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Fast birefringent mode stressed liquid crystal

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We report a stressed liquid crystal (SLC) that produce a large shift in phase retardation at submillisecond speeds. The SLC consists of uniformly aligned micro-domains of a liquid crystal dispersed in a polymer structure. Mechanical stress produces uniform alignment, essentially eliminates light scattering, and substantially improves the electro-optic performance. A 22-μm-thick SLC film switches more than 2 μm of phase retardation in less than 1 ms. The system has a linear voltage response with essentially no hysteresis. © *2005 American Institute of Physics.* [DOI: 10.1063/1.1852720]

Liquid crystal polymer composites have been extensively studied over the last decade because of their intriguing physics and their potential application in novel, fast-switching liquid crystal devices. Polymer networks created by thermal- or UV-curing of a liquid crystal/monomer mixture strongly affect the electro-optic properties of liquid crystal devices. Depending on the concentration of the monomer in the liquid crystal matrix and the details of the phase separation process, a variety of network structures can be created. At low monomer weight concentration ($<6\% - 8\%$), a low density polymer network is formed. According to Fung *et al.*[1] low-density networks dispersed in a nematic host consist of nano-sized polymer fibrils that form bundles with lateral sizes of hundreds of nanometers. The polymer bundles orient along the liquid crystal director. The director may be controlled by a surface alignment or external fields. The network stabilizes the liquid crystal matrix by locking in the director orientation with the bundles, resulting in decreased response time and increased driving voltage. Escuti *et al.*[2] reported a decrease of the on-times of IPS mode from ~90 ms in pure liquid crystal to ~10 ms in 2% polymer stabilized liquid crystals. An even faster response of the polymer-stabilized liquid crystal composites can be achieved by increasing the polymer concentration. However, light scattered by the polymer network and the liquid crystal micro-domains drastically reduces light transmission through the cell.

Several approaches have been explored to increase the switching speed of liquid crystal-polymer composites while maintaining optimum optical properties. Holographic polymer dispersed liquid crystals (PDLCs), having submicron liquid crystal droplets dispersed in a polymer binder, are among the fastest known liquid crystal devices, providing submillisecond response.[3] Fan *et al.*[4] produced devices operating in the near IR wavelength range using relatively high concentrations of polymer ($c \approx 10\%$) dispersed in a liquid crystal. This system can modulate IR light with about 2 ms switching speeds. High transmission was achieved by uniform surface alignment of the liquid crystal domains, eliminating light scattering. We found that while good performance can be achieved with relatively thin cells, uniform

alignment is difficult to maintain using this surface alignment approach with the thicker films required to produce the large phase retardations reported in this paper.

Doane *et al.*[5] and Aphonin[6] oriented liquid crystal domains by mechanical shearing of a polymer dispersed liquid crystal. This approach produced a composite with a well-oriented, relatively dense polymer network without using any aligning layers.[7] These materials scatter light with a clear polarization dependence. In this letter we report using stressed liquid crystals (SLCs) for ultrafast birefringence switching at relatively low driving voltage and with essentially no light scattering for any polarization of incident light.

The production of a SLC cell consists of two main steps. First, a continuous polymer network is formed. The concentration of the polymer is optimized to be high enough to maintain the mechanical stress induced by shearing (we assume by adhering to the substrate) while being as low as possible to maximize the electro-optic performance (maximum phase retardation and minimal light scattering). Before shearing the SLC cell scatters light because of the randomly aligned liquid crystal domains dispersed in the polymer network. In the second step the cell is sheared by moving one substrate relative to the other. Typically the substrates are moved distances on the order of several times the thickness of the cells. This essentially eliminates light scattering and produces uniform alignment of the liquid crystal domains as is seen from the birefringence produced by the cell. We assume that sheering orders the polymer network, which then orients the liquid crystal domains in the direction of shearing. This dramatically reduces the light scattering and decreases the switching time.

We prepared sets of 22-μm-thick SLC cells using a mixture of a cyanobiphenyl liquid crystal (either 5CB, E7, or E44) and the photocurable monomer NOA65 in a weight ratio of 9 to 1, respectively. Lower concentrations of polymer produced a polymer network that could not sustain the mechanical shear. The cell, made of two indium-tin-oxide covered glass substrates, was filled with the mixture and maintained at a constant temperature while irradiated with unpolarized UV light for about 30 min at ~30 mW/cm^2. After the irradiation, all the cells appear opaque. The amount of light scattering depended on the temperature of polymerization. We found that if a cell is polymerized at a tempera-

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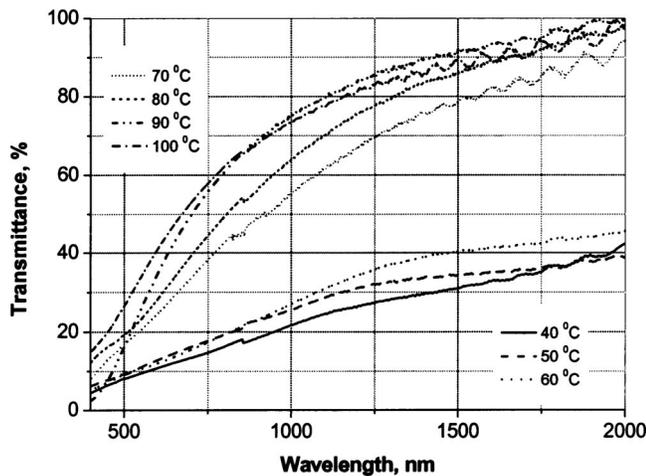[a]Author to whom correspondence should be addressed; electronic mail: johnwest@lci.kent.edu

FIG. 1. The scattering dispersion of the E7-based SLC films as a function of the temperature of polymerization. The $N-I$ transition temperature for the E7 liquid crystal is 67 °C. Those cells prepared at a lower temperature have higher scattering and their characteristics belong to the lower brunch of the transmittance curves. The scattering is much lower for the cells polymerized at a higher temperature than 67 °C. The measurements were done before application of any shearing stress.

ture lower than the $N-I$ transition temperature of the liquid crystal used, the transmittance curve belongs to lower brunch of the curves as shown in Fig. 1. For a cell polymerized at a higher temperature than the $N-I$ transition temperature, the initial scattering is much lower and the transmission curve belongs to the upper brunch of the curves (Fig. 1). Here and later we normalize transmittance measurements on the transmittance of an empty cell.

Initially, all the cells appear optically isotropic when viewed between crossed polarizers. Therefore the unstressed cells cannot be used in a traditional birefringence mode. Also, the unstressed cells are characterized with a slow electro-optical response. For example, for a 5CB based cell polymerized at a temperature of 60 °C application of about 20 V switched it into a transparent state with the relaxation time of about 50 ms. The relaxation time is even larger for the cells polymerized at a lower temperature than 60 °C.

Mechanical shearing drastically changes the optical and electro-optical properties of the mentioned 5CB cell. The cell becomes transparent, highly anisotropic and with greatly increased switching speeds. We quantified the shearing using a specially constructed shearing device where the shearing distance can be controlled with an accuracy of 2 μm. Figure 2 shows the dependence of the film transmittance versus the shearing distance L_{shear}. A dramatic increase of the transparency occurs upon shearing. For instance, the transparency at $\lambda=0.63$ μm increases from 20% for $L_{shear}=0$ to 95% for $L_{shear}=100$ μm. The increase of the transparency occurs because of the uniform orientation of the liquid crystal microdomains along the shearing. Unlike PDLCs,[8] the transmittance of the SLC cell does not depend on the polarization of the probe light indicating that the light scattering resulted from a mismatch between the refractive index of the unaligned liquid crystal domains and not between the individual domains and the polymer network. This indicates that the dimensions of the polymer network are small relative to the wavelength of light.

Starting from a shearing distance of about 20 μm we were able to measure the pretilt angle in the cell. It revealed
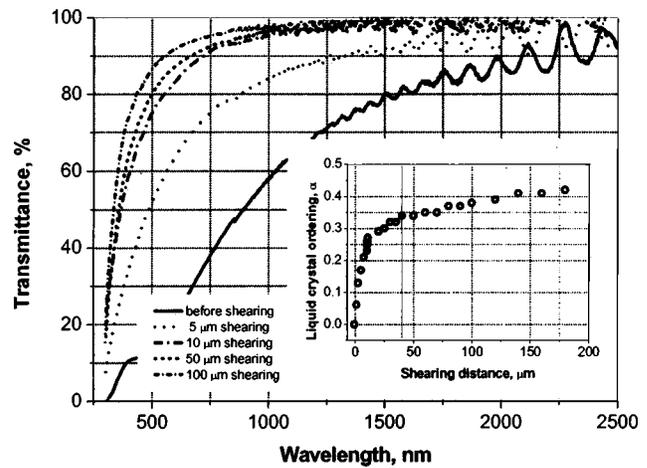

FIG. 2. The dependencies of transmittance of the 5CB-based SLC and the liquid crystal ordering (insertion) on the wavelength of the probe light and different distance of shearing.

a value of about 3°, which changed negligibly with further shearing. To characterize the orientation of the liquid crystal at a smaller shearing we measured the absorption dichroism of the SLC film in the absorption band of 5CB at λ =0.325 μm for different shear distances, L_{shear}. From these data we calculated the dichroic ratio, N, and then the liquid crystal domains ordering as $\alpha=(N-1)/(N+1)$. In our experimental geometry, a change of α with shearing reflects changes of the liquid crystal orientation in both the azimuthal and polar planes. As depicted in the inset of Fig. 2, the ordering of the liquid crystal increases drastically at small shearing ($L_{shear}<20$ μm) and saturates above ∼80 μm.

The switching speed of the liquid crystal cell increased dramatically upon shearing. We measured the switching speed of the cell by placing it between crossed polarizers, and aligning the shearing axis 45° to the polarization axes. The transmitted intensity, I, of the normally incident beam can be calculated by the following equation: $I=I_0 \sin^2 \delta$, where $\delta=\pi\Delta nd/\lambda$ is the angular phase retardation and Δn is a function of the applied voltage. The transmitted intensity will be at a minimum when the phase retardation, Δnd, is an even multiple of the incident light wavelength.
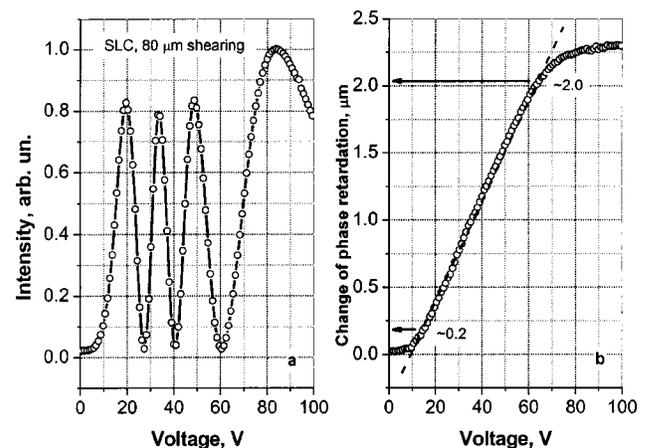

FIG. 3. Static electro-optic response of the SLC cell: (a) the dependence of transmittance of the SLC cell vs applied voltage measured between crossed polarizers; (b) corresponding change of the phase retardation vs applied voltage.
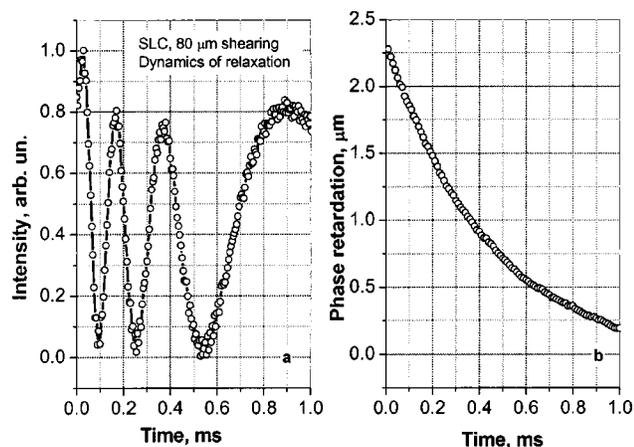
FIG. 4. Dynamic response of the SLC cells: (a) the dependence of transmittance of the SLC cell on time after switching-off the 100 V; (b) relaxation of the corresponding phase retardation change when the applied voltage drops from 100 to 0 V.

As noted earlier, the total phase retardation of the cell can therefore be easily determined from a graph of the transmitted intensity relative to the applied voltage. Figure 3(a) shows the dependence of the transmittance as a function of applied voltage for a SLC cell with $L_{shear}=80$ μm. The calculated dependence of the phase retardation shift versus applied voltage $\Delta nd(U)$ is shown in Fig. 3(b). One can see the phase retardation changes linearly with the applied voltage until $\Delta nd(60V) \sim 2$ μm. The electro-optic response of the SLC cell is much more linear than for cells made with a pure, aligned liquid crystal, greatly simplifying the drive electronics. Also the cell shows essentially no hysteresis in the phase retardation, typically found in PDLC and polymer-stabilized liquid crystal systems. The maximum measured shift in the phase retardation for the 22-μm-thick SLC cell is ≈ 2.3 μm. This value amount to about 60% of the theoretical maximum shift of 3.78 μm (assuming 90% of the liquid crystal and $\Delta n=0.19$, and taking into account the low pretilt angle). We propose that the difference $\Delta nd_{theor} - \Delta nd_{exper}$ is related to the remaining inhomogeneity of the liquid crystal orientation in the SLC, which decreases with increased shearing. This inhomogeneity may be induced by surface alignment at the domain boundaries. Also some of the liquid crystal may remain dissolved in the polymer network and therefore is not aligned by the field.

The relaxation of the cell after removal of 100 V is shown in Fig. 4. The relaxation of about 2 μm of phase retardation occurs within 1 ms [Fig. 4(b)]. For switching between bright and dark states, only a $\lambda/2$-shift is required. At the wavelength of $\lambda=0.632$ μm, the phase shift of 0.316 μm occurs within less than 100 μs. The turn-on time is much less than the turn-off time so the SLC can be driven with the frequency of 10 kHz.

The SLC's electro-optic response can be varied by varying the materials used, their relative concentration, and the preparation conditions. For example, by varying concentration of the components and polymerization temperature, we have produced thin SLC cells (~ 5 μm) for display applications that switch about 0.5 μm within 2 ms with the driving voltage of ~ 5 V. For other applications that require large phase shifts we have made 800-μm-thick SLC cells that produce about 60 μm shift of phase retardation within several milliseconds. As with the thinner cells these thick cells require 1 V/μm to switch.

In conclusion, stressed liquid crystals provide ultrafast response and relatively low driving voltage for devices requiring shifts in the phase retardation. SLC cells are simple to fabricate and reliable, the active area of the film may be relatively large, and the films do not require any liquid crystal orientation layers. The shearing technique can produce uniform alignment in very thick cells (up to 1 mm) allowing very large shifts in the phase retardation. The system has a linear voltage response with essentially no hysteresis.

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