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Shock-free ferroelectric liquid crystal displays with high optical contrast

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Shock-free ferroelectric liquid crystal displays with high optical contrast and low operating voltage are the concern of this contribution. Ferroelectric liquid crystal compositions with a wide temperature range of the chiral smectic C phase are designed. The influence of the chiral fragments of the molecules, especially their length, on the mechanical stability of the SmC* phase has been investigated. The relation between the surface anchoring energy of the aligning polymer and the viscoelastic properties of the SmC* phase has been described. Our qualitative model reflects the resistance of the SmC* layer structure against mechanical and thermal action. © *2010 American Institute of Physics*. [doi:10.1063/1.3530444]

Since Clark and Lagerwall[1] invented some basic principles of ferroelectric liquid crystal displays (FLCDs), much effort has been done in order to bring FLCDs on the status of practical applications.[2,3] In general ferroelectric liquid crystals (FLCs) can be used in a broad range, from single-pixel light choppers up to large area high definition television screens. FLCDs have several advantages compared to nematic liquid crystal displays, such as shorter switching time, wide viewing angle, and memory effect due to bistability. The combination of fast response time and memory effect allows large-size liquid crystal displays (LCDs) with high resolution. However, compared with the existing LCDs, the FLCDs owned an unfavorable reputation because of the poor performance of the gray scaling in moving pictures and in particular the sensitivity against mechanical shock.

To overcome this problem, flexural rigid rodlike chiral molecules with a defined length forming a chiral smectic C (SmC*) phase having a long helical pitch, low melting point, and a broad temperature range of the SmC* phase came into consideration.[4,5] Such chiral smectic C materials can be mixed with low-viscous nonchiral smectic C samples, yet the mixing ratio between the chiral and the low-viscous nonchiral amounts, respectively, has to be optimized. The correlation between the length of chiral molecules and the elastic properties of their SmC* phase on one hand and those of nonchiral molecules and their SmC phase on the other hand has to be found.

The control of the orientation of molecules leads to one of the key technologies for developing FLCDs for practical application.[3] In general a bookshelf or a chevron structure can be created. In our case the quasibookshelf geometry is essential.

The investigation of test cells using different aligning polymers followed by weak, medium, or strong rubbing allowed an understanding of the role of the surface morphology of such layers. Hence the correlation between the surface anchoring energy of the aligning polymers and the viscoelastic properties of FLCs has to be found.

The phase transition temperatures of single compounds and mixtures were measured using a Linkam heating stage and a polarizing microscope (MEIJI ML9400). The solution of the aligning materials was spun onto the indium-tin-oxide substrate (with the resistivity of about $10–20 \ \Omega/\text{cm}^2$) at 3000 rpm and thereafter baked at 200 °C for 1 h in order to allow imidization of the polyimide film. Aligning polymers were unidirectionally rubbed under a velvet-covered cylinder. The thickness of the cells hold between 1.5 and 4 μm, checked in each case interferometrically. In the same way the change of the thickness of the cells (geometrical deviation) was monitored. The microscopic textures of the cells were observed using a polarizing microscope while the electro-optical properties were measured between crossed polarizers using a He–Ne laser. The light transmittance became detected by a photomultiplier and recorded using a digital oscilloscope. The rise and decay times were defined as the change of the transmittance from 10% to 90% and vice versa. During electro-optical measurements the temperature of the cell was controlled with an accuracy of 0.3 °C. The determination of the liquid crystal pretilt angle was based on the measurement of the cell transmission using the rotation stage.[6] In Ref. 7 a spectroscopic method for measuring the twist angle in a liquid crystal cell has been described as well as a method for calculating the azimuthal component of the anchoring energy of the liquid crystal and of the substrate.

For creating eutectic "shock-free" mixtures two chiral compounds (1a) and (1b) are very promising. Their SmC* phase showed low melting point, wide temperature range, and needed elastic properties.

$$H_{2n+1}C_n \!-\!\!\!\!\bigcirc\!\!\!\!-\!\!\!\!\bigcirc\!\!\!\!-\!\!\!\!\overset{\overset{\displaystyle Cl}{|}}{\bigcirc}\!\!\!\!-\!\!\!\!\bigcirc\!\!\!\!-OCH(CH_3)X$$
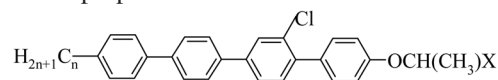
$n = 8$, $X = CH_2OC_3H_7$,

$$\text{(1a)}$$

Cr < 38.7 °C SmC* 115.6 °C SmA 160.3 °C I,

$n = 10$, $X = C_6H_{13}$,

$$\text{(1b)}$$

Cr 39.2 °C SmC* 129.7 °C N* 157.4 °C I.

On the other hand, nonchiral compounds with the required length and the required viscosity of the SmC phase have to be selected.

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TABLE I. Physical parameters of some FLC mixtures.

Mixtures	Temperature range of the SmC* phase (°C)	Spontaneous polarization (nC/cm^2)	Tilt angle (deg.)	Rise time (μs)	Decay time (μs)
A(15)	<0 SmC*+60.4	6	22.1	280	420
A(25)	<0 SmC*+67.5	10	23.4	310	440
A(33)	<0 SmC*+74.5	15	25.1	340	450
A(40)	<20 SmC*+80.7	18	26.1	600	720
A(50)	<20 SmC*+87.9	22	27.2	720	910
B(15)	<0 SmC*+58.4	21	22.1	67	140
B(25)	<0 SmC*+62.5	28	23.4	53	140
B(33)	<0 SmC*+67.5	35	23.8	57	150
B(40)	<20 SmC*+75.4	39	26.1	68	170
B(50)	<20 SmC*+86.9	44	27.2	93	210

As an important result of many trials while creating shock-free FLC mixtures one can state, the viscosity of the chiral SmC* phase while creating the mixture must be twice of those of the nonchiral SmC phase. Discussing on the molecular scale, the length of the chiral molecule (without the chiral fragment) should be comparable to the length of the nonchiral molecule.

2-phenylpyrimidines in general meet this requirement. Therefore two eutectic mixtures of 2-phenylpyrimidines, as examples, were prepared

$$H_{2n+1}C_n \text{—} \overset{=N}{\underset{N}{\bigcirc}} \text{—} \bigcirc \text{—} OC_{2m+1}H_m$$
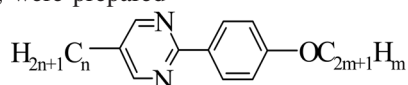
$n = 10$, $m = 8$: 44.4 wt %; $n = 8$, $m = 8$: 23.0 wt %;
 (2a)
$n = 8$, $m = 9$: 32.6 wt %,

$n = 10$, $m = 8$: 24.3 wt %; $n = 8$, $m = 8$: 15.2 wt %;
 (2b)
$n = 8$, $m = 9$: 18.7 wt %; $n = 8$, $m = 6$: 41.8 wt %.

Additionally a typical FLC-mixture (FLC1), not recovering after mechanical deformation, has been prepared

FLC1: $n = 10$, $m = 8$: 30.4 wt%; $n = 8$,

$m = 8$: 20.0 wt %; $n = 8$, $m = 9$: 24.6 wt %;

R,R-4,4″-(2-methylheptyloxy)-terphenyl: 25.0 wt %.

In order to optimize the mixture compositions, the chiral dopants (1a) and (1b), respectively, were added to the mixtures (2a) and (2b) in different amounts, leading to the series A(y) [(1a) combined with (2a)] and B(y) [(1b) combined with (2b)]; **y** is the weight percent of chiral molecules (1a) and (1b), respectively. The main physical parameters of such mixtures are given in Table I; here the gap of the test cell is approximately 2 μm.

Only cells filled with mixtures containing chiral molecules between 25 and 40 wt % do not show zigzag defects and are stable under mechanical deformation, confirmed also by contrast ratio measurements before and after mechanical deformation (see Fig. 1).

In order to study the influence of the anchoring energy of the aligning layer on the mechanical stability, cells with different thicknesses of the aligning polymer (15–90 nm) and a cell gap 2 and 3 μm, respectively, were prepared. The contrast ratio was measured immediately after filling the cell and after applying mechanical deformation. Under this condition the thickness of the cell changed more than 50%.

For controlling the disappearance of defects under various conditions after applying mechanical deformation, the polarizing microscope has been used too.

Figure 2 shows the change of the contrast ratio after mechanical deformation as a function of the anchoring energy for some shock-free materials, A(33) and B(33), as well as for the typical mixture FLC1.

Under the described special conditions, the smectic layers do not break. The designed FLC mixtures are arranged in a quasibookshelf layered structure inside the cell.

If the mechanical deformation has been applied only for a short time (few seconds), the texture has been restored immediately. If strong mechanical deformation has been applied, for example over few minutes, the restoring of the texture is by some delay. However, in both cases there is a complete recovery, monitored via measuring of the contrast ratio.

In ideal shock-free smectic mixtures under severe deformations some areas react only with a layer bending and shifting, without any changes inside the layer. This has been con-
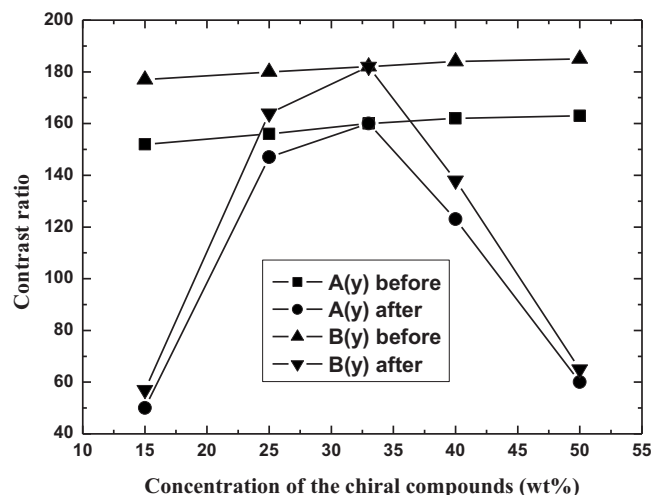

FIG. 1. Contrast ratio of cells containing mixtures A(y) and B(y), respectively, vs concentration of chiral compounds [(1a) and (1b), respectively] before and after application of mechanical stress (geometrical deviation 60%).
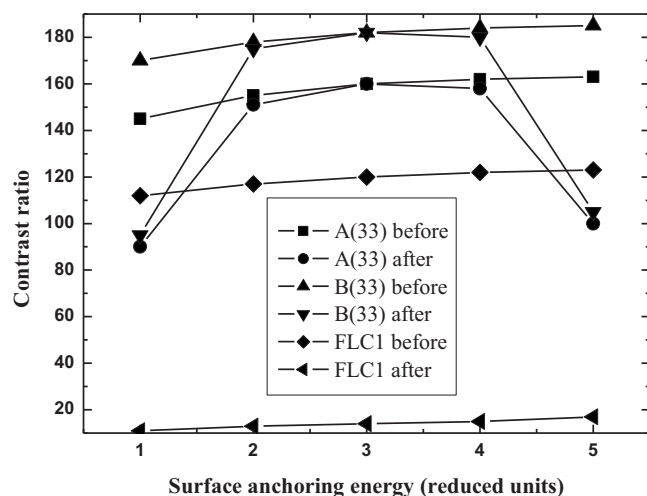
FIG. 2. Contrast ratio vs surface anchoring energy before and after mechanical deformation (geometrical deviation 60%).

firmed by studies under the polarizing microscope. No defects at all before and after the deformation appeared, but a change of the uniformity of the transmitted light under crossed polarizers from black to dark gray. This is due to the inclination of layers. After removing such deformation the cell became uniformly black.

Such conditions are only possible if a special relation between the elastic properties and the anchoring energy exists. Exceeding the pretilt angle of the alignment layer above 1°, no shock-free effect appeared, hence the balance between the anchoring energy and the elastic constant of the FLC disappeared.

The most important requirements can be summarized. The concentration of chiral molecules in mixtures must be between 25 and 40 wt %. The ratio between the length of the chiral fragment of those chiral molecules and the width of nonchiral molecules should be equal 2 to 3. The length of the chiral molecule (without the chiral fragment) should be comparable to the length of nonchiral molecules. The optimal cell gap is between 1.5 and 4.0 μm. The contrast is then completely restored, when the thickness of the aligning layer is between 40 and 60 nm. Important further, the aligning layer pretilt angle must be less then 1°. Under this condition no irreversible destruction of the smectic layers is observable.

To explain this extraordinary behavior, two aspects should be taken into consideration. One aspect is, by considering the mobility of individual molecules, one can assume that under mechanical stress the heavier and longer chiral molecules are fixed inside the layers while the more flexible and smaller nonchiral molecules can be expelled out of the individual layer, but being able to return after releasing the deformation.

The second view is more complicated. As has been demonstrated, the insensitivity against mechanical influence exists only under very restricted conditions, namely, special chiral molecules, special geometric relations between chiral and nonchiral molecules in a narrow concentration range, optimized anchoring conditions, and certain pretilt angle of the aligning polymers. One can assume under these conditions a formation of some sort of supramolecular aggregations takes place. This means under very narrow conditions geometrically optimized supramolecular nuclei appeared inside the smectic layers. Such supramolecular units might create special elastic properties, stabilizing the smectic layers against breaking. This more speculative consideration is supported by a sort of elastic layer bending seen under microscope during the evaluation of the texture under applying mechanical stress. After removing the deformation, the texture is restored showing the initial contrast ratio. At the same time the supramolecular assemblies will move back to their initial volume due to the restoring forces caused by the elastic torque.

Following the concept of combining chiral quaterphenyls with related phenylpyrimidines the combination of chiral pentaphenyls with biphenylpyrimidines would be a very good proof for understanding the entire effect. Scattering experiments as example without and under mechanical stress, a detailed study of the elastic properties of the mixtures and a through out theoretical investigations are further indicated.

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