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LENGTH SCALE DEPENDENCE OF CHIRAL SYMMETRY BREAKING IN FREE-STANDING FILMS OF ACHIRAL SMECTIC C

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The textures of a smectic C phase in free-standing films of the Schiff base 5O.6 are studied as a function of the number of layers and temperature. Periodic stripes with alternating contrast are observed in SmA boundary layers. On further cooling a transition to the splay stripes occurs in the smectic A temperature interval. In the smectic C phase all the periodic textures disappear, whereas in films with one free boundary a periodic arrangement of splay stripes is found. Mechanisms of symmetry breaking are discussed.

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The problem of chiral structures in achiral liquid-crystal films has attracted considerable attention of late [1–5]. Theoretically, such textures can exist due to two mechanisms [2, 5]. The first was proposed in Ref. [2] to explain the periodic textures in free-standing smectic films. This mechanism has a microscopic nature and is associated with coupling of the chiral order parameter ψ with the $2D$ bending deformation. It turns out that the homogeneous state of the smectic C films becomes unstable with respect to $1D$ or $2D$ periodic two-dimensional director field modulations with alternating sign of the chirality order parameter. The spontaneous formation of such structures on decreasing temperature has been observed recently [4]. The stripes observed in Refs. [1, 3] qualitatively correspond to the predictions of Ref. [2]. The stripe structure of Refs. [1, 3] is characterized by alternating contrast in a polarizing microscope.

The second type of chiral symmetry breaking has been observed in Langmuir [6–9] and SmC films with one free boundary [10]. This texture consists of the periodic array of equivalent splay stripes separated by sharp discontinuous walls. The chiral symmetry breaking in this case is a consequence of the head-tail asymmetry of the boundary layers due to the free surface.

The first mechanism is associated with breaking of the point symmetry of the layers and should therefore work on a short length scale. The second mechanism works for some assembly of polar molecules with some minimum radius ξ. In this paper we have found a nontrivial example of a substance for which both kinds of chiral symmetry breaking can take place in free-standing SmC films. We have carried out a detailed study of textures in free-standing SmC films of the Schiff base 5O.6 in relation to the number of layers and the temperature. The stripes with alternating contrast observed inside of the boundary layers of the SmA films are transformed to the splay stripes on decreasing temperature. In extremely thin

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films the splay stripes disappear, which gives us an estimate for ξ. After the phase transition SmA–SmC all the periodic structures are destroyed.

The Schiff base 5O.6 (4-n-hexyl-N-[4-n-pentyloxy-benzylidene]-aniline) has the following liquid-crystal phases in bulk samples (degrees centigrade): isotropic (73) N (61.4) SmA (52.8) SmC (51) SmB (43.4) SmF (40.8) SmG (36) Cr [11]. The temperature of the second order phase transition SmA–SmC has been determined microscopically to a precision of $\pm 0.3^\circ$ in samples with one free boundary and common cells between two cover slips.

The film textures were observed in a Leitz–Orthoplan polarizing microscope between slightly decrossed polarizers and registered photographically. The frame for the production of the free-standing films consisted of two brass rails and two movable brass blades. The number of smectic layers was determined by the multiple beam interferometry technique described in Ref. [12]. Films were produced in the smectic A phase and cooled down to the smectic C phase. Configurations of the plane director field in free-standing films of 5O.6 were studied over a broad interval of number of layers (from 5 to 200).

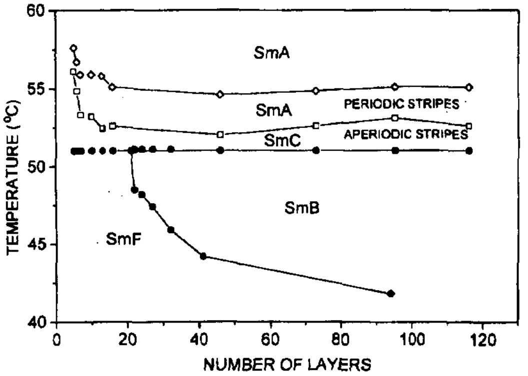

Fig.1. Phase diagram of 5O.6

Fig.1 shows a diagram of phases and textures of 5O.6 for different temperatures and number of layers. Above the SmA–SmC transition temperature we observed stripes with alternating contrast (Fig.2*a*). This state is characterized by a periodicity of about 6 μm which is independent of the number of layers. In extremely thin films with $N \leq 16$ layers the stripes with alternating contrast disappear after the SmA–SmC phase transition, and an aperiodic configuration of domain walls appears (Fig.2*b*). The phase transition SmA–SmC was registered at the temperature of noticeable improvement of the image contrast.

In thick SmA films two types of periodic stripes were observed. On decreasing temperature in thick films the alternating stripes are transformed into a periodic array of splay stripes with a periodicity of about 14 μm (Fig.2*c*). The image in Fig.2*c* is analogous to the splay stripes observed in samples with one free boundary, where the smectic layers were oriented parallel to the substrate (see the inset in Fig.2*c*) [10]. On further cooling the phase transition SmA–SmC occurs, which was registered as a distinct change of the image contrast. The SmA–SmC transition temperature measured in such a way correlates with measurements reported by other groups, in which it was determined by independent methods [11]. At the SmA–SmC transition the periodic stripes disappear and some aperiodic texture of defect walls occurs (Fig.2*d*). It is interesting to note that the director field configuration between the walls is similar to the splay stripes of Fig.2*c*. The other feature of Fig.2*d* is that splay and alternating stripes are present at the same temperature. All the reported texture changes were observed reversibly on heating and cooling. The temperature of formation of the alternating stripes, the SmA – SmC and SmF – SmB transitions, are shifted to higher temperatures as the number of layers is decreased, as is usual for free-standing liquid-crystal films [13]. The temperature interval of the periodic stripes decreases from 2.2 to 1.5°C as the number of layers is decreased. The smectic B phase disappears in films thinner than 21 layers.

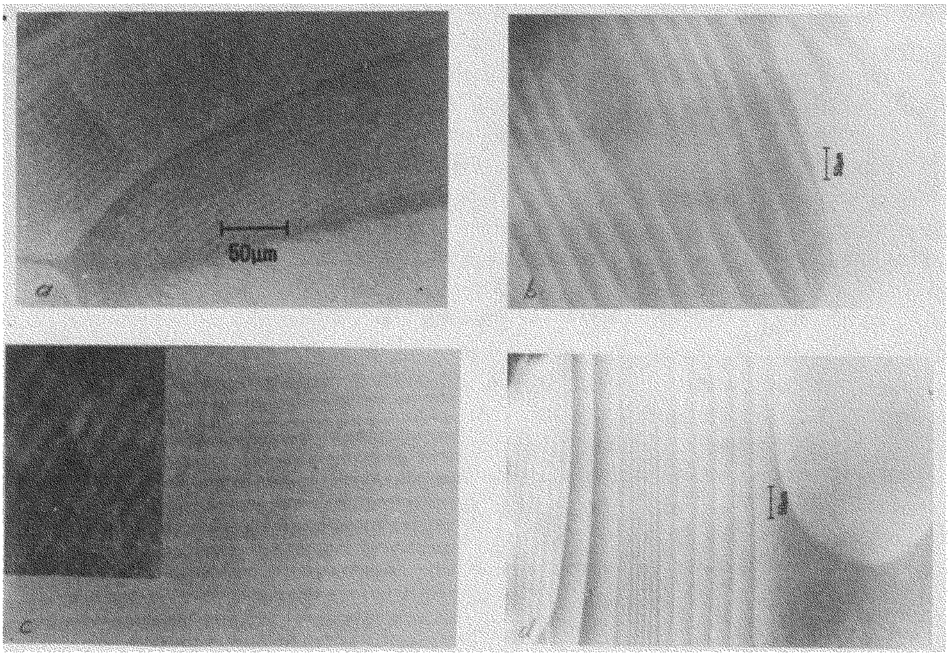

Fig.2 *a*) Stripe state with alternating contrast $N = 5$, $T = 57°\mathrm{C}$; *b*) aperiodic stripes, $N = 5$; $T = 54.6°\mathrm{C}$; c) Splay stripe state, $N = 29$; $T = 53.27°\mathrm{C}$; the magnification is the same as in Fig.2*a*; inset: splay stripes in a film with one free boundary, where the smectic planes are oriented parallel to the substrate [10], $T = 51.5°\mathrm{C}$; *d*) aperiodic stripes, $N = 29$; $T = 51.16°\mathrm{C}$

The alternating-stripes texture is analogous to the observations of Maclennan et al. [3], whereas the splay texture is similar to the so-called “lines” of Ref. [3]. At the same time the general properties of the 5O.6 textures in the smectic C phase are qualitatively different from those in Refs. [1, 3]. First of all, we definitively observe the modulated textures only in the temperature interval of the smectic A phase. It is well known that the exterior layers of the smectic A phase can be tilted [14]. Therefore the periodic stripe textures occur in tilted layers at the SmA boundary. A nontrivial observation is the transformation from the alternating to splay stripes on cooling. This finding is opposite to that of Ref. [3], where the reverse sequence of textures (lines to stripes) on cooling has been reported. The difference is that the authors of Ref. [3] observed the periodic textures in the smectic C phase, whereas we have found them inside of boundary layers on smectic A films.

The occurrence of the alternating stripes can be qualitatively explained by the microscopic mechanism of chiral symmetry breaking proposed in Refs. [2] and [4].

Stripes of this kind are completely analogous to Refs.[1, 3], where it was shown that the stripes are always bent, and therefore the chiral breaking mechanism of the first type works. If the model of Ref. [2] is true, the structure of the boundary should correspond to the SmL phase. The boundary region in which the molecules are tilted grows with decreasing temperature. Our results show that the splay stripes occur when the penetration depth of the surface order reaches some threshold value which should be equal to ξ. This conclusion is supported by two facts: i) the transition from alternating stripes to splay stripes at lower temperatures in the SmA interval; ii) the disappearance of the transition from alternating to splay stripes in extremely thin films ($N \leq 16$). This gives us the estimate $\xi \sim 8$ layers. The penetration length necessary for the first mechanism to work equals 1–3 layers in the case of 5O.6, since the alternating stripes are still seen in the 5-layer film.

Both kinds of stripes are destroyed after the SmA – SmC phase transtion. In thick films this can happen because a periodic arrangement with a high density of defects is inconsistent with the stable 3D configuration of the smectic C phase. The disapperance of the periodic stripes goes along with the coarsening of elongated domains and the diminishing of the number of defect walls. The disapperance of stripes in ultrathin films can be explained by the symmetry of the films as a whole, which constrains the divergence terms like $\nabla \cdot c$ in the free energy, and the second mechanism of chiral symmetry breaking becomes impossible.

The following questions seem to be interesting for the future development of this work: i) why are the splay stripes more stable than the alternating stripes at lower temperatures? Because the opposite type of behavior is possible [1, 3], it is important to know which parameters govern each type of chiral symmetry breaking. ii) The second mechanism is associated with the onset of surface polarization, the role of which has to be studied. This is interesting from the point of view of recent investigations of periodic textures in chiral SmC* films with high spontaneous polarization [15 – 17].

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