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Visualization of the cholesteric texture near a Grandjean line

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The twisted structure near a Grandjean line in a cholesterically ordered liquid crystalline network is visualized by scanning electron microscopy. It is shown that the formation of an extra π -rotation in the molecular helix at a Grandjean line occurs at a localized depth in the sample and gives rise to an edge dislocation of strength $\frac{1}{2}$, dissociated into a pair of two simple line dislocations.

The fixation of the mesomorphic order in a macroscopically oriented liquid crystalline (LC) diacrylate by means of *in situ* photopolymerization has been extensively studied [1-3]. The molecular organization of the monomer as induced by known techniques like orienting layers, external electric or magnetic fields or flow, is maintained during the network formation. The macroscopic ordering in the resulting polymeric film is insensitive to temperature variations. Besides nematic and smectic networks, cholesterically ordered networks are produced by adding an adjusted amount of chiral dopant to the monomer [4, 5].

The various types of frozen-in molecular orientation in such an LC polymeric network have been directly visualized by scanning electron microscopy (SEM) [6]. SEM photographs of a cryo-fractured surface of twisted and tilted foils reveal the change in mean molecular orientation as a function of height through a layered texture perpendicular to the film surface. This layered texture was interpreted to be parallel to the director. In the particular case of a cholesteric polymer film with a natural pitch smaller than the sample thickness, one observes the twisted molecular orientation as a sequence of bands, pointing to subsequent layers in which the director lies along and across the fracture surface, respectively. The origin of this layered texture is not yet fully clarified, but this is beyond the scope of the present paper. It clearly must be sought in the dependence of the mechanical properties on the local molecular orientation, resulting in a fractured surface that reflects the molecular order.

Grandjean had already demonstrated in 1921 that a cholesteric LC material in a wedge-shaped cell with boundary constraints exhibits disclination lines at regular distances [7]. These Grandjean lines occur at those cell thicknesses where the interaction between natural pitch and fixed boundary orientation forces the total molecular twist to increase with π , namely, where the elongated pitch of the band with a twist of $n\pi$ equals the contracted pitch of the band with a twist of $(n+1)\pi$ [8]. In its simplicity, this model assumes a uniform layer structure at both sides of the disclination

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line as illustrated by Kassubek and Meier [9]. In later work the nature of the edge dislocation at a Grandjean line was further investigated [10, 11] and it was claimed to be a line of strength $\frac{1}{2}$ in thin wedges, while in thicker wedges, dislocations of strength 1 were found as well. Whether or not dissociation of the Grandjean lines of strength $\frac{1}{2}$ took place was not proven. Therefore, de Gennes [10] proposed at that time the experiment of polymerizing a cholesterically ordered LC material in a Cano wedge cell for visualization of the molecular ordering by electron microscopy, an experiment which has now been performed after about 20 years and is described in this note.

For this particular experiment we used the LC diacrylate denoted as **2a** in [4]. Its natural pitch, adjusted at $10\ \mu\text{m}$ by adding 1 wt% of chiral dopant **1** [4] to the nematic diacrylate, was determined by the Cano method [12] using a wedge cell, consisting of a glass sphere and a flat glass substrate, both having a parallel rubbed polyimide orientation layer. The polymerization of the cholesteric monomer mixture was performed in this wedge at 100°C by a short exposure to UV.

Figure 1 shows a photograph of the Grandjean lines in a part of the polymeric sample as observed with a polarization microscope. The first band in the bottom left corner of the photograph corresponds to a total molecular twist of 10π , while the molecular twist in the band near the upper right corner equals 7π . The disclination between the bands with a twist of 10π and 9π , in the reproduction of the photograph may not however be too clear. The SEM photograph of this wedge-shaped cholesteric film cryo-fractured along the bar indicated in figure 1, is shown in figure 2. One again observes the banded texture along the height of the sample with a period equal to the distance over which the mean molecular orientation rotates through an angle π . The

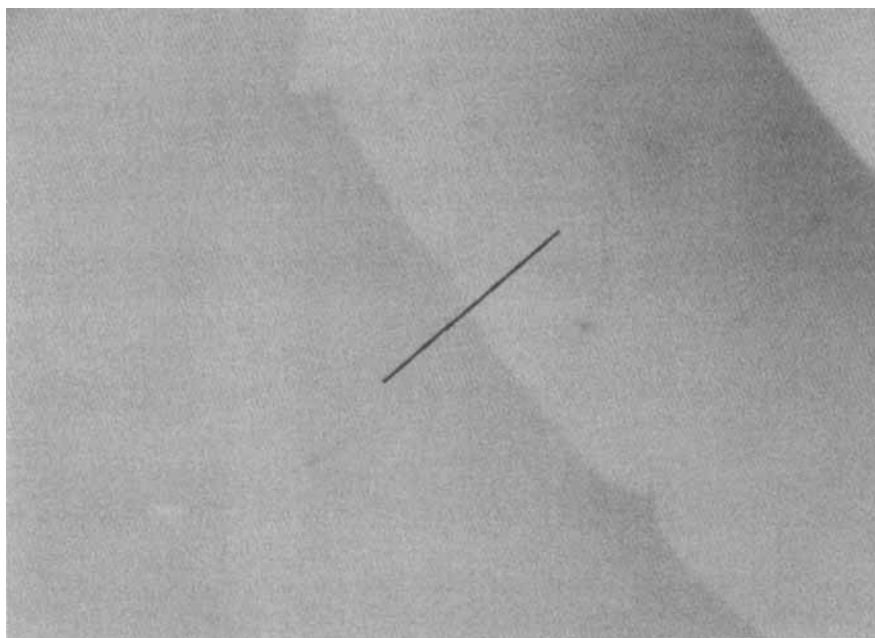


Figure 1. Segment of the circular Grandjean–Cano disclination lines observed through a polarizing microscope with crossed polarizers. The bar indicates the position of the cross-section used for the SEM observation.

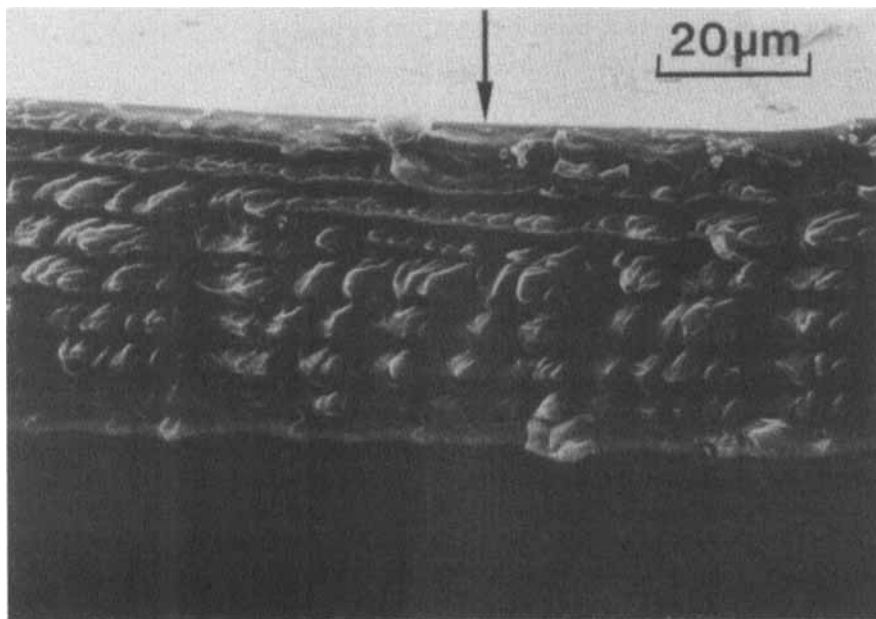


Figure 2. SEM photograph of the cross-section indicated in figure 1. The position of the Grandjean-Cano disclination line is located in line with the arrow.

number of bands, however, and consequently also the total molecular twist, is different at both sides of the photograph, being 8 at the right hand side and 9 at the left hand side. The arrow indicates the sudden formation of a new band in the bulk of the sample, at a location corresponding to the position of the Grandjean line observed in a top view using crossed polarizers. In the particular case shown here, the band decoupling occurs near the central section of the sample, but we also found evidence of band splittings near both substrates. In all our SEM observations, a textured white band with a mean molecular orientation perpendicular to the fracture surface is split up in two new bands and there is no evidence of a splitting of an intermediate black band with a mean molecular orientation parallel to the fracture surface.

From these results it is obvious that the formation of a Grandjean line is not completely described by the simple model of a layered structure resulting from an elongated and compressed pitch with an equidistant period equal to the thickness divided by the number of π -rotations at both sides of the Grandjean line, as suggested in [9] and illustrated in figure 3 (a). The creation of an additional band at a Grandjean line clearly occurs on a local level in the cross-section causing only locally an elongation or contraction of the pitch, as illustrated schematically in figures 3 (b) and (c). Taking into account the evidence that only layers with a mean molecular orientation perpendicular to the fracture surface split up, eliminates the option of figure 3 (b) and points to a molecular ordering near a Grandjean line as shown in figure 3 (c). It means that, using de Gennes' notation [10], the Grandjean line is an edge dislocation of strength $\frac{1}{2}$ dissociated into a $\lambda^- \tau^+$ pair. We found no proof of a Grandjean line of strength $\frac{1}{2}$ dissociated into a $\lambda^+ \tau^-$ pair, which would give a molecular ordering as given in figure 3 (b). In [11], but using a reversed notation, it was suggested that the occurrence of the latter is energetically unfavourable.

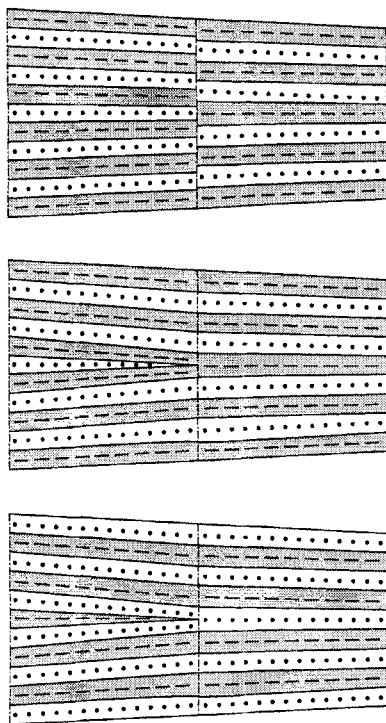


Figure 3. Outline of three possible configurations described in the literature occurring as a consequence of the addition of an extra π -rotation in the cholesteric twist at the location of a Grandjean-Cano line.

Although the phenomenon of Grandjean lines is well established, we believe that this is the first time that the molecular organization near these lines has been visualized, in an experiment already earlier proposed by de Gennes, and elucidating the dissociation of an edge dislocation into a $\lambda^- \tau^+$ pair of simple line dislocations.

We acknowledge Professor Heppke for inspiring us to perform the described experiment.

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