Photo-aligned ferroelectric liquid crystals in microchannels

Daniel Budaszewski,[1,*] Abhishek K. Srivastava,[1] Alwin M. W. Tam,[1] Tomasz R. Wolinski,[1] Vladimir G. Chigrinov,[2] and Hoi-Sing Kwok[2]

[1]*Optics and Photonics Division, Faculty of Physics Warsaw University of Technology, 00662 Warsaw, Koszykowa 75, Warsaw, Poland*

[2]*Department of Electronics and Computer Engineering, Hong Kong University of Science and Technology, Clear Water Bay, Kowloon, Hong Kong, China*

**Corresponding author: danielb@if.pw.edu.pl*

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In this Letter we disclose a method to realize a good alignment of ferroelectric liquid crystals (FLCs) in microchannels, based on photo-alignment. The sulfonic azo dye used in our research offers variable anchoring energy depending on the irradiation energy and thus provides good control on the FLC alignment in microchannels. The good FLC alignment has been observed only when anchoring energy normalized to the capillary diameter is less than the elastic energy of the FLC helix. The same approach can also be used for the different microstructures viz. photonic crystal fibers, microwaveguides, etc. which gives an opportunity for designing a photonic devices based on FLC.

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Nowadays a great deal of interest has been devoted to the investigation of microstructured optical waveguides that merge unique properties of photonic crystal fibers (PCFs) [1] and liquid crystal (LC) materials. This combination, called photonic liquid crystal fibers (PLCFs), is of interest to many research groups all over the world.

In general, PCFs can be designed in various configurations and geometries, in which a fiber core is present as a solid rod or an air hole, both surrounded by a periodic matrix of microholes. But one of the most attractive properties of PCFs is that their air-hole periodical structure can be infiltrated with different substances whose physical properties can be controlled by external factors, such as electromagnetic fields, pressure, or temperature. One of the most suitable materials for infiltrating these microholes are LCs. The experimental evidence of tuning guiding properties of PLCFs obtained by infiltration has been reported since 2003 by many research groups [2–4]. The vast majority of research work has been done by employing nematic liquid crystals (NLCs) that resulted in some practical applications including optical filters, attenuators [5,6], polarization controllers [7], and long-period fiber gratings [8]. The current progress and state-of-the-art research in the field of PLCF technology has been reported by Zagrafopoulos *et al.* recently [9]. Some of the initial studies, in the past include fiber infiltrated with cholesteric and smectic LCs [10–12]. However, alignment quality and stability is still a big issue.

Chiral smectic C (SmC*) LCs, also called as ferroelectric liquid crystals (FLCs), have been of great interest for many research groups mainly due to their outstanding properties such as fast electro-optical response at low electric fields, i.e., an order of magnitude faster than commonly used in NLCs. Other advantages of SmC* LCs worth mentioning are bistability and optical memory, which are very important in LC display technology. SmC* LCs characterized by these parameters can also be considered as promising candidates for other innovative technological applications, particularly for optical fiber devices used in sensors or tunable telecommunication filters. In contrast to typical FLC applications in LC display technology, in confined structures the FLC characteristics and properties play a predominant role in a cylindrical geometry of the microchannels.

The crucial issue in designing and fabricating stable PLCF-based devices and their characterization is the molecular alignment of LCs inside the microstructured PCFs to allow desirable propagation conditions as well as electro-optic modulation. Generally, NLC molecules tend to align homogenously inside the cylindrical cavities that can be obtained either by orientation by flow or by van der Waals interaction between LC molecules responsible for alignment. Liou *et al.* have proposed a method of aligning NLC molecules doped with photosensitive 4-methoxyazobenzene (4MAB). The advantage of this technique is the rewritability of the NLC alignment controlled by irradiation with two different wavelengths [8]. However the azo dyes in the LC medium are not stable [13].

In the case of FLC molecules, these can be aligned in a different way depending on the FLC elastic energy of the helix, tilt angle, helical pitch, and anchoring energy of the alignment. As it was previously reported it is possible to obtain a homeotropic alignment [14], escaped radial alignment [15], or even planar alignment of the FLC molecules inside the optical waveguides by treating the inner side of the microcapillary (MC) with different aligning agents. In the case of the planar alignment of FLC molecules, Wong *et al.* proposed a photo-aligning method based on a photosensitive solution of polyimide with azo-dye [16]. However, the resulting alignment gives quite unstable alignment with low quality of the FLC molecules with small helical pitch in comparison to inner diameter of MC.

In this Letter we disclose a photo-alignment method based on a photosensitive sulfonic azo-dye material SD1 [17,18], which gives an excellent LC alignment in the microchannels and thus could find application in a broad range of devices [19,20].

The physical explanation of photo-alignment process as well as the role of surface interaction with photo-alignment agents has been already described in detail elsewhere [17,21]. An important feature of the SD1 is rewritability of the easy axis of the alignment layer that allows the alignment direction to be controlled and the ability to rewrite the pattern alignment depending on the UV light irradiation azimuth. Furthermore, the anchoring energy of such an alignment layer can be tuned by the irradiance doses.

Recently, it has been revealed that a proper balance between the anchoring energy (normalized to the cell gap) and the elastic energy of the helix with the condition that the FLC layer thickness is larger than the helical pitch of the FLC, which provides a good alignment for FLCs [22–27]. In this condition the helical pitch is not suppressed by the surface and therefore provides excellent alignment conditions. However, application of a small electric field (larger than the critical field of helix unwinding, usually ~0.1–1 V/μm) compels the FLC helix to unwind and provide exactly the same dynamics as for a surface stabilized FLC (SSFLC).

The advantage of ESH mode [23] is its fast response time and high contrast ratio with defect-free geometry at a very low electric field. Therefore, in order to achieve a good alignment in microchannels the anchoring energy normalized to the FLC layer thickness should be comparable with the elastic energy of the FLC material.

To make the problem simpler, first we have chosen single silica glass MC's with inner diameters of 6 μm. The FLC FD4004N from Dainippon Ink & Chemicals Ltd. Japan (DIC) has been chosen for the entire study. The phase transition scheme for this mixture is SmC$*$ → SmA → N$*$ → Iso at temperatures of 72°C, 85°C, and 105°C, respectively. The helical pitch at room temperature for this material is $\rho = 350$ nm, spontaneous polarization $Ps = 61\ \mathrm{nC/cm^2}$, and the tilt angle is $\theta = 22.5°$.

According to the FLC material parameters described here and the electro-optical dynamic response described in [25] and [26] the elastic energy of the FLC helix is $\sim 890\ \mathrm{J/m^3}$ thus for the good alignment our anchoring energy for the FLC layer thickness (i.e., 6 μm) should be $\sim 6 \times 10^{-4}\ \mathrm{J/m^2}$ [26]. Furthermore, the photo-alignment based on the SD1 azo-dye provides precise control on the anchoring energy; the variation of the anchoring energy against the irradiation energy is shown in Fig. 1.

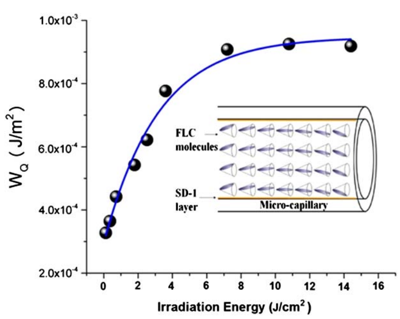

Fig. 1. Variation of anchoring energy for SD-1 against the radiation energy. Insertion shows this schematic alignment of FLC molecules in MC.

In Ref. [26] it was demonstrated that the exposure energy $\sim 3\ \mathrm{J/cm^2}$, at $\lambda = 350$ nm, could provide the required anchoring energy for the FLC in the MC. Thus samples with required conditions have been prepared as follows. Initially a 5 cm long MC with an inner diameter (d) of 6 μm has been cleaned in the UVO cleaner to improve the surface adhesion of the inner wall of the MC. Thereafter, the specimen was infiltrated with 5% wt./wt. solution of SD1 dissolved in N, N dimethylformamide (DMF). From Ref. [20] we know this concentration of SD1 in DMF is the optimal for photo-alignment of LC molecules in PCFs. Later on the SD1 infiltrated samples were placed in the oven at a temperature of 140°C for 20 min to promote the adhesion of azo-dye molecules on the inner surface of the PCF. Afterward the solution was evacuated from the specimen by applying mild air pressure and the samples were heated again for 20 min at a temperature of 140°C to evaporate the remaining amount of solvent. The detailed infiltration procedure has been described in Ref. [20].

After making the SD1 layer inside the MC, the sample was irradiated with linearly polarized deep UV light ($\lambda = 350$ nm) to induce optical anisotropy of the photo-aligning layer and thus a preferred direction of easy axis was established. The azo-dye SD1 molecules under the UV exposure align its chromophore perpendicularly to the polarization azimuth of the impinging UV light. As per irradiation energy estimations described the samples were exposed by the UV light irradiation energy in the range from 1.5 to $4\ \mathrm{J/cm^2}$. This exposure provided uniform alignment easy axis with different anchoring energy [26].

Thus the prepared samples were infiltrated with FLC FD 4004N in the following way. First, one end of the MC was dipped in to the container with FLC and placed in the oven for 1 h at a temperature of 120°C in which FLC was in isotropic phase. After infiltration the samples were placed on a hot plate and cooled down slowly for 2 h, going from a temperature of 120°C to the room temperature. This step is especially crucial in the fabrication of PLCF infiltrated with FLC because it allows the formation of well-aligned SmC* layers inside the MC.

The quality of FLC alignment inside the MC has been analyzed optically by using a polarization microscope with crossed polarizers. The schematic of layer alignment of the FLC molecules in the MC is shown in the inset of Fig. 1. The helix axis orientation was along the MC longitudinal axis, and—according to our analysis—only parallel alignment of the FLC molecules in the microchannel was possible. In the experiment, the MC was exposed to the polarized UV light with a plane of polarization perpendicular to the axis of the microchannel and consequently a parallel alignment was obtained. However, during irradiation of the sample with a plane of polarization of impinging light parallel to the axis of MC (that should result in perpendicular orientation to the MC axis) we have not obtained any alignment. This could be caused by the cylindrical geometry of the MC as well as by boundary conditions. Therefore, it can be concluded that the helix axis of the SmC* LC is parallel to the axis of the microchannel. As shown in Fig. 1, red color of the LC layer is a result of SmC* molecules tilts in each layer that induces small birefringence [Figs. 2(f) and

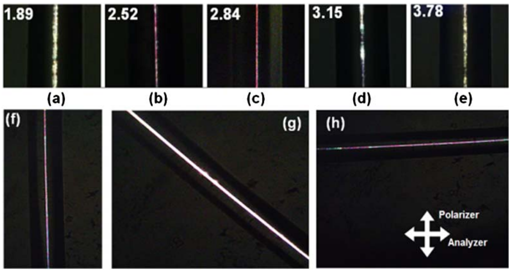

Fig. 2. Microphotographs for the MC ($d = 6$ μm) with uniform alignment of FLC molecules with different irradiation doses (a) 1.89 J/cm^2, (b) 2.52 J/cm^2, (c) 2.84 J/cm^2, (d) 3.15 J/cm^2, (e) 3.78 J/cm^2. (f), (g), and (h) show the MC with irradiation dose of 2.84 J/cm^2 with MC channel and easy axis parallel to polarizer (f), tilted at 45° to polarizer (g) and parallel to the analyzer (h).

2(h)]. Furthermore, the azimuth between the MC and crossed polarizers was 45°, then SmC* helical axis alignment was also at 45° and hence we observed a higher intensity of the light passing through the MC.

The optical microphotographs for the FLC infiltrated MCs, under crossed polarizers, with different exposure energy has been given in Figs. 2(a)–2(e).

The direction of the polarizer and analyzer are as same as shown in Fig. 2(h). As reflected from the Figs. 2(a)–2(e), the alignment quality of the FLC improved at higher exposure doses up to the irradiation energy of 2.84 J/cm^2 and deteriorates with the further increment in the irradiation energy for the SD1 alignment layer. The best alignment has been observed at the irradiation energy of 2.84 J/cm^2, which is very close to the expected irradiation energy mentioned earlier in the manuscript.

Furthermore, the alignment quality of the fiber with irradiation energy 2.84 J/cm^2 [the same capillary shown in Fig. 2(c)] has been illustrated in Figs. 2(f)–2(h). When the capillary was rotated between the crossed polarizers, the changes in light intensity have been observed from red [Figs. 2(f) and 2(h)] to bright yellow. Thus, we have achieved the uniform axial alignment of FLC in the MC.

In a further step we have irradiated MC samples selectively using glass plate with a metallic periodic amplitude mask (with period 2 mm) to create the periodic index distribution of LC in MC. This experiment has been done first, to explore the possibility for the fabrication of the patterned alignment that can be used to fabricate different photonic elements [5–8]. Second, it also confirms the rewritability of the easy axis of the SD1 molecules by further UV irradiation. The advantage of rewritability can be considered especially useful in the context of PCFs infiltrated with LCs. It can be applied for creating and modifying a grating vector of a prefabricated fiber gratings based on a combination of PCFs infiltrated with SmC* LCs. Thus, a prealigned microchannel (without LC) has been covered with a prescribed mask and irradiated with the same UV light source but with a polarization azimuth at 45° to that of previous exposure. Thus, the easy axis of the alignment for the region of the microchannel with open windows realigns in the direction 45° to that of the covered regions. However, the required irradiation energy to get the optimal anchoring energy for the realigned region is higher than that of the first time irradiation. The optimal irradiation energy for the second UV irradiation is 10.47 J/cm^2. The energy absorption by the mask and realigning process can be attributed for this higher energy requirement. It is observed in experiments that 70% of light at the wavelength $\lambda = 350$ nm has been absorbed by the used amplitude mask, and therefore the total energy supplied to the alignment layer is equal to 3.141 J/cm^2, which is relatively higher than the first irradiation.

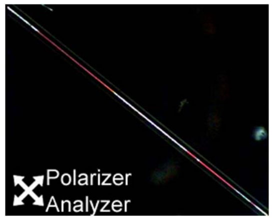

Fig. 3. Pattern alignment in PLCF infiltrated with FLC molecules after double step irradiation process.

The optical microphotograph of the fabricated microchannel with two different periodic alignment domains is shown in Fig. 3.

The photograph was taken by the polarizing microscope with a magnitude of 20× using a high resolution camera. The length of each alignment domain is 1 mm. In the lower right part of the microchannel some defects have been observed, probably due to impurities and dust on the amplitude mask during the UV exposure process. Additionally, the period can be reduced further depending on the divergence of the irradiating light source and diffraction limits [27].

In summary, we have disclosed a method to align FLCs in the microchannels by means of photo-alignment. Photo-alignment based on the sulfonic azo dye SD1 provides good control on the anchoring energy by varying the irradiation energy that delivers optimal value of the anchoring energy and therefore a good FLC alignment has been achieved inside the MC. Moreover, rewritability of the easy axis of the SD1 by another exposure with similar irradiation energy enables us to achieve two multialignment domains. An example of the two-alignment domain with easy axis mutually inclined at 60° is shown in Fig. 3.

Furthermore, the rewritability provides an opportunity to write the alignment of the FLC infiltrated microchannels for different alignments, alignment patterns, or periods. An example of rewriting the easy axis distribution of the FLC infiltrated display cell has recently been disclosed in Ref. [26]. Supplementary irradiation with the UV light, with or without different amplitude or phase pattern, after heating the prealigned FLC filled microchannels beyond the isotropic temperature of the FLC can change the easy axis distribution.

Thus, a good, stable, and rewritable alignment with mono/multiple alignment domains in microchannels opens up a wide variety of interesting physical phenomena and potential applications in modern photonic devices. The future research agenda in the field may include development of the process to achieve the same sort of alignment for PCFs infiltrated with well-aligned FLC that leads to the design of a fast tunable optical element controlled by applying the external electric field. One of the possible applications of periodic alignment of FLC molecules in PLCFs is the fabrication of long-period fiber gratings [8,28].

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