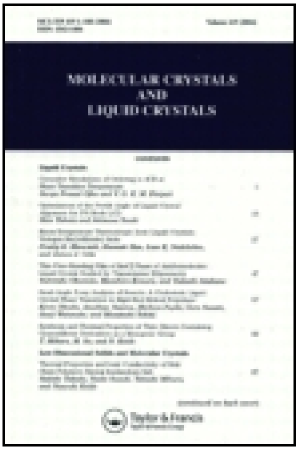

Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl19

Dipping-Induced In-Plane Molecular Alignment of LB Films

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Version of record first published: 24 Sep 2006

To cite this article: Xingze Lu, Kui Han, Yongsheng Xu, Shihong Ma & Wencheng Wang (1997): Dipping-Induced In-Plane Molecular Alignment of LB Films, Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals, 294:1, 23-26

To link to this article: http://dx.doi.org/10.1080/10587259708032239

Mol. Cryst. Liq. Cryst., 1997, Vol. 294, pp. 23–26
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DIPPING-INDUCED IN-PLANE MOLECULAR ALIGNMENT OF LB FILMS

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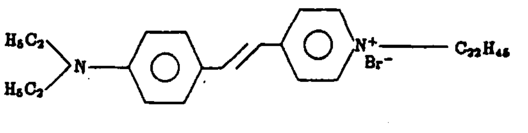
Abstract Super-quadratic growth of SHG intensity with thickness could be realized due to the extra dipping-induced and epitaxy-enhanced in-plane polarization in the hemicyanine LB multilayers.

INTRODUCTION

In surface physics, one usually assumed that molecules adsorbed on substrates with an in-plane isotropy.[1] In fact, flow orientation during transfer of Langmuir monolayer[2,3] would not only modify some concepts and theories in LB technology and nonlinear optics, but also find potential applications in areas such as liquid crystal aligning agent[4] or waveguide frequency-doubler.[5]

SAMPLE PREPARATION

The molecular structure of the hemicyanine (HEM) is:



Arachidic acid $C_{19}H_{39}COOH$ (AA) was used as inert spacer molecules to form stable noncentrosymmetric structures. Four Y-type HEM/AA interleaving multilayer samples A, B, C, D of bilayer numbers N = 6, 12, 24, 36 respectively were prepared at a surface pressure of 30 mN m^{-1} and dipping speed of 3 mm min^{-1}.

DIPPING-INDUCED ANISOTROPY

Fig.1(a) shows the normal-incidence polarized UV-visible absorption spectra of sample D with the incident field parallel (broken line) or perpendicular (solid line) to the dipping direction on the sample. The linear anisotropy parameter was defined $r_L \equiv A_\parallel/A_\perp$, where $A_\parallel$, $A_\perp$ were the areas below the respective absorption curves. We obtained $r_L = 1.66 > 1$ from Fig.1(a) revealing a significant flow orientation along the dipping direction.

The second harmonic generation (SHG) experiments were performed by using a 40ps pulsewidth, 10Hz repetition rate beam at 1.064 μm with different angles ϕ between the dipping direction and incident plane by rotating the sample around its surface normal, at an incident angle $\theta = 45^\circ$ unless otherwise stated. The transmitted SHG signals at 532nm were detected by a photomultiplier tube and a boxcar averager. The measured p-in/p-out SHG intensity $I_{pp}(\phi)$ from sample D is shown by the solid dots in Fig.1(b) displaying an asymmetric "oval" pattern with a maximum at $\phi = 0^\circ$.

Our classical nonlinear oscillator model derived[6]

$$I_{pp} \propto [\, d_h \cos\theta \, \cos\phi + d_z \sin\theta \,]^4 . \tag{1}$$

The fit result using Eq.(1) is given by the solid line in Fig.1(b) and qualitatively agreed with the experimental data.

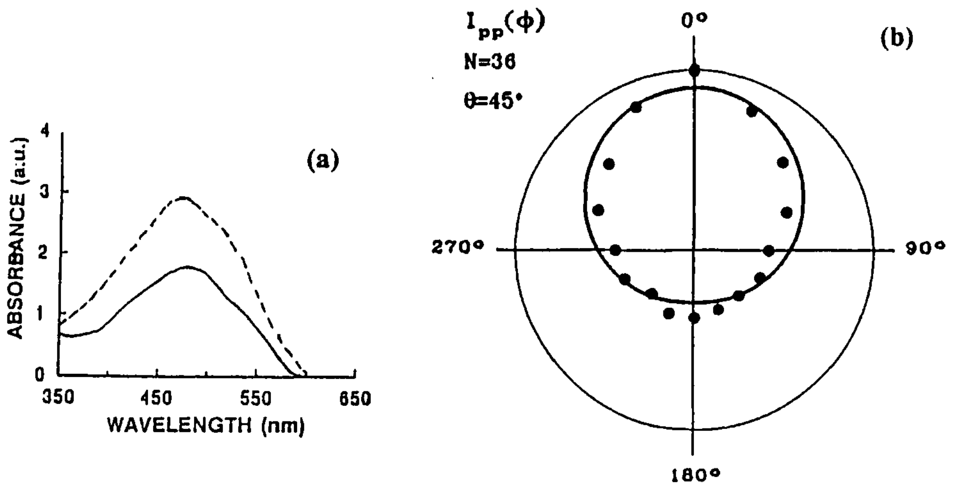


FIGURE 1 Sample D: (a) Polarized absortion spectra; (b) SHG intensity $I_{pp}(\phi)$.

EPITAXY-ENHANCED ANISOTROPY

For samples A, B, C, D, we obtained r_L = 1.10, 1.24, 1.55, 1.66 by absorption measurements. We also define a nonlinear anisotropy parameter $r_{NL} \equiv [I_{pp}(\phi=0^\circ)/I_{pp}(\phi=180^\circ)]^{1/2}$, and obtained r_{NL} = 1.07, 1.22, 1.48, 1.77 measured at $\theta = 45^\circ$ and r_{NL} = 1.06, 1.26, 1.46, 1.68 measured at $\theta = 30^\circ$ by SHG experiments. As shown in Fig.2(a), the degree of anisotropy r_L and r_{NL} increased monotonically and consistently with N, due to the interlayer interaction or "epitaxial evolution" process during deposition:[7]

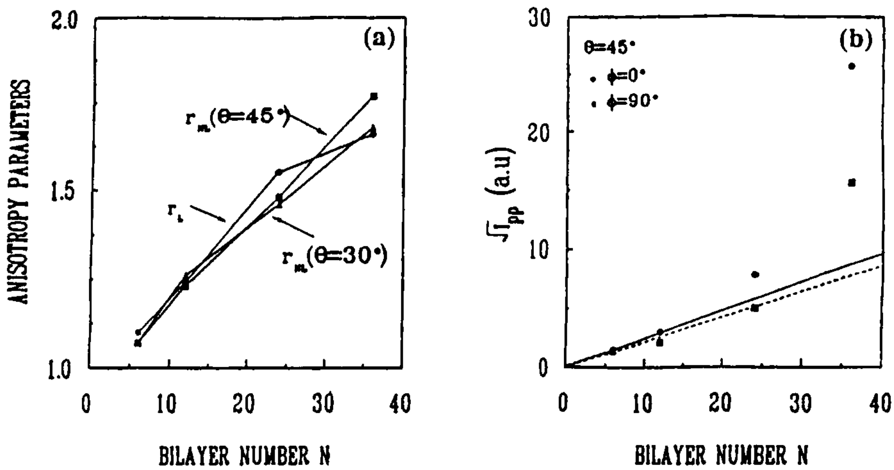


FIGURE 2 N-dependence of (a) Anisotropy parameters r_L and r_{NL}; (b) $\sqrt{I_{pp}}$ measured at $\theta = 45^\circ$.

SUPER-QUADRATICALLY ENHANCED SHG INTENSITY

From nonlinear optics, the SHG intensity

$$I_{2\omega} = \frac{2\omega^2 D_{eff}^2 I_\omega^2 L^2 \operatorname{sinc}^2(L\Delta k/2)}{c^3 \epsilon_0 n_\omega^2 n_{2\omega}} \qquad (2)$$

The L^2-dependence has been considered as an criterion and upper limit for perfectly and invariantly structured multilayer films within the quasi-phase-matching range of optical path L [$L\Delta k \ll 1$, i.e. $\mathrm{sinc}(L\Delta k/2) \approx 1$].

In the presence of the extra L-dependent polarization d_h, however, the effective second-order susceptibility D_{eff} in Eq.(2) should be replaced by

$$D_{eff}'(L) = D_{eff} [\ 1 + d_h(L) \cot\Theta\ \cos\phi\ /\ d_z\ ]^2 . \tag{3}$$

As L (or N) increased, an increase of $d_h(L)$ speeds up the intensity growth and a super-quadratic enhancement could be observed, especially for $\phi = 0^\circ$, if the intensity reduction due to structural deterioration, low transfer ratio and phase mismaching were insignificant.

Fig.2(b) gives the data for $\sqrt{I_{pp}}$ with $\phi = 0^\circ$ (soid dots) and $\phi = 90^\circ$ (solid squares) vs N. Super-linear growth of $\sqrt{I_{pp}}$, i.e. Super-quadratic increase of I_{pp} with N were clearly seen by the data points well above their respective straight lines, especially for $\phi = 0^\circ$ and larger N's.

ACKNOWLEDGEMENTS

This work was supported by the National Natural Science Foundation of China under the grant #19434011.

REFERENCES

1. Y.R.Shen, Nature 337, 519 (1989).
2. N.Minari, K.Ikegami, S.I.Kuroda et al, J. Phys. Soc. Jpn. 58, 222 (1989).
3. K.Ikegami, S.I.Kuroda, Y.Tabe et al, Jpn. J. Appl. Phys. 31, 1206 (1992).
4. Y.M.Zhu, Z.H.Lu, X.B.Jia et al, Phys. Rev. Lett. 72, 2573 (1994).
5. Ch.Bosshard, M.Florsheimer, M.Kupfer et al, Opt. Comm. 85, 247 (1991).
6. X.Z.Lu, K.Han, S.H.Ma et al, J. Phys. D: Appl. Phys. 29, 1576 (1996).
7. I.R.Peterson, G.J.Russell, Thin Solid Films 134, 143 (1988).