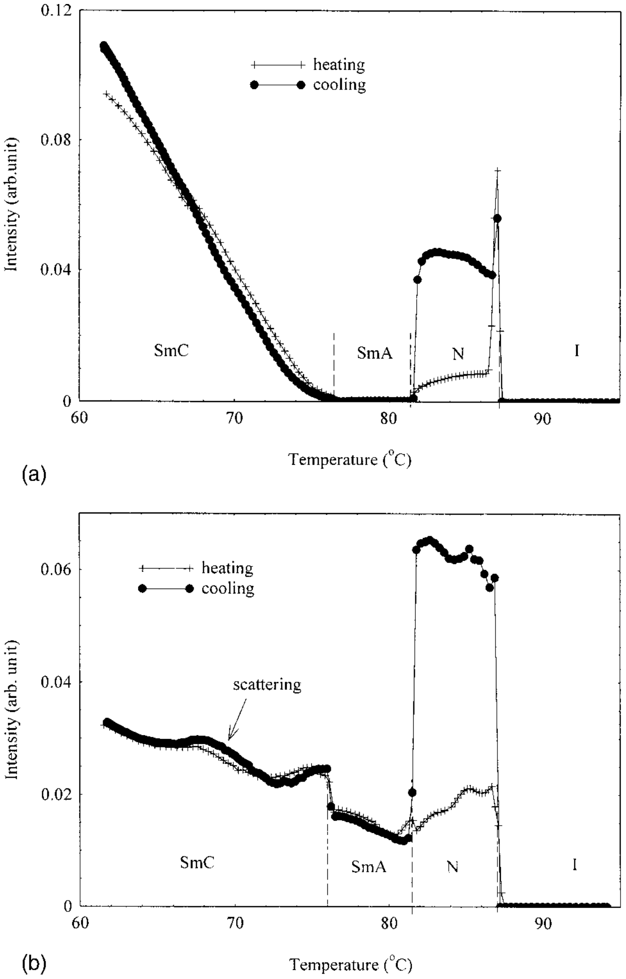

FIG. 5. Temperature dependences of the intensities in the first diffraction spot in two geometries. (a): rubbing is parallel to the wave vector; (b): rubbing is perpendicular to wave vector.

tilt enables to write periodic pattern in both geometries, just like in nematics. Due to the layer structures the director however can rotate only along a cone, which makes the situation more complex than in the nematic.

At the surfaces two factors determine the azimuthal angle of the director. (i) the surface anchoring pulls the director toward the easy axis (ii) the director tilt pushes away the director from the layer normal. Due to the different surface conditions in the active and passive substrates, the director and layer structures are different near to the two surfaces. In the active surface the easy axis is tilted with respect the layer normal by δ_N, which is larger than the director tilt θ, i.e., it facilitates the director tilting and keeps the layers vertical. In the rubbed polymer surface however the easy axis is almost parallel to the rubbing direction, i.e., to the layer normal. This counteracts with the director tilting and leads to a structure, where the layers are tilted with respect to the substrate normal by an angle β, and the director deviates form the rubbing direction by an angle γ. Accordingly the layer tilt angles differ at the different substrates, and the layer ''chevrons'' [4] are not symmetric. The layers are practically vertical near the active surface, but are tilted close to the rubbed polyimide. Such structures are shown in Figs. 2(c), 2(d), and 2(e) and in Fig. 3(d) for $\alpha=0$ and $\alpha=90°$, respectively. The magnitudes of β and γ depend on a number of parameters, like layer and director elastic constants and anchoring energies. Their signs however can be both positive and negative. We propose that the coexistence of the domains with $\pm\beta$ and $\pm\gamma$ result in the observed zigzag defects and the domains with different colors. The zigzag lines separate domains of oppositely tilted layers ($\geqslant$ and $\leqslant$), i.e., β changes sign as we cross a zigzag defect [see Figs. 2(c) and 2(e)]. In the domains with opposite colors the sign of γ, i.e., the angle between the easy axis and the director is opposite [compare Figs. 2(c) and 2(d)]. It is interesting that, in spite of these complications, the diffraction efficiency is even better in the Sm*C*, than in the *N* phase. This probably is due to the fact that the director fluctuations are considerably smaller at low temperatures.

Finally we have to comment on the increase of the angle ξ between the crossed polarizers and the stripes after repeatedly heating the material to the isotropic phase. A change of ξ in the smectic phases means a rotation of the smectic layers. We think that it is not in connection with the smectic-*C* phase, but is due to the so-called ''gliding'' of the easy axis. On the rubbed polyimide the easy axis is determined by the competition between the rubbing and the ''surface memory effect'' (SME) [5]. Due to SME a monolayer of liquid-crystal molecules are absorbed at the surfaces in the direction determined by the flow that occurs during the filling process. If the flow direction is not parallel to the rubbing direction the easy axis of the passive surface will also deviate from the rubbing direction. Heating the material to the isotropic phase we eventually ''erase'' [6] the surface memory, i.e., the easy axis eventually becomes parallel to the rubbing direction. During the cell preparation the grating vector was set parallel to the original easy axis, which deviated slightly from the rubbing direction.

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