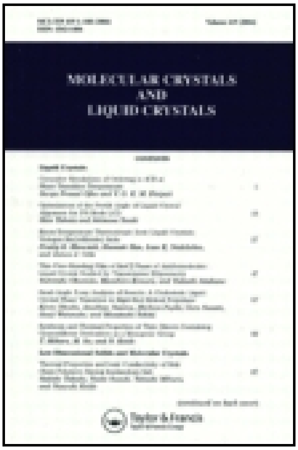


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# Observations on a Nematic Liquid Crystal with an Oblique Orientation of the Director at the Nematic-Isotropic Interface

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# Observations on a Nematic Liquid Crystal with an Oblique Orientation of the Director at the Nematic-Isotropic Interface†

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We report some observations on the nematic isotropic (NI) interface of 4′-heptyl-4-cyanobiphenyl (7CB), 4-cyanophenyl-*trans*-4′-n-heptylcyclohexane (PCH-7) and a mixture of PCH-5 with a nonmesomorphic compound. In the mixture the director makes a tilt angle ≃45° at the NI interface leading to a new structure of nematic drops floating in the isotropic phase. The structure is characterised by two point defects at the poles and a line deflect at the equator. When thin films of this mixture and of 7CB are spread on a slide treated for homeotropic alignment, the NI interface develops a regular network of point defects of strength ± 1 arranged in a square lattice. It is associated with conical distortions in the surface profile, which are generated to lower the elastic energy of the sample as was predicted by de Gennes. The conical distortions can be directly seen at the NI interface of a PCH-7 sample mounted in a hole in a coverslip.

## INTRODUCTION

The NI interface has been a subject of several studies in the recent past. The early investigations[1–7] were concerned with the influence of the boundary condition of the director ($\vec{n}$) at the interface on the director configuration in small nematic drops floating in the isotropic phase. Specifically, a normal boundary condition leads to a 'star' configuration in the one elastic constant approximation.[1] Elastic anisotropy leads to distortions in this structure,[5–7] especially close to

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the centre of the drop. On the other hand, a tangential boundary condition at the interface leads to a 'bipolar' configuration with a point defect at each pole. In general the tilt angle $\theta_t$ of the director at the NI interface can take any value $0 \leq \theta_t \leq 90°$. There have been measurements of $\theta_t$ in a few cases. $\theta_t = 90°$ in N-p-methoxybenzyl-idene-p'-butylaniline (MBBA), i.e. it has a tangential boundary condition.[8] $\theta_t \simeq 61.9°$ in 5CB[9] and 52° in 7CB.[10]

Another interesting problem connected with the NI interface is the coupling between its shape and elastic distortions of the director field in the bulk nematic. de Gennes[11,12] developed the theory of such a coupling in the case of free surface of a nematic, i.e., the nematic-air interface, the distortion in the director field being produced by an alignment of $\hat{n}$ in the bulk by a suitable magnetic field. The origin of this coupling can be easily understood. In view of the observations to be reported in the next section, we consider a nematic film of thickness $d$ spread on a glass plate treated for homeotropic alignment (Figure 1a). If the tilt angle $\theta_t$ at the nematic-air interface $\neq 0$, the orientation of the director is not uniform in the nematic film. The corresponding elastic energy can be lowered by a tilting of the interface (Figure 1b) so that the director configuration is less distorted. Assuming, for the time being, that the director is confined to the $XZ$ plane, the interface is tilted by an angle $d\zeta/dx$ where $\zeta$ is the height of the interface with respect to the unperturbed level (Figure 1b). The tilting of the interface increases the surface area and hence the surface energy. Further, the non-uniformity of the height also increases the gravitational energy of the nematic film. The $\zeta$-dependent part of the net energy is given by,[11]

$$f_s = \int \left[ \frac{A}{2} \left| \nabla \zeta \right|^2 - \frac{K\theta_t}{d} \left| \nabla \zeta \right| + \frac{1}{2} \rho g \zeta^2 \right] dx\, dy \qquad (1)$$

where only the leading terms are retained and $A$ is the inter-facial tension, $\rho$ the density, $g$ the acceleration due to gravity and $K$ the curvature elastic constant in the 'one constant' approximation ($K = K_{11} = K_{33}$).

It is clear from eq. (1) that though tilting is favoured by the elastic term, the height cannot indefinitely increase due to the gravitational term. However if the interfaces of neighbouring regions tilt in opposite directions as shown in Figure 1b and 1c, $\zeta$ need not become very high and the system can gain in net energy by such an arrangement. When two oppositely tilted regions meet, either cusps or dips are formed and the director distribution around them gives rise to

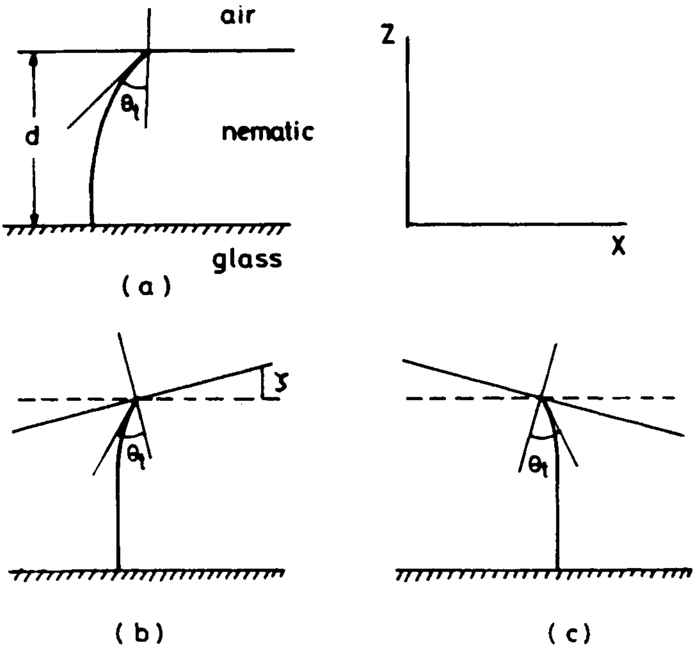



FIGURE 1 A nematic film with different orientations of the director at the two boundaries has a distorted director configuration (a). The associated elastic energy can be lowered by a tilting of the nematic-air interface (solid line in b). Neighbouring regions (b and c) have opposite tilts so that $\zeta$ does not become too large.

disclinations along the cusps and dips. Williams has observed a few broad domains at the free surface of paraazoxyanisole[13] and MBBA[14] when a sufficiently strong horizontal magnetic field was applied across the sample and hence favouring a horizontal orientation of $\vec{n}$ far below the surface. This observation has been interpreted by de Gennes[11,12] as due to the occurrence of surface disclinations.

In the geometry considered in Figure 1, there is no preferred azimuthal orientation of the director in the $xy$ plane. In such a case the surface can be distorted in two dimensions leading to the formation of conical cusps and dips which are associated with disclination *points* of strength $+1$. If there are a large number of such defects, they could be expected to arrange themselves in a regular network. de Gennes predicted that they would be arranged in a square lattice as shown in Figure 2. Indeed simple geometrical considerations show that only such a lattice is compatible with the alternate dips and elevations on the surface. Regular networks of point disclinations have not been found so far on the nematic free surface.

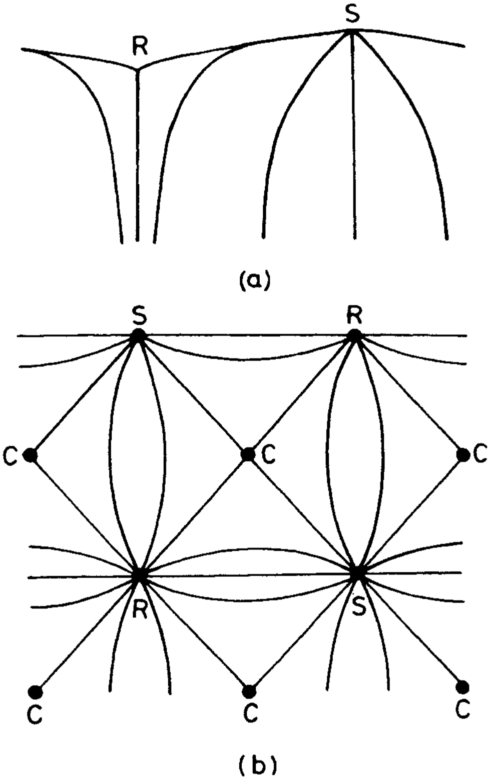



FIGURE 2 (a) A possible arrangement of the director in the sample. (Notice that the director is inclined obliquely with respect to the interface.) (b) A regular network of defects as visualized by de Gennes.[11,12] The lines are projections of the director at the interface. R and S correspond to +1 disclinations and C to −1 disclinations.

For a typical nematic liquid crystal like 5CB, the surface tension $A \simeq 30$ dyn/cm[15] and the density is $\simeq 1$ gm/cc.[16] On the other hand the interfacial tension at the NI interface is $\simeq 10^{-2}$ dyne/cm,[17,8] and the density jump between the nematic and isotropic phase is $\simeq 10^{-3}$ gm/cc.[16] Hence compared to the nematic free surface both the terms in eq. (1) which would disfavor tilting of the interface are weaker by three orders of magnitude for the nematic-isotropic interface. Point disclinations at the NI interface have been studied by Meyer.[4] We recently found regular networks of point disclinations at the NI interface of a mixture of a nematogen with a nonmesomorphic compound.[18] In this paper we report further observations on the networks supporting the structure proposed. We have also found the networks in some pure nematogens. In addition, observations are also reported in a new geometry which leads to a direct visualisation of the deformation of the NI interface.

## OBSERVATIONS AND DISCUSSION

Our first observations were made on a mixture of PCH-5 with ≈10% of n-heptyl cyanide which is a nonmesomorphic compound. The advantage of such a mixture is that the nematic and isotropic phases coexist over a considerable range of temperatures and hence it is relatively easy to observe the NI interface. By careful observations on nematic drops we could conclude that the director makes an angle of ≈45° with the interface.[19] Further, this boundary condition leads to a new structure of the nematic drops. It is characterized by two point defects at the poles and a line defect at the equator (Figure 3).

For making observations on the network of surface disclination points, the sample was spread on a glass slide treated for homeotropic alignment (cleaned with chromic acid only). The thickness of the film was ≈10μ (fixed using mylar spacers). A Mettler FP52 hot stage was used for temperature control and the observations were made using a Leitz Ortholux II Pol BK polarizing microscope.

A typical pattern observed between crossed polarizers in shown in Figure 4. The striking resemblance between this pattern and the one predicted by de Gennes (Figure 2) is immediately obvious. Indeed we have now found similar defect networks at the NI interface of pure 7CB and PCH-7. The pattern found in the case of the latter compound is shown in Figure 5. It is also clear from this photograph that one set of alternate +1 defects are defocussed while those corresponding to the other set alternate +1 defects are focussed. This is clearly due to a lens action, the former set of points acting like concave lenses while the latter act like convex lenses. Indeed with the director orientation under the $S$ and $R$ points being as shown in Figure 2a, it is clear that the effective refractive index for light propagating close to these points in the $N$ phase should be lower than that for the isotropic phase, whatever be the polarization of the incident beam. Consequently the elevations ($S$) are defocussed and the depressions ($R$) are focussed and this observation provides another supporting evidence for such a break-up of the interface. Observation between crossed circular polarizers also brings out the difference in the focus between the two sets of points (Figure 6). Another interesting observation is shown in Figures 7a and 7b. Between crossed *elliptic* polarizers dark brushes can occur either along SCS or RCR

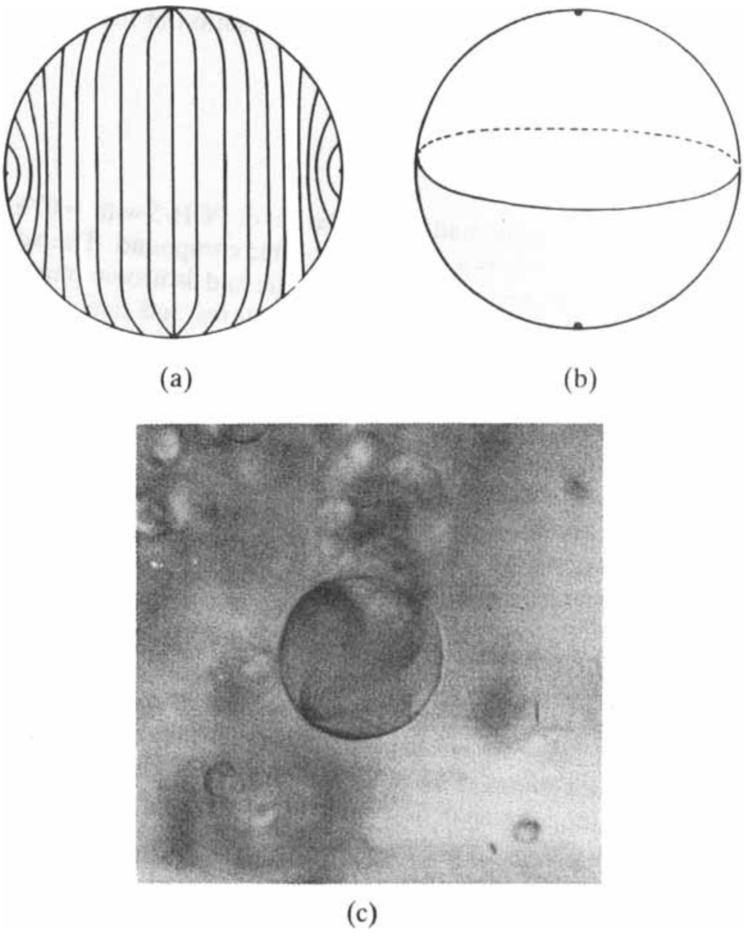


FIGURE 3 (a) Schematic diagram of the director orientation pattern in a principal section of a nematic droplet with tilted boundary condition. This configuration leads to two polar point defects and an equitorial line defect as shown in (b). (c) A spherical droplet showing one of the polar point defects and a part of the equatorial defect. Magnification x800.

(see Figure 2) depending on the ellipticity and azimuth of the incident beam. This can either mean that the light beam is incident at an oblique angle to the sample or that the director distribution is more complicated than that depicted in Figure 2, probably involving a twist deformation also. We may note here that Yokoyama et al.[17] have found evidence for a twist deformation in the undulation structure

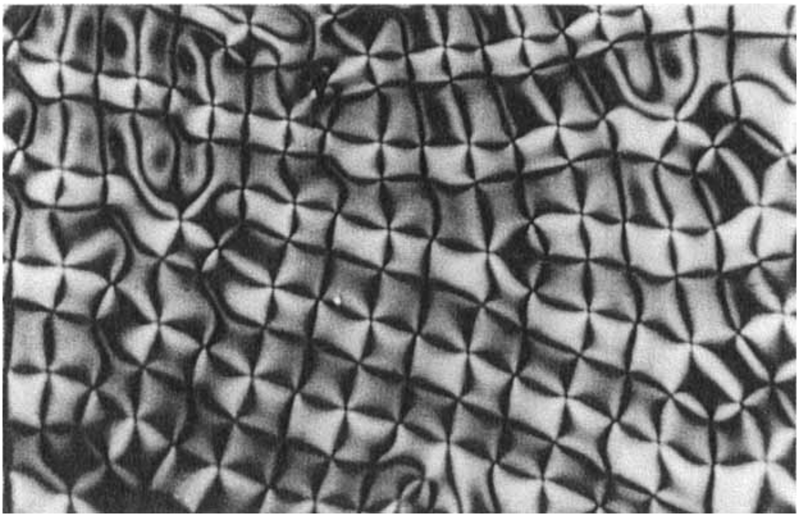


FIGURE 4 A regular network of point disclinations at the NI interface of PCH5 with 10% n-heptyl cyanide. Note the similarity with Figure 2b. Crossed polarizers; Magnification x350.

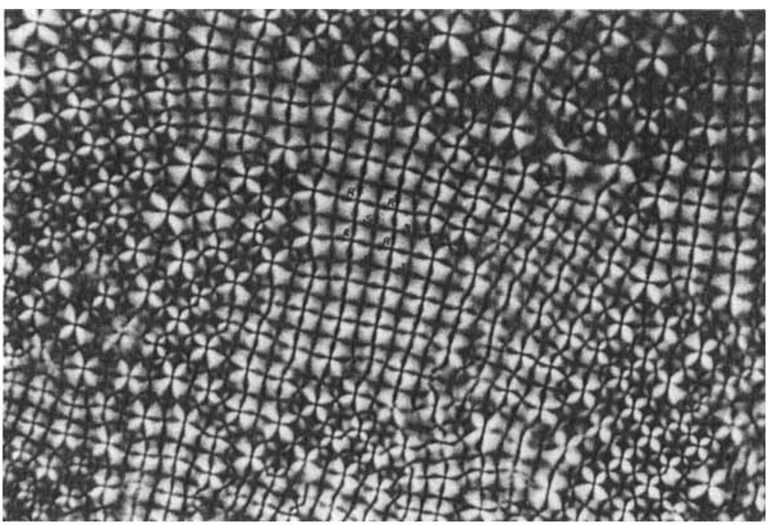


FIGURE 5 A regular network of point disclinations in PCH 7 showing the square lattice. Notice focussing and defocussing effects at points corresponding to *R* and *S* respectively. Crossed linear polarizers; Maginification ×175.

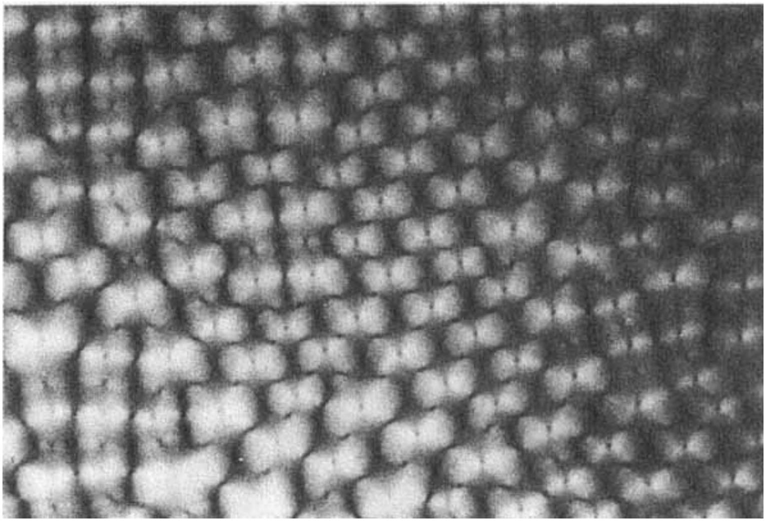
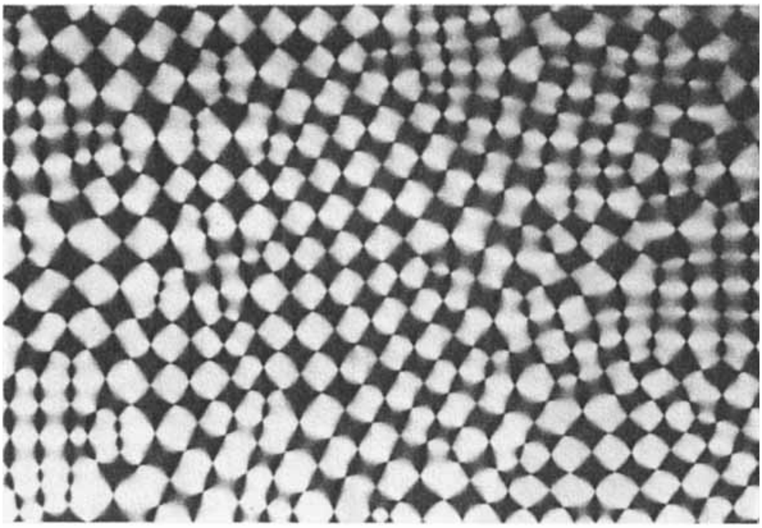


FIGURE 6 Same sample in Figure 5 between crossed circular polarizers; Magnification ×175.

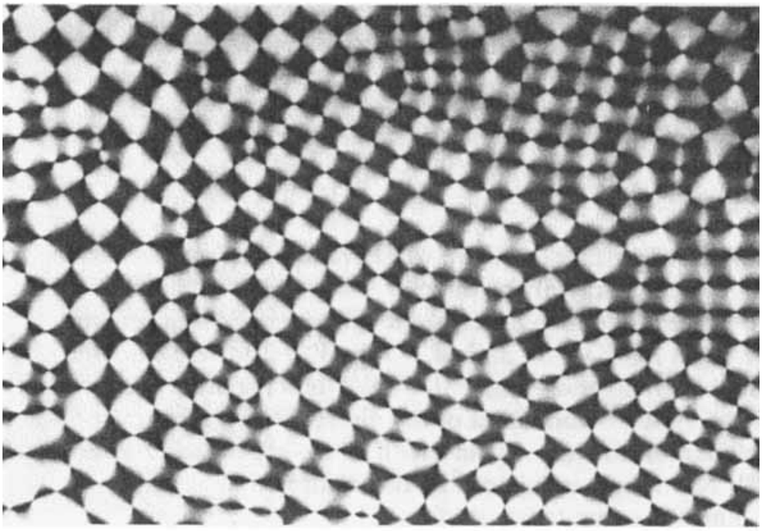
at the NI interface of 5CB, produced by an external electric field acting normal to the interface.

It would be interesting to see the rugged nature of the interface directly by making observations in a transverse direction. The sample was taken in a new geometry for this purpose. A hole of diameter ≃1mm was drilled in a microscope coverslip whose thickness is ≃120μ. A film of PCH-7 is mounted in the hole and cooled slowly from the isotropic phase. The nematic phase starts forming from the center in such a hole due to the natural temperature gradient in such a geometry. The director is oriented practically normal to the nematic-air interface and in favourable cases the NI interface clearly breaks up into alternate conical elevations and depressions (Figure 8). In this case the conical spikes have a height of ≃50μ and the angle of the cone is ≃10°, and occur at two different levels in the thickness of the liquid film. This demonstrates *directly* the break up of the NI interface to minimise the elastic energy of the sample.

In conclusion, we have given in this paper both indirect and direct evidence for the break up of the NI interface into a rugged pattern. It is appropriate here to point out that recently Faetti and Palleschi[20] have inferred the presence of irregularly spaced surface disclination lines at the NI interface of 7CB, by studying the reflection of a laser from such a surface. As they have pointed out a quantitative descrip-



(a)



(b)

FIGURE 7 A sample of PCH 7 between crossed elliptic polarizers. The dark brushes occur either along (a) SCS. . . . or (b) RCR depending on the ellipticity and azimuth of the incident beam. Polarizer at (a) 37° and (b) 53° with respect to the $\lambda/4$ plate. Magnification $\times 175$.

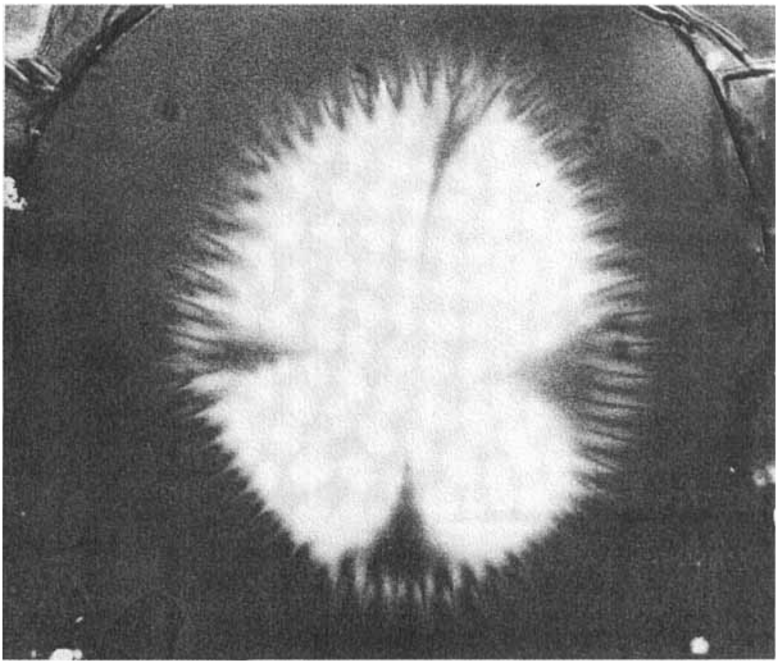


FIGURE 8 Picture of PCH 7 sample mounted in a hole in a coverslip. The nematic drop is surrounded by the isotropic phase in the lateral direction and air in the vertical direction. Notice the breaking of the NI interface into spikes. Magnification ×125.

tion of the surface structure will have to take into account the finite anchoring energy of the director for the tilted orientation at the NI interface. Assuming as usual the anchoring energy per unit area to be given by $W_S \simeq W \operatorname{Sin}^2(\theta - \theta_t)$, $W \sim 10^{-3}$ erg/cm$^2$ (see ref. 10) or the extrapolation length[12] $L = K/W$ is of the order of a few microns. If the elastic deformation of the director takes place over a distance comparable to $L$ (i.e. in our case if $d \leqslant L$) the tilt angle at the interface will be strongly influenced by the deformation.

Further quantitative studies on the interfacial properties are in progress.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