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PRELIMINARY COMMUNICATION

The relationship between liquid crystal alignment in the nematic and smectic C* phases

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The pretilt angle in the nematic phase is related to two different surface dependent factors, while the quality of surface stabilized ferroelectric liquid crystal (SSFLC) devices is dependent on only one of them. We have synthesized two series of polyalkyleneimides and found that the pretilt angles for thick nematic cells are not related to the cone angles in SSFLC cells.

Control of the alignment of nematic liquid crystals has been an established art form for many years. Millions of twisted nematic and supertwisted displays are produced every year; the overwhelming majority of them use rubbed polymers to align the nematic phase. The invention of the ferroelectric smectic liquid crystal display (SSFLC) [1], and subsequent problems in producing well-aligned smectic samples clearly highlighted how little is actually known about the mechanisms responsible for the surface alignment of liquid crystals on polymer surfaces. So far, liquid crystal display (LCD) manufacturers rely on tested procedures and recipes, but as the performance of the displays has to improve, an understanding of the mechanism underlying the surface alignment is needed.

Some workers have found a relationship between the pretilt angle in the nematic phase and the quality of SSFLC devices [2–6]. We have recently shown [7] that there is no general relationship between the pretilt angle in the nematic phase and the observed cone angle in the smectic C* phase.

To quantify the alignment in the nematic phase, we measured the pretilt angle with the room temperature nematic mixture ZLI 1132 (E. Merck). The crystal rotation method [8] was used to determine the pretilt. The alignment in the smectic phase was characterized by the cone angle (angle between the two extinguishing states) for cells with no field applied. The quarter plate method [9] was used to measure this. The room temperature smectic C* material ZLI 3774 (E. Merck) was used.

The pretilt in the nematic phase gives us information about how much the optical axis tilts out of the plane, while the data from the smectic C* phase will tell us how far the optical axis tilts away from the rubbing direction within the plane. However, both these approaches rest on the assumption that the optical axis has the same orientation throughout the cells. It has long been recognized that this is not true for the smectic C* phase (see, for example, [3–6]). We have recently shown [10] that, due to the strong anchoring of the mesogens on the polymer surface, the symmetry of the nematic phase is broken near the surface. The optical axis we measure by the crystal rotation method is the director of the nematic phase. This is the symmetry axis of the nematic phase. As

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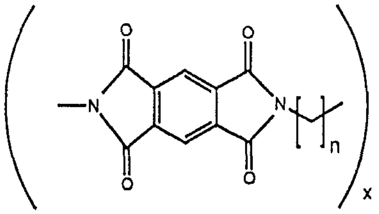
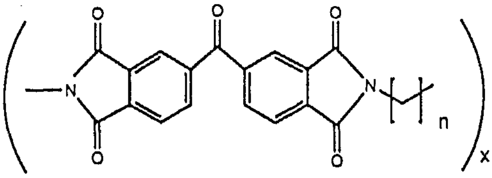


(I)



(II)

the symmetry of the nematic phase is broken close to the surface, the optical axis of the nematic phase is no longer constant through the cell.

The two different aromatic tetracarboxylic acids based on phenyl (PMDA – 1,2,4,5-benzene tetracarboxylic acid dianhydride) and benzophenone (BTDA – 3,3′,4,4′-benzophenone tetracarboxylic acid dianhydride) were condensed with a number of alkylene diamines with alkylene chains ranging from two to ten methylene groups to give the polyalkyleneimides (**I**) and (**II**), respectively. For the determination of the pretilt angle we used 120 μm thick cells with antiparallel rubbing directions. For the determination of the cone angle in the smectic C* phase we used 1·9 μm thick cells with parallel rubbing directions. Our experimental procedures for the synthesis and cell construction have been described in detail elsewhere [11, 12].

Figure 1 gives the variation of the pretilt angle with alkylene chain length for the BTDA series (**II**). The results are similar to those shown by other polyalkyleneimides [11], with a clear odd–even effect. The cone angles for the smectic material are given in figure 2. In this case, there is no odd–even effect. Except for an unexplained dip for the octylene chain, the cone angle is independent of the alkylene chain length.

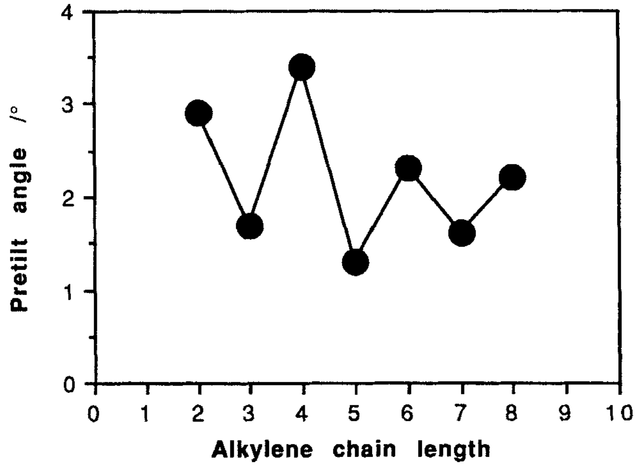


Figure 1. The pretilt angles found with ZLI 1132 for the members of the BTDA series (**II**).

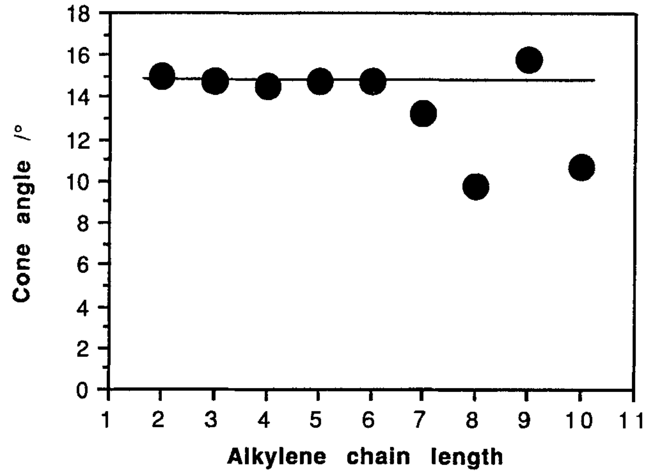


Figure 2. The cone angles found with ZLI3774 for the members of the BTDA series (II).

We now turn our attention to the PMDA series (I). Figure 3 shows that there is no odd–even effect when we look at the pretilt angles in the nematic phase. Instead there are some random variations across the series. Going to the smectic C* phase, the picture is completely different (see figure 4), and we observe a most dramatic odd–even effect.

The results here can be understood by applying our recently proposed model [13] for the pretilt angle in the nematic phase. We have shown that the pretilt angle of the optical axis in the bulk can be expressed as

$$\beta_{\mathrm{obs}} = a_1[(\beta_0 - \alpha) + (d^*/d)\alpha] + \tau. \qquad (1)$$

Here β_{obs} is the observed pretilt of the optical axis of the nematic phase, and a_1 is the in-plane order of the surface layer as discussed by Barmentlo *et al.* [14]. β_0 is the angle between the optical axis of an individual molecule and the surface (surface pretilt angle), and α is the angle between the optical axis and mechanical axis of individual molecules.

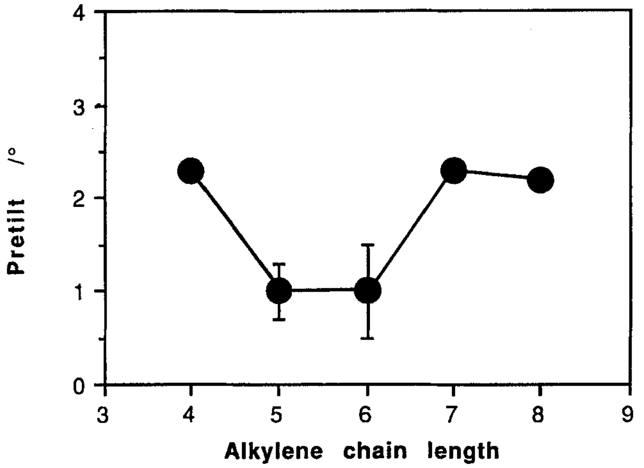


Figure 3. The pretilt angles found with ZLI 1132 for the members of the PMDA series (I).

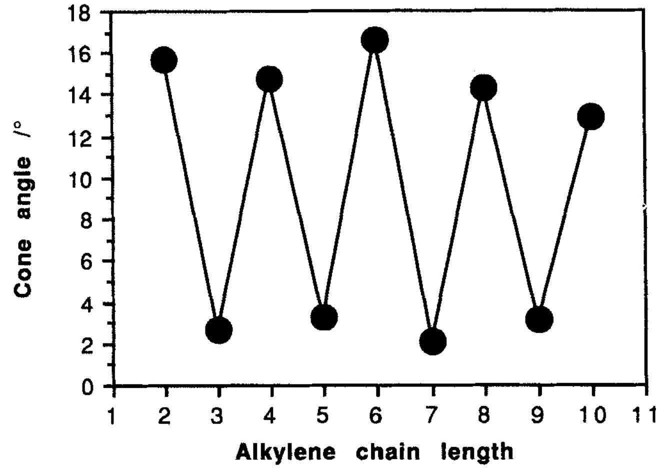


Figure 4. The cone angles found with ZLI 3774 for the members of the PMDA series (I).

d is the cell gap, and d^* is a surface layer thickness where the averaging from the alignment at the surface to the bulk alignment takes place. τ is the contribution from the physical deformation of the polymer.

We can neglect any contribution from physical deformation of the lightly rubbed polyimide alignment layer. d^* is typically less than 1 μm; with the 120 μm cell gap used for determining the pretilt angle, we can thus neglect the last terms and simplify equation (1) to

$$\beta_{\mathrm{obs}}=a_1(\beta_0-\alpha). \qquad (2)$$

This equation tells us that there are two different ways of achieving a high observed pretilt angle; we can increase the in-plane order (a_1) or we can increase the angle between the surface and the molecules in the first monolayer (β_0).

By annealing the polyimides in the PMDA series we increase the observed pretilt angle [7] due to an increase in a_1; this annealing also removes defects in the smectic C* alignment and improves the stability towards electric pulses [15]. The smectic phases have positional order as well as orientational order. There has to be a match between the periodicity of the smectic phase and the surface if we want good alignment. A well-ordered surface layer is a prerequisite for defect free smectic C* alignment [16]. Thus we can improve the alignment of the smectic C* phase by increasing the in-plane order (a_1). If the surface pretilt angle (β_0) stays constant, this will give us an increase in the observed pretilt angle β_{obs}. However, when we change the chemical structure of the polyimide, there are no a priori reasons to expect β_0 to stay constant. There will thus be no general relationship between a high pretilt angle in the nematic phase and the quality of SSFLC cells. A high in-plane order is a prerequisite for a defect free smectic C* alignment. However, a defect free alignment is no guarantee for a bistable alignment. We have so far not been able to derive equations similar to equation (1) for the cone angle in the smectic C* phase. It is clear though that both the in-plane order (a_1) and the surface layer thickness (d^*) must play important roles in these thin cells (1·9 μm) with an ordered liquid crystal. There is no reason why the surface pretilt angle (β_0) should play any important role.

A high observed pretilt in the nematic phase is dependent upon two factors—a high in-plane order and/or a high surface pretilt angle. Good quality SSFLC cells are dependent on only one of these factors, the high in-plane order. We do not therefore expect any general relationship between a high pretilt of the optical axis in the nematic phase and the bistability in the smectic C* phase. As our data clearly show there is in fact no such relationship. Data from Koden *et al.* [6], for a number of unspecified polyimides also show that there is no relationship between the pretilt angles in the nematic phase and the cone angles for the uniform states in SSFLCs.

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