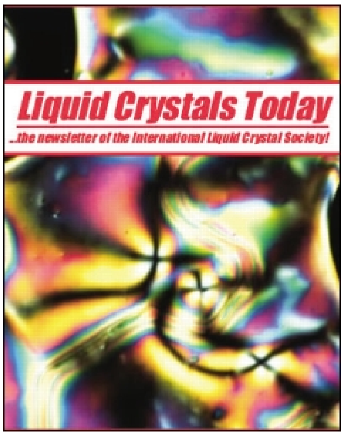

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Electrically Switchable Holograms: Novel PDLC Structures

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Polymer-dispersed liquid crystal (PDLC) films have become the subject of much scientific investigation and commercial development. These systems begin as a homogeneous mixture of liquid crystal (LC) and monomer or oligomer, and possibly some other minor components, and upon curing produce a composite system consisting of a well-defined LC phase embedded in a polymer matrix.

An important class of these systems proceed to their final state by a process known as polymerization-induced phase separation (PIPS). Those which are initiated by photopolymerization also contain a photoinitiator in their starting recipe. When uniformly irradiated, polymerization ensues and LC domains are phase-separated from the growing polymer. These PDLCs are typified by a random distribution of LC domains. The LC droplets are usually of a size comparable to an optical wavelength. They produce random scattering of light, i.e. they are milky white in appearance. The film can be made transparently clear by the application of a suitable voltage. This forms the basis for optical displays (see Paul Drzaic's article in *Liquid Crystals Today*, Vol. 5, 1995).

In contrast to the standard PIPS method for fabricating PDLCs, we have studied a new system which is irradiated holographically, i.e. by a coherent light pattern which is intrinsically anisotropic. This breaks the symmetry of the resultant LC diffusion, nucleation, and subsequent phase separation. The PDLC pattern that is formed results in a coherent scattering of light, which in the limit of a thick film is called the Bragg diffraction regime. In other words, the incident coherent light pattern records a PDLC volume hologram. The size of the LC domains are typically much smaller than an optical wavelength, resulting in optically clear samples. This leads to a new class of PDLC devices: switchable holograms.

Holographic Recording Mechanisms

We use a fast-curing multifunctional acrylate monomer in our prepolymer syrup to record holograms in a single step process. The recipe consists of the monomer, LC, chain-extending monomer, photoinitiator dye, and co-initiator. The homogenized syrup is sheared between glass plates containing nominally 10–20 μm spacers. We record the holograms using standard techniques. Exposures of the order of a few milliwatts per square centimetre for 60–200 s are enough to produce Bragg gratings with a high diffraction efficiency (70–90%).

The photopolymerization reaction proceeds with the formation of an excited triplet-state dye followed by electron transfer from the co-initiator to the dye, thereby producing a free radical. This triggers a rapid free radical polymerization reaction leading to changes in the chemical potential of the system, reducing the miscibility gap between the LC and the monomer. Diffusion of monomers to the polymer-rich regions takes place at the same time as LC molecules diffuse away from high intensity regions. Gelation follows a build-up of the 3-D polymer network, and phase separation of LC microdroplets occurs in low intensity regions. It is possible to control the size and to a large extent the shape of the LC droplets by manipulating the polymerization kinetics.

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Microstructure

The microstructure of a PDLC holographic grating is illustrated in the scanning electron micrograph of Fig. 1. The grating consists of well-defined PDLC channels periodically arrayed between dense polymer regions. These channels provide the refractive index modulation which produces Bragg diffraction.

An intriguing aspect of the anisotropic counter-diffusion of LC and monomer during the hologram recording phase is its apparent control over LC droplet shape. Droplets tend to form in the shape of a prolate spheroid with the long axis preferentially along the grating vector (i.e. a vector perpendicular to the PDLC planes). Internal shear stresses that are applied when the polymer is still soft can distort an otherwise nearly spherical droplet in a direction along the shear. Such internal stresses may be a natural result of the anisotropic diffusion and polymer network growth during grating formation. The net result is a uniaxial domain aligned preferentially along the grating vector.

The optical properties of PDLC holograms depend on the detailed nematic configurations in the small LC droplets. Several configurations have been considered, from bipolar for tangential surface anchoring to the axial configuration when normal surface anchoring is stable. It had previously been thought that, as LC droplets with a homeotropic or radial configuration decrease in size to less than 100 nm, a size dependent symmetry breaking would occur yielding a bipolar director structure. This is because the elastic deformation energy cost becomes too large to maintain the normal surface anchoring. However, other configurations are apparently possible. Recent NMR studies of PDLC gratings indicate a potentially radial-like nematic structure near the surface of an ellipsoidal droplet, with the director bending inward to a central line-segment defect of size comparable to that

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of the droplet. However, the actual nematic configuration in nanometre-sized droplets is still an open scientific question.

Although the LC droplet structure is complex, it is not resolved at optical wavelengths. Hence the distribution of very small LC domains presents an effectively smooth refractive index modulation to incident optical light. The strength of the index modulation and its periodicity, as well as the thickness of the sample, place the resultant diffraction of light in the Bragg regime. Here there is only one diffraction order when the light is incident at the Bragg angle. The diffraction efficiency is very sensitive to this angle, with angular widths ~1°. The amount of random optical scattering is small (<10%) because of the fine LC droplet size.

Interestingly, the Bragg diffraction in these holograms highly favours p-polarized light. This is in stark contrast to ordinary holograms wherein s-polarization is favoured. This confirms the uniaxial nature of the droplets and further supports the hypothesis that the droplet symmetry axes align preferentially along the grating vector.

Electro-Optical Response

The diffraction of light in a simple transmission hologram is illustrated in Fig. 2. Bragg diffraction can produce a large angular displacement of an optical beam (tens of degrees). A diffraction efficiency of 100% is theoretically achievable. Diffraction efficiencies >98% have been obtained experimentally. When a suitable voltage is applied, the effective index of the LC domains matches that of the polymer host. This effectively erases the hologram, and light propagates straight through it like in an ordinary glass window. With the voltage turned off, the hologram is recovered. This forms the basis of a simple optical switch. On/off switching ratios of >25 dB have been measured.

The critical electric field for switching depends on a number of parameters, including droplet shape and size. The addition of surfactants is believed to lower the critical field by lowering the surface anchoring energy of the LC and perhaps by increasing the electrical conductivity of the polymer host. Critical fields of 3–5 V/µm have been thus far achieved. Lower critical fields are believed possible.

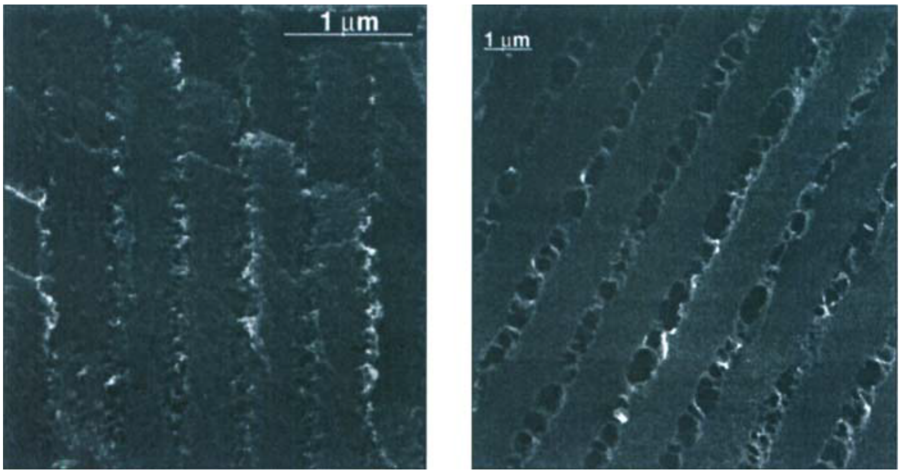

Figure 1. Low voltage, high resolution scanning electron micrograph of a PDLC holographic grating. The sample has been treated to remove the liquid crystal, exposing the voids wherein the liquid crystal resides. The grating consists of periodic PDLC channels interspersed between dense polymer regions. The grating spacing is ~0.5 µm and ~1 µm for the photos on the left and right, respectively. Different reaction kinetics lead to different droplet sizes as shown. (Courtesy of Dr Timothy Bunning, Science Applications International Corporation.)

Another important parameter for devices is switching speed. We have measured response times in the range of 20–40 µs for simple as well as complex PDLC holograms. Response time scales quadratically with droplet size, and the small LC domain size in these PDLC holograms is believed responsible for this fast response in nematic liquid crystals.

Applications

The simple optical switch illustrated in Fig. 2 becomes the basic building block for fabricating more complex programmable beam steering devices. However, this simple switch by itself can perform useful functions. One example is a 2x2 switch for a telecommunications optical fibre network. Current switches are electromechanical and can be prone to failure or high bit error rates in extreme environments such as on-board a naval vessel. An electro-optical switch with no moving parts should prove to be more robust for this type of environment. Fig. 3 shows a prototype of such a switch made with a PDLC hologram for a single mode 1.3 µm fibre. Simple holographic switches can be arrayed to perform as an *N*x*N* programmable switch. These could serve as reprogrammable optical interconnects in the backplane of a parallel processor. Ideas have been formulated regarding the use of PDLC holograms in future optical computing applications.

More complex holograms can be recorded. A projection image hologram recorded in this PDLC material is shown in Fig. 4. The image quality indicates that this material may be useful for the graphic arts. An added feature over other photopolymer holographic materials is that the image in a PDLC hologram can be repeatedly erased and restored electrically, as we have recently demonstrated.

In fact, an attractive feature of this PDLC material is that complex switchable holographic optical elements can be fabricated in a single recording step by simply modifying the recording beams. Several complex structures can be conceived. One example is a switchable holographic lens. In such a lens, a suitable voltage switches the focal length between a few millimetres and infinity. A stack of *N* such lenses in conjunction with a conventional lens yields a digital zoom lens with 2^N focal lengths. Complex diffractive designs can also be used to control the aberrations in a conventional optical system. Such designs are under consideration for use in new 3-D optical data storage applications.

We have also successfully recorded electrically controlled reflection holograms. These types of holograms are useful as notch colour filters. They are also under consideration as the basic elements for full colour reflective displays. It has been predicted that a system based on holographic PDLC technology can achieve >50% luminance in a full-colour system. There is general agreement that this is the requirement for commercial acceptance.

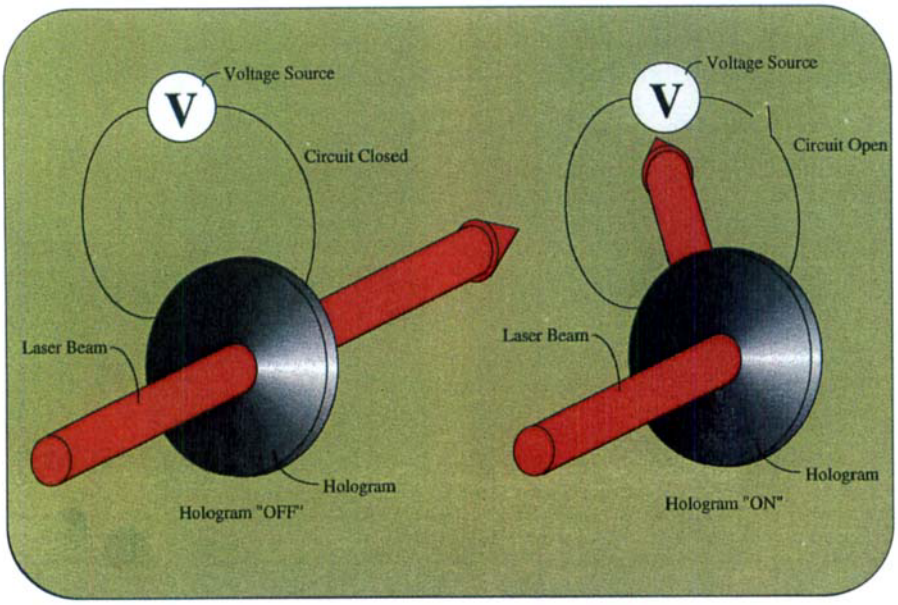

Figure 2. Schematic illustration of diffraction switching in a transmission PDLC hologram. With no voltage applied, the PDLC planes produce a wide deflection of the beam by Bragg diffraction. Upon application of a suitable voltage, the hologram is effectively erased, and the element acts as a simple transparent window. The hologram is recovered when the voltage is switched off.

Future Developments

The impact of this materials technology is just beginning to be felt in certain niche applications. Future research will focus on improvements of system parameters as well as new devices. We expect soon to increase the on/off dynamic range to >30 dB, and ultimately to 40 dB. The switching voltage is also expected to drop, and present goals call for values below 1 V/µm. Switching speed will be addressed with response times approaching 1 µs. Finally, since many applications are in the telecommunications area, the spectral sensitivity for recording is being pushed to cover wavelengths in the near-infrared region. Additional application areas under exploration include switchable filters, imaging spectrometers, optical processing, phased array radar, and automotive lighting.

One of the exciting aspects of this research is that it has trapped the liquid crystal in a quasi-solid-state polymer diffractive device. This is done automatically in the single step fabrication process and hence simplifies the handling of liquid crystals. They become in essence a part of the solid polymer. This makes devices attractive for many applications in the visible as well as the near- and mid-infrared spectral regions. The added dimension of electrically controlled diffraction efficiency opens up a wide range of new designs for diffractive optics. Given the current interest and activity in several applications, more are expected in the future.

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***Richard Sutherland** and **Lalgudi Natarajan** are senior scientists at Science Applications International Corporation, where they are involved in the research and development of nonlinear materials for optical limiting, tunable filters, and other applications. Dr Sutherland is the author of a book,* Handbook of Nonlinear Optics, *recently published by Marcel Dekker. Both authors have published extensively on PDLC holograms, including a co-authored chapter in* Polymeric Materials Encyclopedia, *published by CRC Press.*

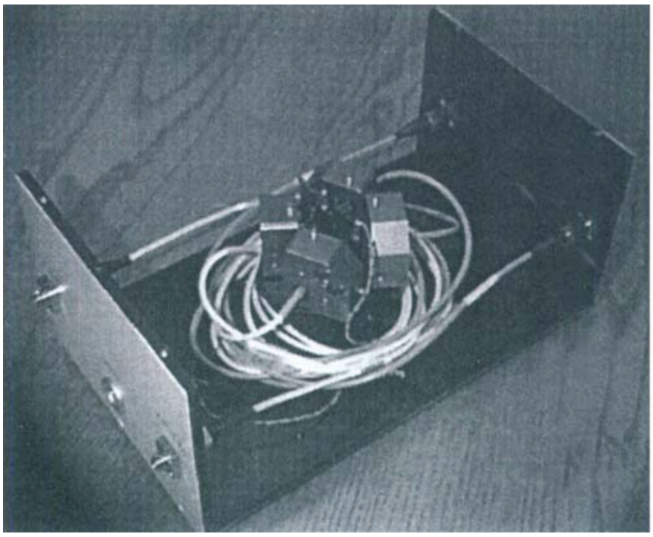

Figure 3. Photograph of a prototype 2x2 single mode 1.3 µm fibre optic switch based on an electrically switchable PDLC transmission hologram. (Photograph courtesy of Dr Lawrence Domash, Foster-Miller, Inc.)

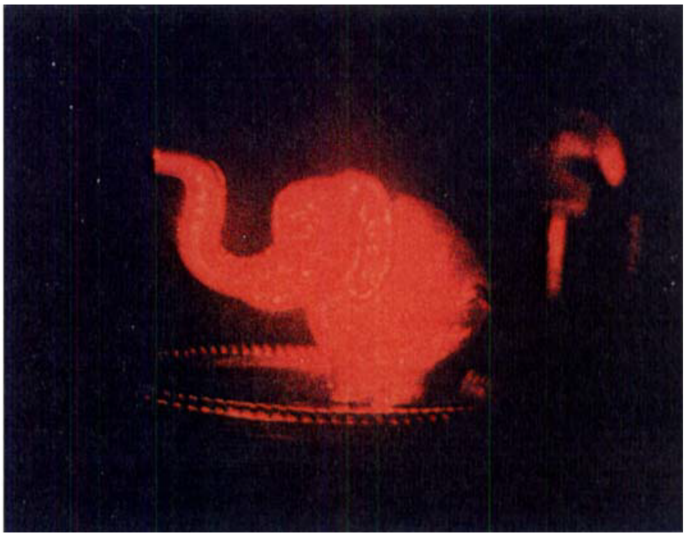

Figure 4. Image of a projection hologram formed in a PDLC material. The image can be repeatedly erased and restored by the application of an electric field. (Courtesy of Mr Vince Tondiglia, Science Applications International Corporation.)