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# Molecular Crystals and Liquid Crystals

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W. Zheng <sup>a</sup> , I. Underwood <sup>a</sup> , B. F. Macdonald <sup>b</sup> , R. J. Cole <sup>b</sup> , D. G. Vass <sup>b</sup> & R. J. Cole <sup>c</sup>

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<sup>&</sup>lt;sup>a</sup> Department of Electrical and Electronic Engineering, The University of Edinburgh, Edinburgh, UK

<sup>&</sup>lt;sup>b</sup> School of Physics, The University of Edinburgh, Edinburgh, UK

<sup>&</sup>lt;sup>c</sup> Centre for Materials Science and Engineering, The University of Edinburgh, Edinburgh, UK

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# THE EFFECT OF RUBBING STRENGTH ON THE FORMATION OF ZIGZAG DEFECTS IN SURFACE STABILISED FERROELECTRIC LIQUID CRYSTALS

W. Zheng and I. Underwood Department of Electrical and Electronic Engineering, The University of Edinburgh, Edinburgh EH9 3JF, UK

B. F. Macdonald, R. J. Cole, and D. G. Vass School of Physics, The University of Edinburgh, Edinburgh EH9 3JZ, UK

R. J. Cole Centre for Materials Science and Engineering, The University of Edinburgh, Edinburgh EH9 3JL, UK

The effect of the rubbing strength on the formation of the zigzag defects in surface stabilised ferroelectric liquid crystal (SSFLC) devices is demonstrated. Atomic force microscopy images of rubbed polyimide (PI) films reveal that the surface topography is modified by mechanical rubbing and reflection anisotropy spectroscopy reveals the surface anisotropy of the rubbed polyimide films increases with rubbing strength. Strongly rubbed PI films are found to generate zigzag lines and the density of these defects in the SSFLC layers is found to increase with rubbing strength.

Keywords: atomic force microscopy; ferroelectric liquid crystal; FLC alignment; reflection anisotropy spectroscopy; rubbed polyimide; zigzag lines

#### I. INTRODUCTION

Zigzag lines are common defects that appear in most surface stabilised ferroelectric liquid crystal (SSFLC) devices. These undesired defects are the dividing lines between the two chevrons which have their apex pointing in opposite directions. The chevron is believed to be formed due to a

Address correspondence to W. Zheng, Department of Electrical and Electronic Engineering, The University of Edinburgh, Edinburgh EH9 3JF, United Kingdom.

competition between the pinning of the liquid crystal molecules on the substrate and the shrinkage of the smectic layers as the liquid crystal enters the Sc\* phase [1-4]. The prerequisite for zigzag free layers is that the FLC smectic layers must be properly controlled to form the bookshelf structure or a one-domain chevron, with either C1 or C2 structure, over the entire cell. In reality, an ideal bookshelf structure can rarely be achieved, and the chevron structures normally form in most cases. At present, most SSFLC devices are constructed using substrates which are coated with a thin film of polymer and then rubbed. Infrared dichoric measurements of mechanically rubbed polyimide (PI) surfaces suggest that the rubbing procedure causes directional orientation of the polymer chains in the upper layer of the film [5]. It is widely thought that the ability to promote the alignment of liquid crystal molecules is generated by this orientation within the alignment layer [6]. Mechanical rubbing of a PI film will also result in changes to the physical properties of the film, especially the surface morphology and it has been reported that anchoring energies change with rubbing strength [7]. The examination of rubbed PI films by means of reflection anisotropy spectroscopy (RAS) [8] and reflection ellipsometry [9,10] reveals that mechanical rubbing generates surface anisotropy. These changes in the molecular alignment and surface topography of the alignment layer affect the molecular alignment of the ferroelectric liquid crystal (FLC) layer as it is in direct contact with the PI film. It has recently been suggested that the surface topography of the alignment layer may be a factor responsible for the formation of zigzag defects within SSFLC devices [11].

In this paper, we demonstrate that the rubbing strength (RS) plays an important role in the formation of zigzag defects by examining the surface of PI films modified by mechanical rubbing using atomic force microscopy (AFM) and RAS.

#### II. EXPERIMENTAL

In the present studies, a polyamic acid (Aldrich 431206) was diluted to 5 wt% in N-methyl-2-pyrollidinone, and spin coated on to ultrasonically cleaned 1 cm<sup>2</sup> ITO glass slides. The imidization of the amic acid occurs by soft baking at 100°C for 30 minutes followed by thermal curing at 250°C for 100 minutes. The surfaces of the PI films were then rubbed using a custom made rubbing machine, composed of a velvet nylon cloth covered rotating drum and a flat sample holder which is moved laterally under the drum. The rubbing strength can be evaluated using the following equation [12]

$$RS = N\Lambda(2\pi R\omega/v - 1),\tag{1}$$

where N is the number of rubbing cycles,  $\Lambda$  is the pile impression of the velvet fibres,  $\omega$  is the rotation speed of the drum which was fixed at 40 rpm in the present studies, v is the lateral movement speed of the substrate and R the radius of the drum. As  $\Lambda$  and v are normally preset for each processing, RS is therefore determined by the number of rubbing cycles, N.

The rubbed PI films were analysed using a phase modulating reflection anisotropy (RA) spectrometer. A linearly polarised light beam propagates along the direction normal to the sample surface (z-direction) with the polarisation plane parallel to yz plane. Sample are aligned with rubbing direction parallel to the y-direction. The difference between the reflectivity for light linearly polarized along two perpendicular directions ( $\Delta r$ ) is measured and the result is given in terms of the ratio of the change in the samples reflectivity and the average reflectivity (r)

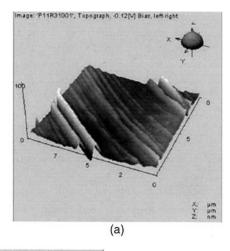
$$\frac{\Delta r}{r} = 2\frac{r_x - r_y}{r_x + r_y},\tag{2}$$

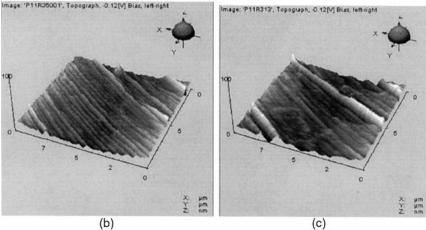
where  $r_x$  and  $r_y$  are the complex Fresnel reflection amplitudes for light polarised along x and y, respectively. Samples studied prior to surface treatment returned a zero RAS signal indicating the surface is isotropic, as expected.

The surface morphology of the PI films was examined using an AFM (Molecular Imaging PicoSPM) and the FLC alignment effects were examined using a polarising optical microscope (Olympus BH2). Cells were made using rubbed PI coated substrates and were assembled in the antiparallel configuration using 2.4  $\mu m$  spacers to ensure a constant predetermined gap. The FLC samples were made by introducing a ferroelectric mixture (Felix015/100 from Hoschst) into the cells at 105°C (about 10°C above the clear temperature of the FLC) through capillary action. To avoid thermal turbulance during the molecular alignment stage, the samples were cooled with the temperature carefully controlled. In these studies, the cooling rate for all samples was set at 1°C min $^{-1}$  using a Linkam TMS 93 temperature system (Linkam Scientific Instruments Ltd).

### III. RESULTS AND DISCUSSIONS

Figure 1 shows AFM images of rubbed PI films and clearly demonstrates mechanical rubbing to cause changes in the surface topography of the PI films. As can be seen, after one rubbing cycle (RS =  $150\,\mathrm{mm}$ ) microgrooves parallel to the rubbing direction have been formed. In some regions large quantities of the PI material has been excavated from the surface leaving grooves with width  $>1\,\mu\mathrm{m}$ , as illustrated in Figure 1(a). With subsequent rubbings more grooves are produced (Fig. 1(b)) and as this continues

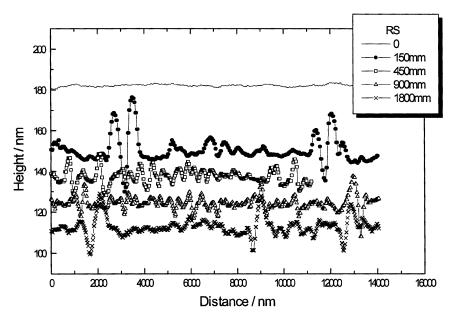




**FIGURE 1** AFM images for PI films rubbed with rubbing strength of (a) 150 mm, (b) 900 mm, and (c) 1800 mm.

some degree of damage may be caused in the PI film, as can be seen in Figure 1(c). More details of these topographical changes can be obtained from a surface profile analysis.

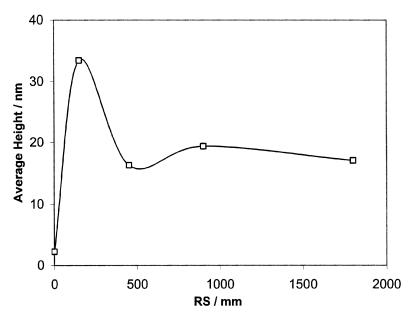
Figure 2 shows the AFM cross section profiles of the rubbed PI films in the plane orthogonal to the rubbing direction. After the first rubbing cycle, the surface profile of the PI film contains valleys whose width range from  $160\,\mathrm{nm}$  to  $1.3\,\mu\mathrm{m}$ . As the RS increases, the number of grooves increases, and the width of the grooves reduces and become mores uniform (see Fig. (1b) and Fig. (1c)). The surface of the PI film before rubbing is



**FIGURE 2** The cross section profiles of the rubbed PI films in the plane orthogonal to the rubbing direction.

reasonably flat with an average peak-to-valley height  $(H_{PV})$ , i.e. the average depth of the microgrooves, of 2.2 nm. As illustrated in Figure 3, the first rubbing cycle produces microgrooves with  $H_{PV}=33\,\mathrm{nm}$  on average for the 10 µm scan width, but further rubbing cycles lead to a significant reduction of  $H_{PV}$ . Although the surface topography will lead to only a small variation in pile impression (and hence rubbing strength) across the surface, it would appear that peaks in the corrugated surface suffer much higher abrasion rates than troughs, leading to a reduction in  $H_{PV}$  with RS. Quantitative analysis of this trend is complicated by the production of surface debris and gauging of the surface. Certainly after extensive rubbing the complete removal of polymer from regions of the surface will contribute to a continual reduction of  $H_{PV}$ .

Real time  $in\ situ$  experimental probes are needed to learn more about the detailed origins of the surface topography of rubbed polymers. RAS is an optical reflectivity technique and in principle can fulfil this role. Results of an RAS study of rubbed PI surfaces as a function of RS are shown in Figure 4. Before rubbing, the samples studied here were found to be isotropic in the plane of the surface and so RAS measures only the optical anisotropy induced by surface rubbing. It can be seen that the largest increase in measured anisotropy occurs after the first rub (RS = 150 mm).



**FIGURE 3** The average peak-valley height as a function of rubbing strength for PI films.

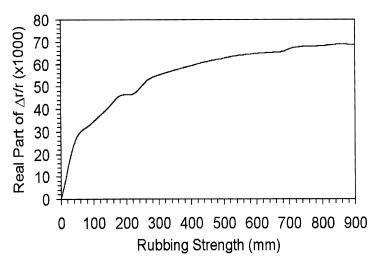


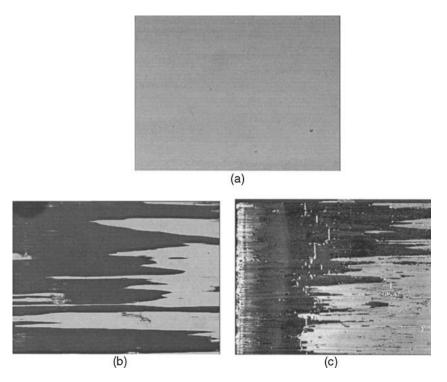
FIGURE 4 The RA spectra as a function of the rubbing strength for PI films.

Although the relation between the surface anisotropy and degree of the molecular ordering of the PI film has not been investigated here, previously published infrared dichroic measurements of the rubbed PI revealed that the backbones of the polymer chains are oriented parallel to the rubbing direction [5]. From our previous studies [13] we know it is possible to measure anisotropy in a PI sample, using RAS, whilst seeing no evidence of topographical features. We believe RAS measurements from the rubbed the PI films reveal mechanical rubbing to generate optical anisotropy due to the orientation of the polymer chains rather than the "form birefringence" due to the topographic grooves. The observed increase in RAS amplitude with RS implies that the degree of molecular orientation in the polymer film also increases with RS.

The objective of employing a rubbed PI film as an alignment layer in liquid crystal devices is to promote the alignment of the liquid crystal molecules on the surface of the polymer. The orientation of the liquid crystal molecules in the device is determined after they contact the surface and in general all samples made with substrates coated with rubbed PI films show good alignment when considering only the orientation of the molecules. This was confirmed by heating the liquid crystal in the cells to the nematic phase, and examining the texture using a polarising microscope. When the sample is rotated, the viewing field of the polarising microscope becomes dark and bright alternatively indicating that the liquid crystal has been aligned. Figure 5(a) which shows a FLC layer, after the liquid crystal has cooled down and entered the Sc\* phase, in a cell made using substrates coated with PI film rubbed with RS = 150 mm. As can be seen, this photomicrograph shows no evidence of zigzag lines proving that a defect free FLC layer can be produced in a cell constructed using PI films which have been subjected to a weak RS. In cells constructed using PI films which have been subjected to a stronger RS, zigzag lines can be clearly seen (Fig. 5(b)). Further increases in the RS causes the density of the defects to increase proportionally, as illustrated in Figure 5(c).

The production of a zigzag defect free FLC layer, sandwiched between weakly rubbed PI films, indicates a one-domain chevron structure is present in the cell. Similarly, the appearance of zigzag lines in the samples constructed from PI films which were treated with high RS indicates that the one-domain-chevron in the cells has been destroyed and replaced with multi-domain chevron structure.

It has been suggested that zigzag free FLC alignment can be achieved using rubbed PI films which can generate a large molecular pretilt [14] (although moderate pre-tilt which matches the difference between the smectic cone angle and the layer tilt angle can also promote defect free C2 alignment). However, for the PI used in the present studies, the maximum pretilt that can be generated is less than 5° [15] and so for this



**FIGURE 5** FLC alignment in the cells made using substrates treated with rubbing strength of (a) 150 mm, (b) 900 mm, and (c) 1800 mm.

case, the zigzag free FLC does not result from the large pretilt angle. A theoretical model predicted that for an alignment film which generates a small pretilt angle, a small surface anchoring energy is required to guarantee a zigzag free FLC layer [16]. Our observations suggest that a weak RS may meet the low anchoring energy requirement and favour the formation of the one-domain chevron structure, whereas a strong RS may cause an increase in the anchoring energy [7,17], destroying the one-domain chevron structure and therefore generating zigzag lines.

#### IV. CONCLUSION

Complementary surface characterisation tools have been used to assess the effect of mechanical rubbing of PI films; AFM measures the surface topography while RAS essentially reveals molecular alignment at the surface. Both techniques show that the early stages of rubbing have the strongest

effect on the surface. PI films treated with a low rubbing strength enable the formation of a one-domain chevron, and hence, can produce a zigzag free FLC layer.

We found that increasing the RS lead to a decrease in the average trough to peak height of the surface grooves and that the resulting FLC cells displayed zigzag lines due to domains with different chevron structures.

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