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Atomic Force Microscopy of Rubbed Polyimide Aligning Films for Liquid Crystal Displays

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Abstract The surface of polyimide (PI) films for aligning liquid crystals were studied by means of atomic force microscopy (AFM). The surface of the unrubbed PI films consisted of polymer clusters of different sizes which are randomly distributed over the film area. After rubbing, however, these polymer clusters formed long chains along the rubbing direction. The cluster chains were separated with periodicity about 100 nm for weak rubbing strength. Deeper grooves are also present, separated with periodicity about 2 μm that could be related to the microstructure of the fibres from the rubbing cloth. On increasing the rubbing strength further on, the cluster chains coalesce into wider ones. The rubbed PI films show optical retardation, which was increased rapidly with increased the rubbing strength and reached a constant value of about 1.4nm.

INTRODUCTION

Rubbed polyimide films are utilized widely in the fabrication of liquid crystal displays as aligning layers.[1] Rubbing is the simplest available method for obtaining homogeneous alignment of liquid crystals and widely used in the production of liquid crystal displays because of its high productivity. Although many theoretical and experimental studies on liquid crystal alignment have been performed, the actual alignment mechanism in the case of rubbed polyimide films is still not fully understood.[2-10] One assumption is that the alignment of liquid crystal molecules is induced by grooves or scratches formed mechanically on the polymer surface by the rubbing process. Another one is that alignment is due to the orientation of the polymer molecule chains.[5,6]

Obviously, knowledge about the surface state and morphology at the nanometer level is necessary in order to reveal the relationship between the rubbing strength and the surface anchoring strength of the liquid crystal. In this study, it is difficult to use ordinary electron microscopy due to the low height resolution. Instead scanning tunnelling microscopy (STM)[11] has been used to image rubbed polymer surface.[12,13] However, the

STM requires conducting samples, therefore the polymer films must be coated with a conductive layer. In contrast, the atomic force microscope (AFM)[14,15] does not need a conductive sample, so finer surface structures can be investigated. The AFM generates images with nanometer resolution by raster scanning of a sharp tip over the surface area at constant tip-sample force.[14] The sharp AFM tip, which is fixed on a soft spring, is pressed against the sample and the force is kept constant (typically 10^{-7} to 10^{-9} N) by measuring the deflection of the spring. The soft spring is usually in the form of a microfabricated cantilever. To detect the deflection of the cantilever, a common method is by optical techniques. In the AFM the interaction is dominated by relatively short range forces, giving a resolution, in favourable cases, down to the atomic level. D. S. Seo et al. have made an AFM study of rubbed polymers showing that grooves are present on the surface.[16]

In this work, we describe an AFM study on the influence of rubbing strength and the surface structure of rubbed polyimide films.

EXPERIMENTAL

The polyimide (HT210, Toray Industries) films were deposited on indium-tin-oxide (20nm thick) coated glass substrates by spinning on a dilute solution (typically 6% by weight). The solvent was evaporated at 80°C for 30 min., and the film was cured at 300°C for 1 hour. The approximate thickness of the resulting polyimide films was about 100 nm. Rubbing was done with a nylon velvet. The rubbing strength (RS) was calculated using the expression[17]

$$RS = \gamma \times L \tag{1}$$

where γ is a characteristic of the rubbing process including the rubbing pressure, the fibre density in the rubbing material, the coefficient of friction etc. L is the total length of the rubbing fibre in contact a with certain point of the substrate and is expressed by

$$L = N \times l\left(\frac{2\pi rn}{60v}\right) \tag{2}$$

where N is a cumulative number of rubs, l is the contact length of rubbing fibre, n is the number of revolutions per minute (r.p.m.) of the roller, r is the radius of the roller and v is the velocity of the substrate stage.

Standard Si_3N_4 AFM tips, with a force constant of 0.0032 N/m, were used in a

commercial AFM.[18] The force applied in all images was about 5 nN and the scanning frequency was kept in the interval 0.5-1 Hz. The images were tilt corrected and low-pass filtered.

The degree of molecular orientation in the polyimide films was also estimated by measuring the optical retardation of the films using Senarmont ellipsometry method (λ =632nm).[9,19] The measuring set-up is schematically shown in Figure 1. The axis of the polarizer is set at 45° to the rubbing direction and parallel to the optical axis of the quarter-wave optical plate. The analyzer is rotated about an angle (θ) until the minimum transmission is obtained. The optical retardation (δ) is calculated by the following equation

$$\delta = \frac{\lambda \cdot \theta}{180^{o}} \tag{3}$$

where λ is the wavelength of the incident light.

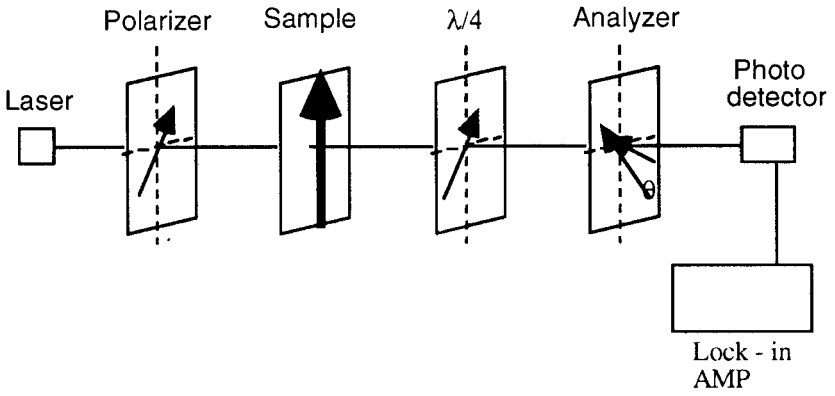
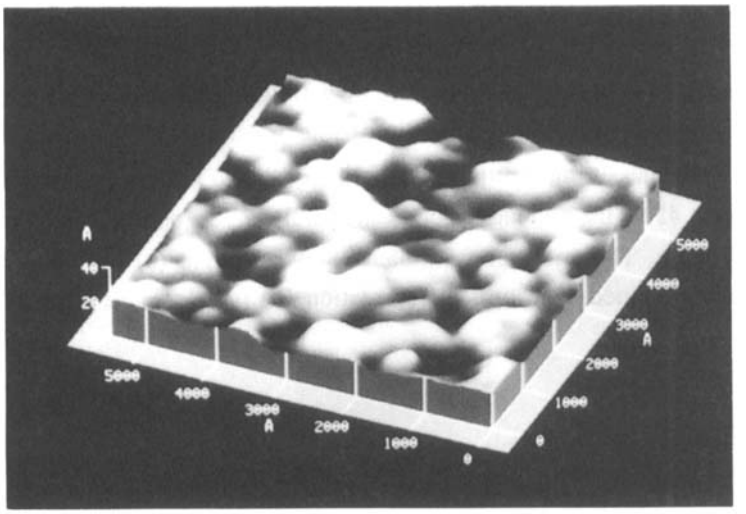


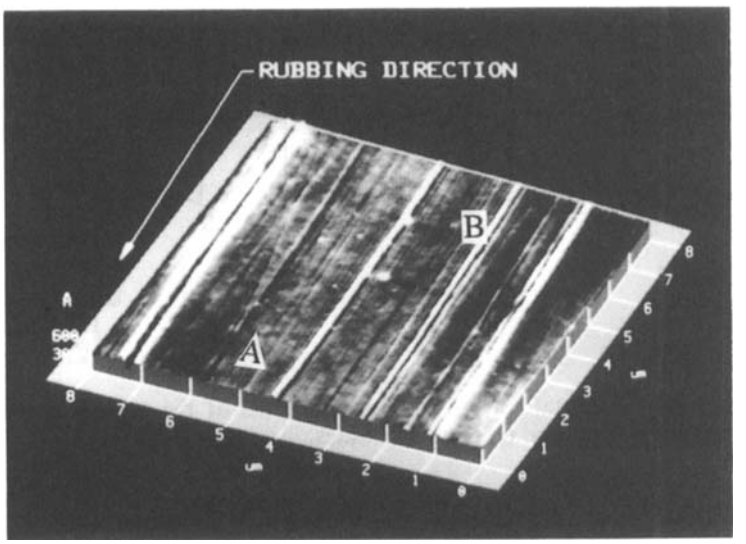
Figure 1 Experimental set-up for measurements of the optical retardation of polyimide film deposited on glass substrates.

RESULTS AND DISCUSSION

The unrubbed PI films consist of randomly distributed clusters of different sizes, as shown in the AFM image (Figure 2a). The sizes of the clusters are much bigger than an individual polyimide molecule. The cluster formation process starts in the solution where a number of polyimide molecules spontaneously form clusters with spherical shape. After deposition of the polyimide solution film onto glass substrate and complete solvent evaporation the shape of the remaining clusters becomes strongly deformed.

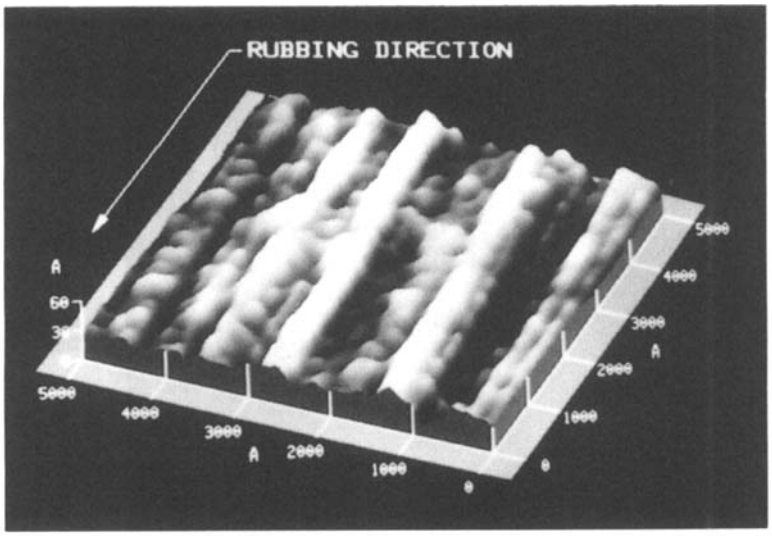


a



b

Figure 2 a: AFM image of the unrubbed polyimide showing a randomly distributed surface (grey scale: 1.6 nm). b: Larger view of the rubbed surface with grooves separated by deeper grooves (grey scale: 15 nm). Note that the distance between the deeper grooves is the same as the diameter of the subfibres (about 2.0 μm) shown in Figure 4a (grey scale: 570 nm)



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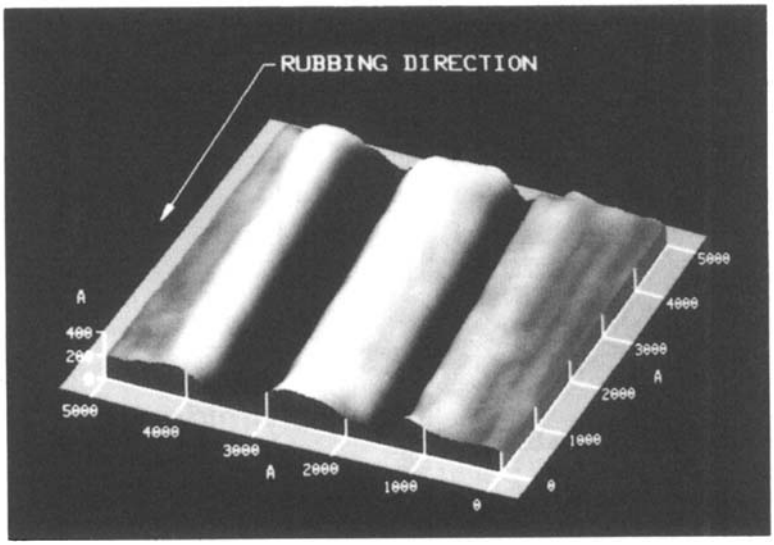
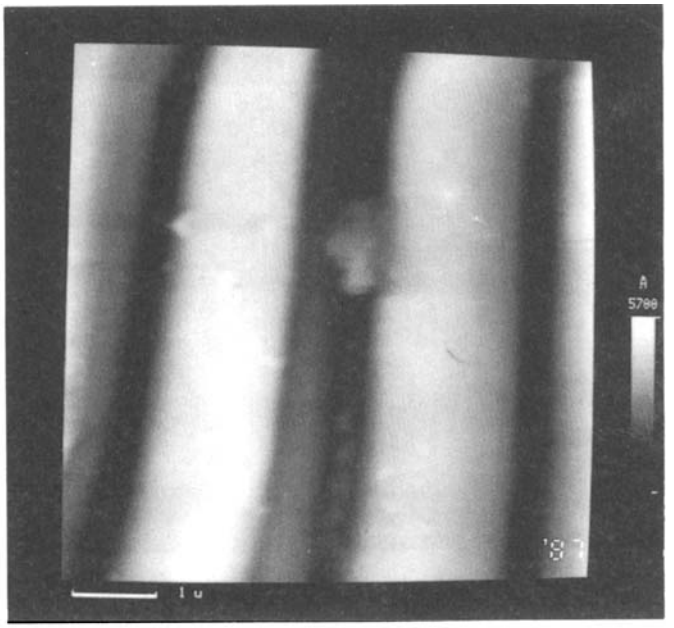


b

Figure 3 a: Cluster chains and grooves with higher magnification (labelled A in Figure 2b). b: A deep groove with higher magnification (labelled B in Figure 2b).



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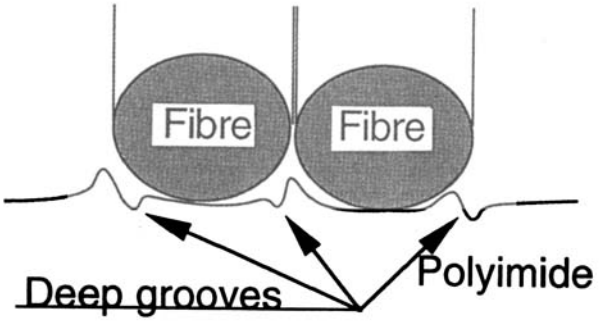

b

Figure 4 a: AFM image of a small part of a rubbing fibre (diameter about 20 μm), which are used in the rubbing machine. The fibre consists of a number of smaller sub-fibres (diameter about 2.0μm). b: The mechanism suggested to explain the process of forming the deep grooves. The high pressure in the centre of the fibres locally melts the polyimide films.

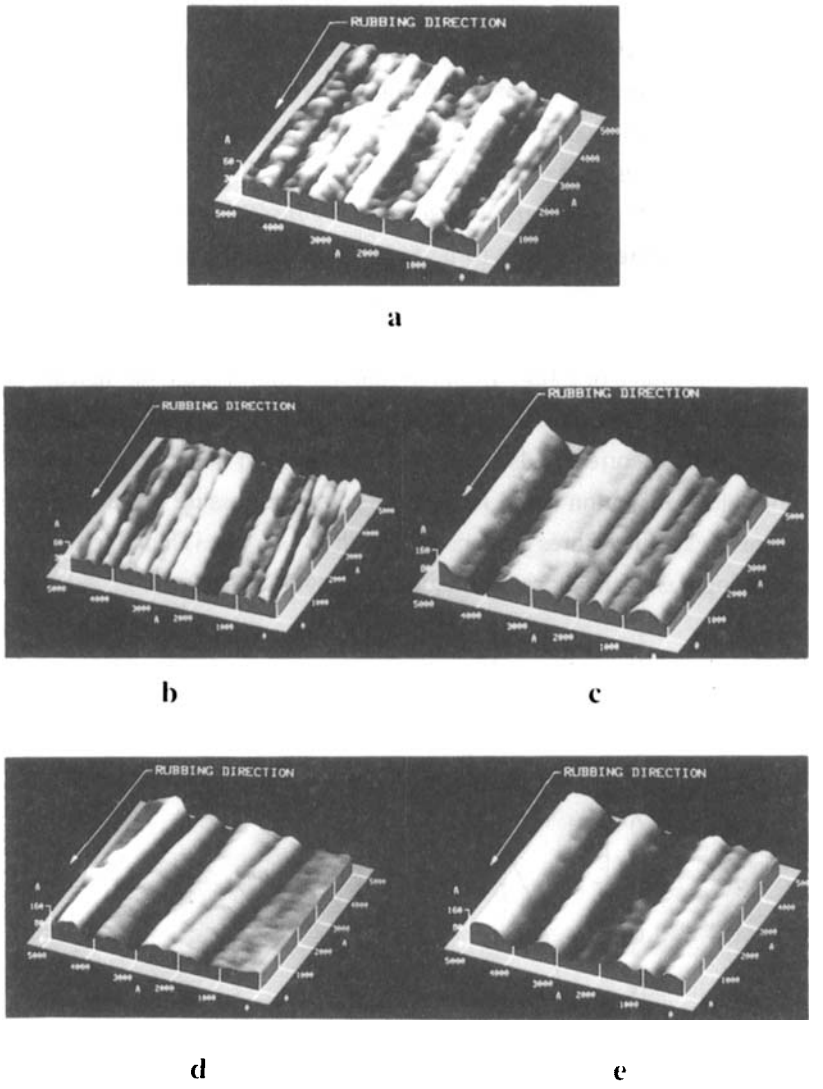

Figure 5 A sequence of AFM images showing the effect of increasing rubbing strength (RS). (a) Even at weak rubbing strength the alignment of clusters is visible, but some degree of disorder still remains (RS=7 cm). (b-e) With increasing rubbing strength the cluster chains coalesce into wider ones. (b) RS=21 cm (c) RS= 36 cm (d) RS=43 cm (e) RS=57 cm

After rubbing, the surface of PI films is drastically changed: long chains of connected clusters are formed and aligned along the rubbing direction (Figure 2b). Two distinct features of the rubbed surface are visible; wide smoother parts with shallow grooves and parts with deep grooves. Also, the grooves are paralled to the rubbing direction. The AFM images also reveal that the smoother part of the surface relief consists

of fine cluster chains with cluster size of about 50 nm (Figure 3a).

These cluster chains are aligned along the rubbing direction with a certain period, although some of them have coalesced into thicker bundles. Due to the relatively small rubbing strength (RS=7cm), some parts of the surface still contain randomly distributed clusters (see upper left part of Figure 3a).

The part with deeper grooves seen in Figure 2b (labelled B in Figure 1b) and magnified in Figure 3b seems to be built up of bundles of thin cluster chains. The surface of the bundles appears to be smoother than the one of the fine cluster chains as shown in Figure 3a.

An AFM image of a fibre from the rubbing cloth (Figure 4a) reveals that the fibre has a complicated structure. It consists of a number of fine subfibres with a diameter of about 2μm, which corresponds to the distance between the deep grooves in Figure 1b. One possible explanation could be that the rubbing pressure has a maximum value in these points where the fibres are incontact with the surface. Then, the friction force, due to the rubbing, results in a local heating of the polyimide surface.

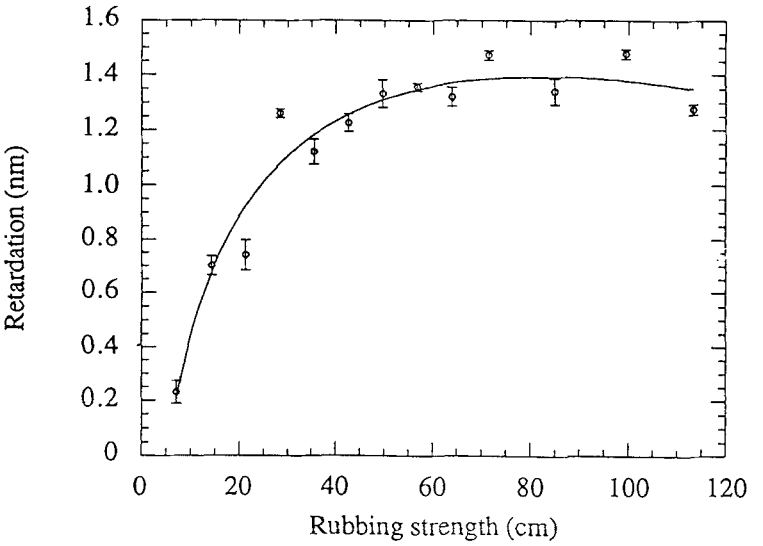

Figure 6. The optical retardation as a function of rubbing strength. The optical retardation increases with increased rubbing strength up to about RS=50 cm

Therefore, at these points the clusters could be deformed and easily moved along the rubbing direction. We suggest that the deep grooves are formed because the clusters are pushed and piled up between the fine subfibres as shown schematically in Figure 4b.

The effect of increased rubbing strength is demonstrated in Figure 5. Even at weak

rubbing strength, the anisotropic alignment of the clusters is visible, although some degree of disorder still remains (RS=7 cm, Figure 4a). The cluster chains are quasi-periodically aligned along the rubbing direction with a spacing of about 100 nm.

With increasing rubbing strength, two or more cluster chains coalesce into thicker bundle chains. As mentioned above, the local heating and shearing force cause formation and alignment of cluster chains. For the same reason, the fine cluster chains coalesce into thicker bundles when the rubbing strength increases.

The rubbing process causes not only changes in the surface morphology but also partially aligns the polyimide molecules along the direction of rubbing. The alignment of polyimide molecules after rubbing is similar to the orientation of polymer molecules due to cold drawing of bulk polymers.[4,6] The optical retardation of rubbed polyimide films is due to both those factors. The higher is the rubbing strength the larger is the optical retardation of the polyimide films (Figure 6). It is observed that the optical retardation is increasing rapidly with the rubbing strength up to RS=50 cm and then saturates, reaching a constant value of about 1.4 nm.

CONCLUSION

The surface structure of unrubbed and rubbed polyimide films is directly observed using an AFM. It is found that the unrubbed surfaces of polyimide films consist of randomly distributed clusters with size much larger than the one of an individual polyimide molecule. The rubbing process drastically changes the surface structure of polyimide films. It forms well aligned cluster chains and some of them coalesce into wide bundle chains on increasing the rubbing strength.

The rubbing strength was also correlated with the optical retardation of the rubbed polymer films. The rubbing process causes not only changes in the surface morphology but also partially aligns the polyimide molecules. The optical retardation of rubbed polyimide films is due to both those factors, however, which of them is the most important for the optical retardation is a question for further study.

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