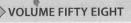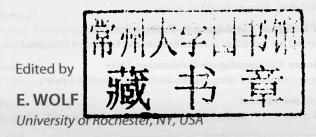# EMIL WOLF

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PREFACE

The present volume contains review articles on the following subjects:
Low-dimensional silicon structures for use in photonic circuits, phase
anomalies in microoptics, invisibility physics, dynamic photonic materials
based on liquid crystals, and subwavelength atom localization. Experimental,
as well as, theoretical researches are reviewed.

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March 2013

CONTENTS

CHAPTER ONE

Dynamic Photonic Materials Based on Liquid Crystals

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Contents

1. INTRODUCTION

Optics and photonics involve material sciences and device technology, at the basis of displays, computing devices, optical fibers, precision manufacturing, enhanced defense capabilities, and a plethora of medical diagnostics tools. Opportunities arising from optics and photonics offer the potential for an even greater social impact in the next few decades, related to solar power generation, efficient lighting, and faster internet. Continuously increasing data capacity requirements in telecommunications and in the next-generation of dynamically reconfigurable networks increases demand for highly compact, non-mechanical, and high speed optical devices. New materials exhibiting enhanced optical properties are key to these developments. In particular, liquid crystals (LCs) have attracted a great deal of attention in the last three decades. This is due to their capability both to behave as smart anisotropic materials, exhibiting self-organizing properties along with fluidity, and to fulfill conditions imposed from outside, due to their responsiveness to a wide variety of external perturbations, like AC, DC, and optical fields (Gennes & Prost, 1995). Indeed, the large birefringence ($\sim$0.5) of LCs allows for the realization of tunable photonic devices for both optical communications and optical sensing systems. LCs and polymers have become an exciting field of research with practical applications in flat panel displays and active optical devices. Thanks to achievements obtained in the micro/nano fabrication processes, such as Intensity and Polarization Holography, Electron Beam (E-Beam) Lithography, Focused Ion Beam (FIB), and Dip-Pen nanolithography, several composite photonic structures exploiting LCs properties have been realized. Liquid Crystals are currently playing a significant role in nanoscience and nanotechnology, too. They can be utilized as bridge between "hard matter" and "soft matter," due to the fact that nano-structured materials do not induce significant distortions of LC phases. Various nanomaterials have been dispersed and studied in LCs to enhance their physical properties. Furthermore, alignment and self-assembly of nanoparticles themselves can be achieved inside the LC, since it acts as a tunable solvent for the dispersion of nanomaterials. As an anisotropic medium, it provides a support for the self-assembly of those materials into large organized structures, even

in multiple dimensions. Most importantly, in order to exploit the distinctive characteristics and capabilities of LC technologies, a variety of means for alignment and confinement of LCs have been investigated and employed. In this chapter, we review our achievements in the fabrication and characterization of LC-based photonic devices, underlining, in particular, the "active way" we utilize to control their properties. The most important aspects and novelties are highlighted in different sections of the paper. We begin with an overview of electro-responsive and light sensitive chiral nematic LCs. We continue by reporting on the optical and electro-optical properties of holographic structures containing several kinds of LC phases and, finally, we show how it is possible to exploit and control the plasmonic nanomaterial properties by means of LCs utilized as active host media.

2. PHOTONIC DEVICES BASED ON CHOLESTERIC LIQUID CRYSTALS

2.1 Electro-Responsive CLCs

In Cholesteric Liquid Crystals (CLCs), also called chiral nematic LCs, the molecules are arranged in a helical structure such that in each plane of the system the directors are aligned (and lay in that plane) and the director orientation changes progressively along the direction perpendicular to the planes (such direction, **h**, constitutes the axis of the helix). If the helix axis is along **z** and **n** is the director orientation, the angle θ between **n** and a reference direction in the xy plane can be expressed as follows:

$$\theta = (2\pi/P)z, \tag{1.1}$$

where the parameter P is the pitch of the helix, that is the distance along **h** over which the orientation of the molecules rotates by 2π. In each xy plane (constant z) the system only has orientational order but no translational order. Because of the periodicity in the director orientation in **z**, CLCs behave as one-dimensional photonic bandgap system and propagation of light of certain wavelengths and polarizations states is forbidden (Blinov, 1983; Yeh & Gu, 1999). In particular, for a CLC system in a planar state (**h** ∥ **z**, perpendicular to the plane of the cell, xy) and at normal incidence (**k** ∥ $\mathbf{e}_z$, where **k** is the propagation wavevector of the light beam), circularly polarized light of wavelength between $n_o P$ and $n_e P$ (n_o and n_e are the ordinary and extraordinary refractive indices of the material, respectively) with the same handedness as the helix is reflected by the CLC layer, while the opposite sense of circular polarization propagates through the CLC
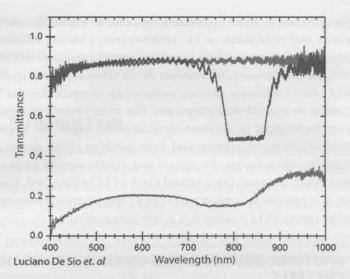
Figure 1 Transmission spectrum of a CLC cell (80% E7, 20% R811) at normal incidence: sample in the planar homogeneous state with $\lambda_0 = 825$ nm, no field applied (blue line); sample in a focal conic state at $E = 1.5$ V/μm (green line); sample in the homeotropic state at $E = 5$ V/μm (red line). All field square waves at 1 kHz. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this book.)

unaffected. For unpolarized light, an ideal sample reflects 50% of the light and transmits the remaining 50% in the wavelength range $n_oP < \lambda < n_eP$, whereas the sample is transparent outside this range (Figure 1, blue line). The center of the reflection band occurs at:

$$\lambda_0 = \langle n \rangle P, \tag{1.2}$$

where $\langle n \rangle$ is the average refractive index, $\langle n \rangle = (n_e + n_o)/2$ and the bandwidth is given by:

$$\Delta\lambda = \Delta nP = (n_e - n_o)P. \tag{1.3}$$

Because of the dielectric anisotropy of the material, the director orientation of the liquid crystals can change in the presence of an electric field, and this effect has been used to design various types of electro-responsive devices based on CLCs. For a CLC with a positive dielectric anisotropy ($\Delta\varepsilon > 0$), the helical structure is not stable when an electric field is applied parallel to the helical axis ($\mathbf{E} \parallel \mathbf{h}$) (Blinov, 1983). When an electric field is first applied to a planar aligned (homogeneous) CLC cell, the sample becomes scattering at a field above a critical value. The axis of the helix becomes tilted (from

the original normal condition) and the sample assumes a multi-domain focal conic texture (Figure 1, green line), which does not transmit light. When the field is further increased above a second critical value, the helical arrangement of the molecules is destroyed and molecules (and overall director) become aligned parallel to the field direction (perpendicular to the cell). The sample becomes transparent (Figure 1, red line) and the large change from a reflective to transparent state by field is the basis for numerous display-related applications. However, upon removal of the field, recovery of the initial reflective state can be very slow and in many cases the sample remains trapped in the disordered focal conic texture (scattering state), limiting the utility of the system for practical applications. Two major approaches have been explored to overcome this shortcoming. The first, the use of dual frequency LCs, utilizes an applied field to drive alignment in both directions enabling fast switching between the transparent and reflective conditions (Xu & Yang, 1997). In these materials, the sign of the dielectric anisotropy depends on the electric field frequency (Bücher, Klingbiel, & VanMeter, 1974; De Jeu, Gerritsma, Van Zanten, & Goossens, 1972). Below a crossover frequency, $\Delta\varepsilon > 0$, and a homogeneous cell can be switched from the planar (reflective) to the homeotropic (transparent) state. For frequencies above the crossover point, $1\varepsilon < 0$, an applied field will reorient the LC molecules from a homeotropic (transparent) orientation back to parallel (to the plane of the cell), facilitating the restoration of the standing helical structure. Thus, toggling between frequencies, with an applied field in both cases, will enable switching between the reflective and transparent states. Switch-off times on the order of 100 ms have been achieved using this type of material (Xu & Yang, 1997). Dual frequency CLCs have also been used to fabricate fast-switchable devices that operate between the reflective and scattering states (Gerber, 1984; Hsiao, Tang, & Lee, 2011). The shortfalls to this approach are specialty materials whose electro-optical properties are very sensitive to temperature and complex drive schemes. The second approach for increasing speed has been incorporating a loose polymer network within the CLC cells. Shortening of the recovery time by several orders of magnitude has been demonstrated (Beckel, Natarajan, Tondiglia, Sutherland, & Bunning, 2007; Hikmet, 1998; Sathaye, Dupont, & de Bougrenet de la Tocnaye, 2012). Typically, the cell is fabricated with a mixture of photopolymerizable monomers which are then polymerized using light after initial fabrication of a specific cell design (typically planar homogeneous). These systems are generally referred to as polymer-stabilized CLCs (PSCLCs). Figure 2 shows the transmission spectrum as a function of applied voltage for a PSCLC reported by Hikmet
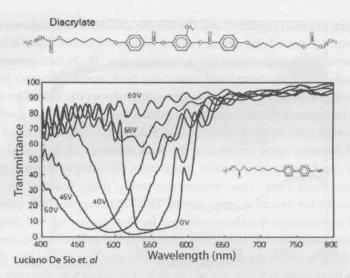
Figure 2 Transmission spectra as a function of applied voltage for a PSCLC system containing the monoacrylate monomer shown in the inset and a small amount of diacrylate shown above the spectra. *Reproduced from Hikmet and Kemperman (1998).*

and Kemperman (1998), which was fabricated using UV irradiation. The dip in the transmission spectrum (corresponding to the reflection band of the CLC system) initially shifts to shorter wavelength when a voltage of 40–50 V is applied (the blue-shift is due to tilting of the helix axis), but the system is still highly transparent in the rest of the spectrum. At 60 V, the sample is brought into a homeotropic state and becomes transparent. Because of the presence of the polymer network, the planar homogeneous configuration can be fully recovered upon removal of voltage with switching-off times as short as a few ms (Beckel et al., 2007; Hikmet & Kemperman, 1998). The recovery time depends on the pitch and elastic constant of the liquid crystal as well as on the content of diacrylate monomer in the initial mixture. Quicker response times are typically observed for larger contents of diacrylate monomers and thus higher crosslinking density (Beckel et al., 2007; Guillard, Sixou, Reboul, & Perichaud, 2001; Hikmet & Kemperman, 1999). The polymer network, however, cannot be too dense and rigid, otherwise the strength of the interaction between the free liquid crystal molecules and the polymer network overcomes that of the field–induced reorientation and the system is no longer switchable. One drawback is that the switch-on time and voltage for the transition from the reflective to the transparent regime are typically larger in PSCLCs than in CLCs without polymer network (Hikmet & Kemperman,

1999). Polymer stabilization has also been used to achieve broader reflection bands (Broer, Lub, & Mol, 1995; Broer, Mol, van Haaren, & Lub, 1999; Hikmet & Kemperman, 1998) and to reduce the temperature dependence of the pitch. It has also been utilized to fabricate reflective reverse-mode systems which offer a much higher contrast ratio and lower operating voltages than the typical transmissive systems (Ren & Wu, 2002).

In the cases discussed above, the pitch length and thus the position of the reflection band was determined by the composition of the CLC system and was not significantly affected by the field when $E \parallel h$ (apart from a small blue-shift in the band position that can be seen in some cases at low field and corresponds to a tilt of h, but not changes in the pitch itself). A wider range of applications would be accessible if the reflection band could be tuned rather than just switched reversibly in different regions of the spectrum. Phototunable changes in the helical twist power and birefringence are discussed elsewhere in this article. A few electro-responsive tuning architectures have been explored and are discussed below. The development of full color (e.g., red, green, and blue), addressable CLC reflectors to decrease the complexity of stacked red–green–blue CLC pixels for full color displays has been the major driver behind exploration of tunable CLC's. If an electric field is applied perpendicular to the helical axis ($E \perp h$) of a CLC with a positive dielectric anisotropy ($\Delta\varepsilon > 0$), the helical structure is deformed and the director of the liquid crystal molecules undergoes a partial reorientation toward the field direction, resulting in a elongation of the pitch [the dependence of θ on z is no longer described by Equation (1.1)] (Blinov, 1983; Meyer, 1968). If the CLC is in a planar homogeneous state, at normal incidence, this results in a shift of the reflection band toward longer wavelengths (Figure 3) (Li, Desai, Akins, Ventouris, & Voloschenko, 2002; Xianyu, Faris, & Crawford, 2004). In this device configuration, an electric field with the appropriate orientation (in-plane field) is often achieved using a pattern of interdigitated electrodes on one of the cell substrates. The red-shift in band position increases with an increase in field strength until the limiting value in field strength for which the helix is completely unwound (infinite pitch, homogenous alignment with $n \parallel E$), the reflection band disappears, and the sample becomes transparent under unpolarized light (the sample is a uniaxial birefringent slab). Shifts on the order of 300 nm for systems with reflection band in the visible have been achieved based on this approach (Li et al., 2002). Relatively fast response times (on the order of ms) have been reported for this process (Li et al., 2002), even without polymer stabilization. However, depending on the target application, the use of interdigitated electrodes may pose