O films with adjustable tilt of optical axis for LCD compensation

Oleg Yaroshchuk (SID Member)
Leonid Dolgov
Jacob Ho
Hoi-Sing Kwok (SID Fellow)
Vladimir Chigrinov (SID Fellow)
Haruyoshi Takatsu
Hiroshi Hasebe (SID Member)

Abstract — A method of preparation of positive O films with the tilt angle of the optic axis continuously controlled in the range 0–90° is proposed. It is based on the use of reactive mesogens and alignment materials that provide a wide range of pretilt angles. The method developed allows for further improvement in the viewing-angle characteristics of LCDs with O compensation films.

Keywords — *Compensation film, O film, reactive mesogen.*

1 Introduction

Liquid-crystal displays (LCDs) excel in low power consumption and operational voltage, high luminance and resolution, color gamut, and excellent contrast at direct viewing. However, oblique viewing of LCDs leads to a loss of contrast, occurrence of gray-level inversion, and color shifts. These undesired effects result from the intrinsic birefringence of the liquid-crystal (LC) layers, which causes an angular dependence of the phase retardation for the light passing though LCD device. There are known various techniques to improve the viewing-angle performance of LCDs. The preferred technique uses supplementary birefringent films, which compensate undesirable phase retardation for obliquely incident light.

The most popular for TN-LCD compensation film is Fuji film, which consists of an aligned discotic layer with the optic axis tilt smoothly changed in the film-thickness direction. This film provides substantial improvement in the angular dependence of contrast and the gray-level stability. At the same time, primary Fuji film, commercialized in 1999, causes a rather strong color shift. According to Schadt *et al.*,[1,2] optical phase retardation typical of Fuji film can be attained by using two positive O films with crossed optical axes. The authors made these films from calamitic reactive mesogens (RM) similar to those published by Broer.[3] The compensation of a TN-LCD with a stack of two crossed O films results in a viewing-angle dependence of the contrast similar to that of Fuji film. In parallel, the color shift is greatly improved. Further improvement can be attained by using O films with a non-uniform tilt in the thickness direction adjustable at only one[2,4] or both interfaces.[5] In addition to advancements in TN-LCDs, calculations performed[4,5] predict great improvement for the angular dependence of the contrast of OCB- and VA-LCDs compensated by O films.

Thus, optimization of LCDs with integrated O compensators, from the viewpoint of contrast angular dependence, gray-level stability, and color shift, requires the technology of O films, providing a continuous variation of O film parameters, such as thickness, birefringence, and optic-axis profile. The present paper offers such technology. We use an approach developed for conventional liquid crystals,[5,6] which provides a continuous change in the LC pretilt angle from 0° to 90°. This variation is attained by using a mixture of two polyimides designed for planar and homeotropic alignment (polyimides p-PI and h-PI, respectively). The p-PI/h-PI layers are backed and unidirectionally rubbed to impart alignment function. The pretilt angle is continuously tuned with a change of ratio of PI concentrations. We demonstrate that this method well suits reactive mesogens, thus allowing us to prepare O films with variable optic-axis profiles and almost complete control of the tilt angle of the optic axis in the range 0–90°.

2 Experimental

2.1 Alignment substrates

We combined p-PI and h-PI precursors purchased from the Japan Synthetic Rubber Co., providing excellent alignment and continuous pretilt-angle control for conventional nematic liquid crystals.[6,7] The p-PI and h-PI give nominal LC pretilt angles of 88° and 5°, respectively. These polyimides were diluted in standard solvent provided by JSR, allowing them to mix. The concentration of h-PI precursor was varied from 0 to 100% by weight to provide a different pretilt angle of the RM. The resulting solution was spin-coated on glass slides to form the alignment film. The film was first pre-annealed at 90°C for 5 min and then annealed at 230°C for 90 min where most imidization took place. Finally, the PI layers were unidirectionally rubbed by a conventional rubbing machine.

O. Yaroshchuk and L. Dolgov are with the Institute of Physics, National Academy of Sciences of Ukraine, Department of Physics of Crystals, prospect Nauki 46, 03028 Kyiv, Ukraine; telephone +380-44-525-2424, fax –1589, e-mail: olegyar@iop.kiev.ua.

J. Ho, H-S. Kwok, and V. G. Chigrinov are with Hong Kong University of Science and Technology, Department of Electronic & Electrical Engineering, Clear Water Bay, Kowloon, Hong Kong.

H. Takatsu and H. Hasebe are with Dainippon Ink and Chemicals, Inc., Corporate R&D Div., Japan.

2.2 Reactive mesogen films

The aligned RM films were obtained (1) by filling RM in a sandwich cell (confined films) and (2) by disposing RM on a single substrate (coated films).

To prepare *confined films,* sandwich cells were assembled from two substrates so that the aligning PI films were from the inner side of the substrates, and the rubbing directions were anti-parallel. The gap between substrates was maintained by 4.5-μm spacers. The cell was glued with Norland 65 photoadhesive by printing it on the edge of one substrate before cell assembly. The glue was hardened by UV illumination (7 mW/cm^2, 365 nm, 1 min). The prepared cells were filled with a low-viscosity nematic RM UCL-011 from DIC. The filling was realized in low vacuum at 50°C. After filling, the cell was illuminated by UV light with an intensity of 7 mW/cm^2 at 365 nm to solidify the RM film.

The *coated RM films* were obtained by spin-coating RM solution on the rubbed PI layer. In these experiments, along with RM UCL-011, we used two viscous nematic mixtures, UCL-017 from DIC and RMM256C from Merck, designed for planar alignment. All mixtures were dissolved in toluene at 30 wt.%. The films were spin-coated at 3000 rpm for 30 sec and then kept at 60°C for 1 min to remove residual solvent and improve RM alignment. The film thickness, detected by the interference method, was 1.5–2 μm. For solidification, the films were illuminated by UV light (7 mW/cm^2, 365 nm) in argon atmosphere.

2.3 Film characterization

The alignment of RM layers was tested by viewing samples between a pair of polarizers and by sample observation in polarizing microscope. The anisotropic properties were studied by the transmission null ellipsometry technique. In these experiments, the light beam from the He–Ne laser (λ = 0.63 nm) passed through an optical system consisting of a fixed polarizer, an anisotropic sample, a quarter-wave plate, and a rotating analyzer and was registered with a photodiode. The sample was set so that its slow axis or, more precisely, rubbing direction, was oriented vertically or horizontally. The polarizer axis formed an angle of 45° with the sample's slow axis and 0° with the slow axis of the retardation plate. The analyzer rotation angle φ, corresponding to the minimal transmittance of the laser beam, was experimentally measured for different incidence angles of this beam on the sample, *i.e.*, for different sample rotation angles α. The measured φ *vs.* α curves were fitted in frame of the most appropriate uniaxial model. The fitting yielded retardation $(n_e - n_o)d$ and a tilt angle of the optical axis θ. The details of this ellipsometric method can be found in our previous papers.[8,9] The tilt angle of the optic axis for the confined films was also determined by a well-known crystal rotation method.[10]

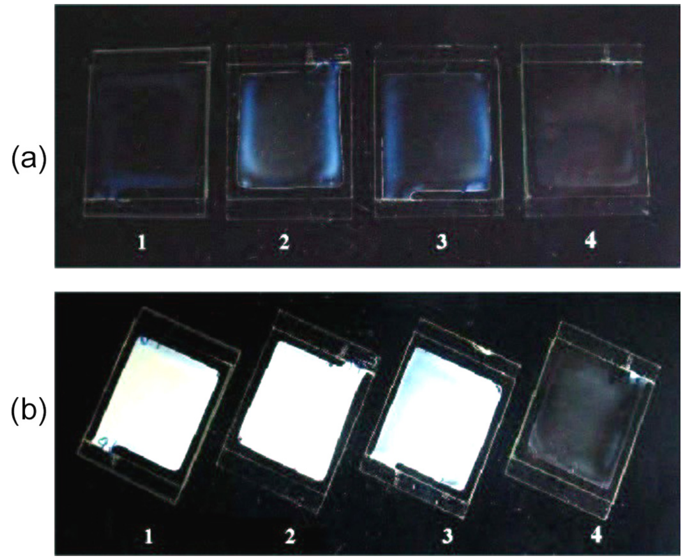

FIGURE 1 — The cells filled with RM UCL-011 viewed between a pair of crossed polarizers: (a) the in-plane projection of the optical axis is parallel to the polarization direction of incident light; (b) the in-plane projection of the optical axis is rotated with regard to the polarization direction of incidence light. The tilt angle of the optic axis is 3, 33, 56, and 89° in the cells 1, 2, 3, and 4, respectively.

3 Results and discussion

3.1 Confined O films

Figure 1 shows a set of samples, viewed between a pair of crossed polarizers, corresponding to different ratios of the p-PI and h-PI precursors in the alignment film. There is evident excellent uniformity of RM films. One can also observe variation of the tilt angle of the optic axis with a ratio of p-PI and h-PI concentrations.

Figure 2 demonstrates the ellipsometric curves for the three samples from the series shown in Fig. 1. The experimental curves (dots) are fitted well in the frame of the model of the uniaxial orientational structure with a tilted optical axis (O film model). This means that the intrinsic liquid-crystalline order of the RM is preserved in the photopolymerization process and thus the RM layer can be considered as a frozen liquid crystal. The fitting parameters corresponding to the experimental ellipsometric curves are summarized in Table 1. One can see that the tilt angle of optic axis θ, accounted from the film surface, varies from 3° to 89°, running from sample 1 to sample 4. Excellent fitting of the experimental curves in the frame of an uniaxial model

TABLE 1

Cell No.	Concentration of h-PI (wt.%)	Fitting of ellipsometric curves		Fitting of crystal rotation curve
		Tilt angle (°)	$(n_e - n_o)d$ (nm)	Tilt angle (°)
1	0	3	225	3
2	10	35	225	33
3	15	55	225	56
4	20	65	225	64
5	100	89	225	89

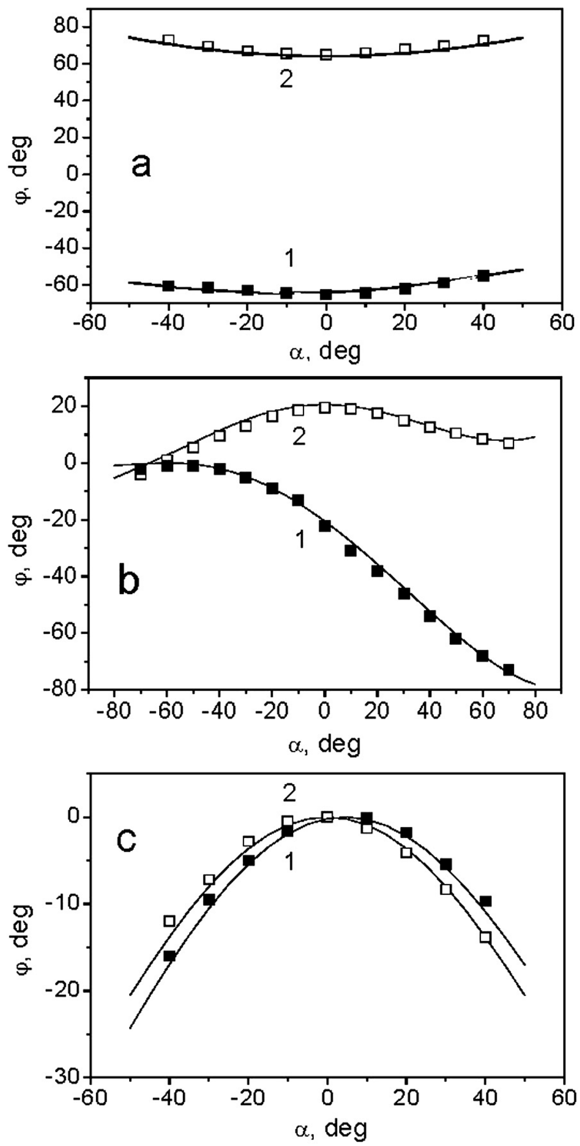

FIGURE 2 — Measured (dots) and modeled (solid lines) analyzer *vs.* light-incidence-angle curves for cells 1(a), 2(b), and 4(c) from Fig. 1. Curves 1 and 2 correspond to two measuring positions of the cell, when in-plane projection of the optic axis is oriented horizontally and vertically, respectively.

implies high uniformity of the optic axis across the film. This allows for the application of a conventional crystal rotation method to evaluate the tilt angle of the optic axis θ. The corresponding values of θ are also presented in Table 1. There is evident excellent coincidence of data obtained by the null ellipsometry and crystal rotation methods.

3.2 Coated O films

Figure 3 shows that coated RM films, same as the confined films, are of good optical quality. No considerable alignment defects are detected on the macroscopic and microscopic levels as well. The difference in the brightness of the films presented in Fig. 3 is caused by the difference in the optic-axis tilt.

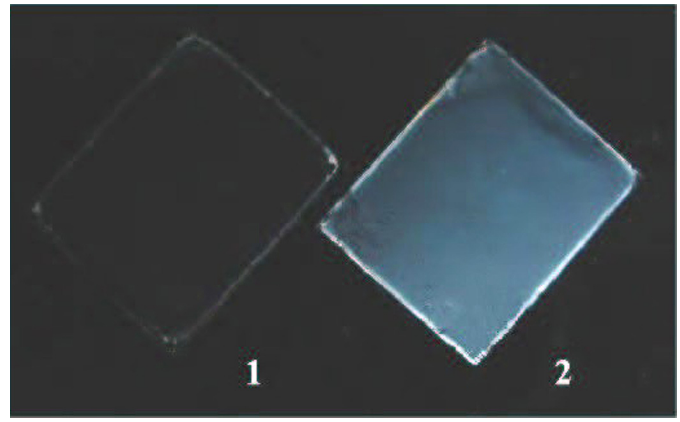

FIGURE 3 — The films of UCL-011 (1) and UCL-017 (2) coated over rubbed p-PI/h-PI (4:1) layers viewed between a pair of crossed polarizers. The in-plane projection of the optical axis is rotated with regard to the polarization direction of incidence light.

The ellipsometric curves measured for films 1 and 2 (Fig. 3) are shown in Fig. 4(a) and 4(b), respectively. The solid lines correspond to φ *vs.* α curves calculated in the frame of an uniaxial O film model and fitted to experimental data. One can notice that fitting is not as good as in case of

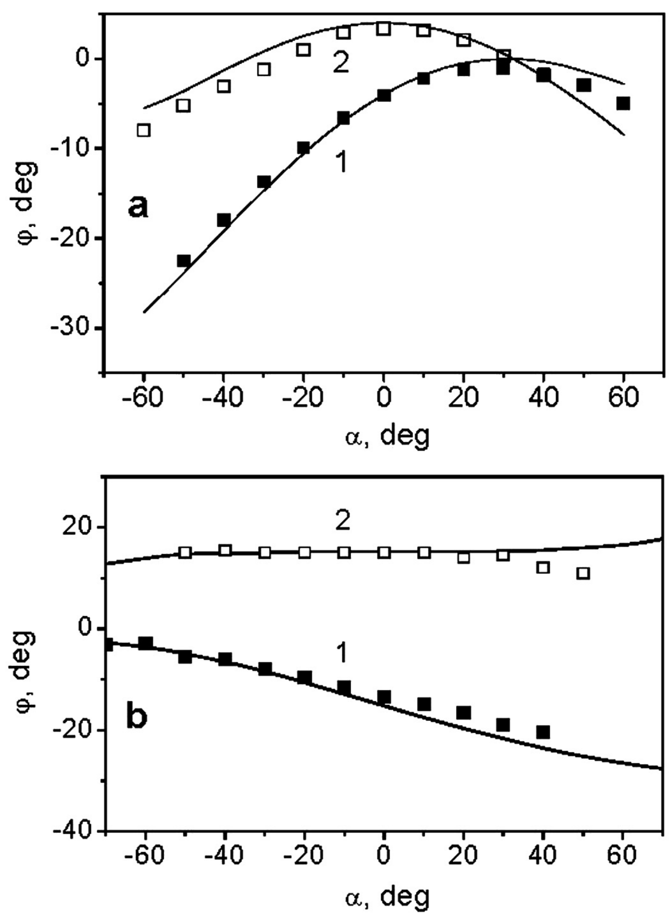

FIGURE 4 — Measured (dots) and modeled (solid lines) analyzer *vs.* light-incidence-angle curves for cells 1(a) and 2(b) presented in Fig. 3. Curves 1 and 2 correspond to two measuring positions of the cell when in-plane projection of the optic axis is oriented horizontally and vertically, respectively.

TABLE 2

Cell No.	Concentration of h-PI (wt.%)	RM	Fitting of ellipsometric curves	
			Tilt angle (°)	$(n_e - n_o)d$ (nm)
1	20	UCL-011	70	85
2	20	UCL-017	30	78
3	20	RMM256C	30	80

confined films. This suggests that the coated films are not ideally uniaxial. For this reason, the fitting yields averaged parameters of the coated films, which must be treated with some care.

The fitting results obtained for different types of RM films are given in Table 2. There is evidence that the effective tilt of the optic axis for UCL-011 film is much higher than that for the films of UCL-017 and RMM256C planar mixtures, which usually contain planarization agents.[11] Furthermore, the tilt angle of UCL-011 film coated on the PI substrate is even higher than that of the film confined between two aligning PI substrates, providing the same angle of LC pretilt (70° *vs.* 65°). This implies a non-uniform alignment of the optic axis (RM director) across the open films. In contrast to the confined UCL-011 layer uniformly aligned [Fig. 5(a)], the coated UCL-011 film presumably has a splayed structure [Fig. 5(b)]. It is formed because of different anchoring conditions on the bottom and top surfaces; the bottom pretilt angle is determined by the PI substrate, while the top one by the surface free energy of the RM in contact with air. In the latter case, a homeotropic or high tilt alignment is usually realized.[9,12] To suppress a homeotropic alignment on the border with air, reactive mesogen mixtures are usually doped with planarization agents, surfactants in the most common case.[11] The UCL-017 and RMM256C films, containing planarization additives, have a structure shown in Fig. 5(c). This is consistent with a low effective angle of the optic-axis tilt obtained for these films. Thus, along with classical uniaxial O film, splayed O films are realized. The optic-axis profile in these films can be controlled by the ratio of h-PI and p-PI alignment materials and planarization additives to the RM.

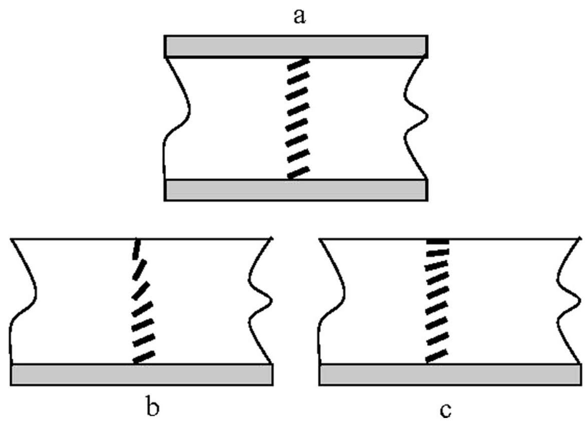

FIGURE 5 — Optic axis profiles in the prepared O films.

4 Conclusions

We attained RM alignment with the pretilt angle controllable in practically the full range from 0° to 90°. Optically, these RM layers act as positive O films, which transform to positive A film and positive C film as the pretilt angle approaches 0° and 90°, respectively. The structure of the RM layer is preserved in the solidification process. This allowed us to obtain solid O films with a controllable optic axis. The prepared O films are confined between two aligning substrates or coated on a single aligning substrate. In the first case, RM alignment is uniform along the film normal, while in the second case a splayed structure is formed, which can be controlled by the alignment material and the content of the RM mixture. Despite the use of glass substrates in our experiments, the approach is applicable to plastic substrates. Moreover, RM film can be striped off to be used separately from the alignment substrates.

The potential of this developed approach is not limited to the structures presented in this paper. It can be used to produce twisted O films by analogy with twisted A films earlier produced.[13,14] Furthermore, it can be also extended to photoalignment by using photosensitive p-PI and/or h-PI materials as it was recently realized for conventional LC[15]. In this way, patterned O films can be easily obtained, which, compared with known solutions,[16,17] open new compensation possibilities for recently developed LCD modes (*e.g.*, transflective and stereoscopic LCDs).

Thus, in general, our approach yields different types of O films with adjustable optical characteristics. This allows further improvement of traditional LCDs and newly developed LCDs as well. The proposed versatile approach is rather simple and can be easily realized by using common facilities of LCD manufacturing.

Acknowledgments

The support from HKUST (grant CERG 612406) and NAS of Ukraine (grant No 93/07-H) is acknowledged.

References

1 J. Chen, K. Chang, J. DelPico, H. Seiberle, and M. Schadt, "Integral-color plastic LCD: the ultimate flat-panel display," *SID Symposium Digest Tech. Papers* **30**, 1054–1057 (1999).

2 T. Bachels, J. Fuenfschilling, H. Seiberle, M. Schadt, G. Gomez, and E. Criton, "Novel photo-aligned LC polymer compensation film for wide-view TN displays," *J. Soc. Info. Display* **11**, 519–524 (2003).

3 D. Broer, J. Boven, G. Mol, and G. Challa, "In-situ photopolymerization of oriented liquid-crystalline acrylates," *Macromol. Chem.* **190**, 2255–2268 (1989).

4 K. Hosaki, T. Uesaka, S. Nishimura, and H. Mazaki, "Comparison of viewing angle performance of TN-LCD and ECB-LCD using hybrid-aligned nematic compensators," *SID Symposium Digest Tech. Papers* **37**, 721–724 (2006).

5 T. Ishikawa and S.-D. Mi, "Compensation of various LCD modes by positive O-plates," *SID Symposium Digest Tech. Papers* **37**, 1166–1168 (2006).

6 F. Yeung, J. Ho, Y. W. Lee, F. C. Xie, O. Tsui, P. Sheng, and H. S. Kwok, "Variable liquid crystal pretilt angles by nanostructured surfaces," *Appl. Phys. Lett.* **88**, 051910–051912 (2006).

7 F. Yeung, F. Xie, J. Wan, F. Lee, O. Tsui, P. Sheng, and H. S. Kwok, "Liquid crystal pretilt angle control using nanotextured surfaces," *J. Appl. Phys.* **99**, 124506–124509 (2006).
8 O. Yaroshchuk, T. Sergan, J. Lindau, S. N. Lee, J. Kelly, and L. C. Chien, "Light induced structures in liquid crystalline side-chain polymers with azobenzene functional groups," *J. Chem. Phys.* **114**, 5330–5337 (2001).
9 O. Yaroshchuk, T. Sergan, and J. Kelly, "Ellipsometry studies of 3D orientational structures in thin liquid crystal layers," *Mol. Cryst. Liq. Cryst.* **367**, 141–150 (2001).
10 G. Baur, V. Wittwer, and D. W. Berreman, "Determination of the tilt angles at surfaces of substrates in liquid crystal cells," *Phys. Lett. A* **56**(2), 142–144 (1976).
11 O. Parri, D. Coates, S. Greenfield, and I. Bonny, U.S. Patent No. 5,622,648 (1997).
12 J. West, G. Magyar, J. Kelly, S. Kobayashi, Y. Iimura, and N. Yoshida, "Dichroic ultraviolet absorption of thin liquid crystal films," *Appl. Phys. Lett.* **67**(2), 155–157 (1995).
13 H. Hasebe, K. Takeuchi, and H. Takatsu, "Properties of novel UV-curable liquid crystals and their retardation films," *J. Soc. Info. Display* **3**(3), 139–143 (1995).
14 O. Yaroshchuk, J. Ho, V. Chigrinov, and H. S. Kwok, "Azodyes as photoalignment materials for polymerizable liquid crystals," *Jpn. J. Appl. Phys.* **46**(5A), 2995–2998 (2007).
15 J. Ho, V. Chigrinov, and H. S. Kwok, "Variable liquid crystal pretilt angles generated by photoalignment of a mixed polyimide alignment layer," *Appl. Phys. Lett.* **90**, 243506 (2007).
16 B. M. I. van der Zande, C. Doornkamp, S. J. Roosendaal, J. Steenbakkers, A. Op't Hoog, J. T. M. Osenga, J. J. Van Glabbeek, L. Stofmeel, J. Lub, M. Shibazaki, K. Asahara, M. Shibazaki, T. Inada, M. Yoshiga, and S. Kawata, "Technologies towards patterned optical foils applied to transflective LCDs," *J. Soc. Info. Display* **13**(8), 627–632 (2005).
17 B. M. I. van der Zande, J. Lub, H. J. Verhoef, W. P. M. Nijssen, and S. A. Lakehal, "Patterned retarders prepared by photoisomerization and photopolymerization of liquid crystalline films," *Liq. Cryst.* **33**, 723–737 (2006).

Oleg Yaroshchuk received his Ph.D. degree in solid-state physics from the Institute of Physics (IOP), National Academy of Sciences of Ukraine, in 1990. He is currently a senior research fellow and leader of an informal group in the Department of Crystals of IOP. His research area includes alignment methods for liquid crystals and liquid-crystal polymers, heterogeneous liquid-crystal media, and non-linear optics. He has published more then 120 scientific papers and authored more then 15 inventions. He is a member of SID and ILCC.

Leonid Dolgov received his M.S. degree in physics from Chernihiv State University in 1998. He joined the Institute of Physics of the National Academy of Sciences, Kyiv, Ukraine, in 2001, as a Ph.D. student. He is currently a scientific research at the Liquid Crystal Department. His research interests include light-scattering liquid-crystal composites. In 2007, he defended his Ph.D. thesis in the physics of heterogeneous liquid-crystal media. He has authored 12 papers.

Hoi-Sing Kwok obtained his Ph.D. degree in applied physics from Harvard University in 1978. He joined the State University of New York at Buffalo in 1980 and became a Full Professor in 1985. He joined HKUST in 1992 and is currently Director of the Center for Display Research. He has over 300 refereed publications and holds over 10 patents in laser optics and LCD technologies. He is a Fellow of the Optical Society of America and a Fellow of IEEE.

Vladimir G. Chigrinov obtained his Ph.D. degree in solid-state physics from Shubnikov Institute of Crystallography, USSR Academy of Sciences in 1978. In 1988, he defended the doctoral degree and in 1998 became a Professor at the Shubnikov Institute of Crystallography, where he was a Leading Researcher since 1996. He joined HKUST in 1999 and is currently an Associate Professor. He has authored two books, 14 reviews and book chapters, 131 journal papers, and 47 patents in the field of liquid crystals since 1974. He is a member of Editorial Board of *Liquid Crystal Today* and is Vice-President of the Russian SID Chapter.

Jacob Y. L. Ho received his B.S. degree in engineering physics from the HK Polytechnic University in 1993, MPhil degree in physics, and M.Sc. degree in electronic engineering from the HK University of Science and Technology (HKUST) in 1995 and 1999, respectively. He is currently a senior technician in the Department of Electronic Engineering of HKUST.

Haruyoshi Takatsu received his B.S. and M.S. degrees in applied chemistry from Waseda University in 1971 and in 1973, respectively. He joined Dainippon Ink & Chemicals in 1973 and has been engaged in the research and development of liquid crystals. He received his Ph.D. in liquid crystals from Waseda University in 1986. He was a chief researcher and in 1996 received the Technology Award from the Japan Chemical Industry Association on Development and Commercialization of Nematic Tolans. He received an Achievement Award from the Japanese Liquid Crystal Society in 2002. He is an inventor of 149 issued Japanese patents and 51 United States patents relating to liquid-crystal materials and displays. At present, he is General Manager, R&D, of the Liquid Crystal Technical Department of Dainippon Ink & Chemicals and has been working on the research and development of liquid-crystal materials and displays.

Hiroshi Hasebe received his B.E. and M.E. degrees in functional polymer chemistry from Shinshu University in 1989 and 1991, respectively. He joined Dainippon Ink & Chemicals in 1991 and has been engaged in research and development related to liquid crystals. He received his Ph.D. degree in liquid crystals from Shinshu University in 1998. He is an inventor of 23 Japanese patents and three United States patents in the field of liquid crystals.