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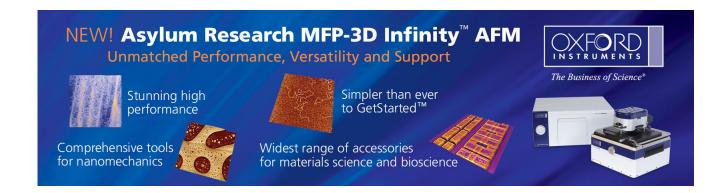
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Nanoimprinting-lithography-induced self-aligned liquid crystals for novel multifunctional optical films

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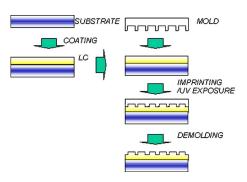
Polymerizable nematic liquid crystal (LC) was adopted as a resist for our newly developed nanoimprinting lithography process. Liquid crystal molecules were self-aligned during imprinting process without precoated alignment layer. The large patterned area was able to achieve 4 × 4 cm² at very low impriting pressure, 1.5 bar. The optical anisotropic was verified through polarized optical microscopy and the retardation was measured by polarized ultraviolet/visible spectrometer. The micrograted LC films prepared by this novel method offer the potential applications for a LC alignment layer within LC cells, as well as preparation of optical anisotropic films. © 2006 American Institute of Physics. [DOI: 10.1063/1.2173222]

A liquid crystal (LC) alignment layer is one of the most critical elements in forming high-performance LC devices. A good alignment layer cannot only induce LC molecular order, but can also enhance its order to a greater degree. This enhancement can directly affect contrast ratio in display applications. Generally, the preparation of an alignment layer for LC cells can be characterized into contact¹ and noncontact modes². Until now, contact modes dominate the mainstream of the LC cell preparation process, in which the polymeric layer yields a robust and uniform alignment ability under mechanical buffing. 3-5 Berreman's result, 6 suggesting that LC molecules can be aligned, is mainly due to the grooves of buffed surface morphology. His concept of LC alignment can be found true in many alternative LC alignment methods, such as oblique evaporation of silicon oxide, microgroove, ⁸⁻¹⁰ surface relief grating, ^{11,12} etc. Here, we report on a novel LC alignment technique, in which we utilize a LC as an imprinting resist that can be self-aligned under 1.5 bar imprinting pressure on a large area of 4×4 cm² without a precoated alignment layer. Hence, the novel alignment technique has great potential for future LC cell fabrication and optical anisotropic film preparation.

Studies have shown that nanoimprint lithography (NIL) applications can create surface relief patterns. ^{13–16} In addition, light-emitting molecules were able to show certain directional alignment on the surface of a submicron patterned polymer matrix. ¹⁷ All patterned polymer matrices mentioned above were mainly made from amorphous polymeric materials with relatively high imprinting pressure, typically at 100 bar. ¹⁸ The high pressure applied on the resist is to overcome the hardness and viscosity of polymeric resist and to create a relief pattern by physical deformation. Along with this approach, LC was incorporated as a resist material in

this study. Our newly adopted NIL process, creating an alignment surface, is depicted in Scheme 1.

Molds with different pitches for the imprinting step were fabricated by electron-beam lithography or photolithography. The pitches were from 1.8 to 0.35 μ m with the line/space ratios from 0.4 to 2, as shown in Fig. 1. A toluene solution, comprised of SLM-90519 (reactive LC oligomer, Wacker Chemical) and Irgacure 369 (photoinitiator, Ciba), was prepared for both LC resist and LC polymer films. Under our standard process condition, we have successfully transferred the pattern of the mold to the liquid crystal resist with as little as 1.5 bar of imprinting pressure at 120 °C for 20 min on 1.7 μ m LC resist. The atomic force microscopy (AFM) results, as shown in Fig. 1, indicate that the period of molds and replicas are not identical. This is due to the shrinkage of LC resist during ultraviolet (UV) curing. The 4×4 cm² patterned area prepared by 1.8 \(\mu\mathrm{m}/\mathrm{period}\), and 200 nm in depth, showed optical anisotropy under a polarizing optical microscope (POM). Light transmission was fully dependent on the azimuthal angle of the patterned area under crossed polarizers. The results in Fig. 2 clearly showed light blockage (dark state) in the patterned area, when grating lines were parallel



SCHEME 1. (Color online) NIL fabrication for surface microstructures using the LC as a resist.

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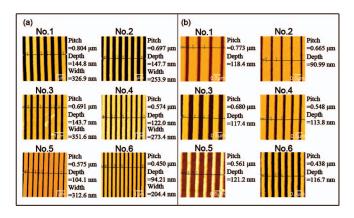


FIG. 1. (Color) AFM pictures for (a) grating patterns of molds, and (b) replicas of molds' patterns on LC resists.

to one of the polarizers' axes. The maximum light transmission (bright state) occurred when grating lines of the patterned area are placed at either 45° or 135°. In all cases above, the Grandjean texture of the unpatterned area appeared in all azimuthal angles under a POM. This is a direct evidence demonstrating that the novel NIL process is capable of aligning a LC resist. The driving force for a self-aligned LC resist can be attributed to the laminar flow and capillary effect during the hot pressing inside the mold. ¹⁹ During the pressing stage, the mold served as a confinement for the rodlike LC molecules. The various shear rate distributions derived from mechanical pressing yielded the preferable orientation along with the flow direction, causing shear-induced alignment behavior. ^{20,21}

The optical anisotropy of LC grating films made from a LC resist under this novel NIL process was further examined. The measured retardation outlined in Fig. 3 was compared to a commercial quarter-wave plate (JSR/ARTON). The data suggest that the grating film demonstrated a relatively good performance in the rest of visible range, yet superior performance within the wavelength of 400–500 nm. Furthermore, retardation of a grating film can be tuned by solid content in a LC resist solution. A larger active area of a

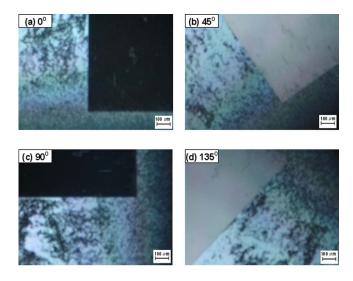


FIG. 2. (Color online) Pictures of the LC grating film under POM. The patterned area is dark in (a) and (c): The grating line direction is parallel (0°) and perpendicular (90°) to the axis of polarizer (b) and (d) The azimuthal angles at 45° and 135°, respectively.

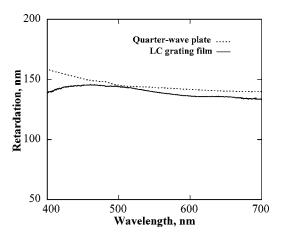


FIG. 3. The retardation value of grating film with 20% solid concentration and conventional.

grating film can be achieved in the future, depending on available molds and equipments.

We also examined LC grating films for their aligning capability by comparing them to buffed polyimide (PI) substrate and an isotropic grating film, made from Desolite 4D5-57, under the same NIL treatment. A polymerizable nematic LC (SLM-90519), the same material utilized for the LC grating film, was doped with dichroic dye (S-428, Mitsubishi Chemical) as an order indicator for evaluation, and was prepared into $3.6\pm0.3~\mu m$ thick films by spincoating. The order parameter, S, is determined by the absorption of A_{\parallel} and A_{\perp} using $S = (A_{\parallel} - A_{\perp})/(A_{\parallel} + 2A_{\perp})$ at 500 nm of the wavelength under polarized UV/visible Spectrometer.²² The high-order parameter, S=0.62, can be achieved by a LC grating film, which was comparable to the PI buffed surface (S=0.67), but much better than the one using an isotropic grating film (S =0.33), as shown in Fig. 4(a). This result suggests that the LC grating film possesses a similar alignment capability, compared with the PI buffed surface. The discrepancy between LC and non-LC resists with same microgrooves, in order parameter, led us to investigate further. In Fig. 4(b), we found that the LC molecules on the LC grating film had a better extinction ratio than isotropic grating film at all annealing and curing temperatures. Both grating films made from Desolite 4D5-57 and SLM-90519 exhibited the same relief pattern under NIL treatment, except for their molecular arrangements. LC molecules on the isotropic grating film demonstrated limited preferred orientation, due to their molecular confinement on the microgroove surfaces. In addition to the confinement effect, the order of the LC molecules on the LC grating film was enhanced by molecular arrangement. The self-aligned sample of SLM-90519, however, possesses a greater degree of molecular alignment ability, which is absent in an isotropic sample of Desolite 4D5-57. This is why the LC on SLM-90519 has much higher-order parameter than the LC on Desolite 4D5-57. These results suggest that microgroove surfaces can induce the alignment ability of LC materials. However, the alignment capability down to the molecular level is the key to achieve well-ordered LCs. Based on the crystal rotation method, the pretilt angle of the LC on the LC grating film, polyimide, and isotropic grating film were 0.1°, 1.7°, and 0.2°, respectively.

In summary, the novel NIL technique for LC alignment and preparation of optical anisotropic LC films was successfully achieved in this study. Without any prealignment layer,

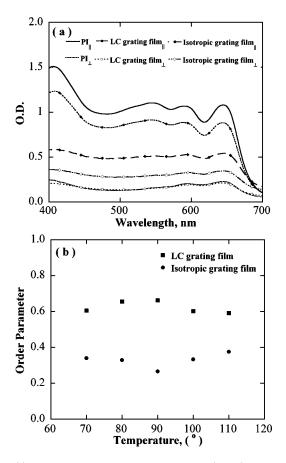


FIG. 4. (a) Linear dichroism of LC with dichroic dye (S-428) on LC grating film, isotropic grating film and PI treated alignment layer. (b) The order parameters of LC with dichroic dye (S-428) on both LC and isotropic grating film at different curing temperatures.

this simple approach can align a LC effectively. Specifically, applying low imprinting pressure at 1.5 bar on a LC resist, yielding a molecular self-aligned grating film, is much easier to operate in the practical sense. Moreover, the LC resist possesses an aligning capability that can also be utilized as an alignment layer itself. Our results also suggest that to achieve a good LC alignment, one should not solely rely on the grooved surface, but rather take into the account the surface alignment ability on the molecular level. The novel NIL technique reported in this letter opens a new gateway to LC fabrication, as well as optical device fabrication. ²³

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