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Photonics and lasing in liquid crystal materials

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Owing to fundamental reasons of symmetry, liquid crystals are soft materials. This softness allows long length-scales, large susceptibilities and the existence of modulated phases, which respond readily to external fields. Liquid crystals with such phases are tunable, self-assembled, photonic band gap materials; they offer exciting opportunities both in basic science and in technology. Since the density of photon states is suppressed in the stop band and is enhanced at the band edges, these materials may be used as switchable filters or as mirrorless lasers. Disordered periodic liquid crystal structures can show random lasing. We highlight recent advances in this rapidly growing area, and discuss future prospects in emerging liquid crystal materials. Liquid crystal elastomers and orientationally ordered nanoparticle assemblies are of particular interest.

Keywords: liquid crystals; mirrorless lasing; photonic band gap

1. Introduction

Liquid crystals, constituting a new phase of matter, were discovered in 1888. They remained a scientific curiosity until the late 1960s, when liquid crystal displays (LCDs) were invented. Today, liquid crystals are the technology of choice for the multibillion dollar flat panel display industry. The development of LCDs has fuelled much liquid crystal research in the past 40 years. The subsequent maturing of the LCD technology, however, resulted in increased exploration in new directions in the liquid crystal research. This in turn gave rise to beautiful results and unexpected advances in a wide variety of areas, to the extent of suggesting a renaissance in the field. Photonics and lasing in liquid crystal materials represents one such advance.

2. Photonics and photonic band gap materials

Photonics is the science and technology of controlling photons, in the way that electronics is the science and technology of controlling electrons. Although the flow of electrons can be controlled in a variety of ways, such as via thermionic valves, it was semiconductors and transistors that really opened the door to the

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realization of current state-of-the-art electronic devices. Photonic band gap (PBG) materials, with the potential of allowing comparable control over light, are the subject of intense interest today.

Periodic PBG structures were invented independently by Yablonovich (1987), whose goal was to prevent photon emission, and John (1987), who focused on photon localization. PBG materials are characterized by the property that classical light propagation in them is forbidden for some range of frequencies; this region is the PBG. Although normal metals, where the dielectric permittivity is negative below the plasma frequency and light propagation is therefore forbidden, are PBG materials, interest today is focused primarily on periodic dielectric structures. The origins of the band gap in periodic structures are deeply embedded in mathematics. According to Floquet's theorem (Floquet 1880), solutions of second order ordinary differential equations with periodic coefficients are of the form

$$\sim e^{i\eta z}\phi(z), \tag{2.1}$$

where ϕ is a periodic function of position z. If η is real, the solutions are stable and periodic, but in the regions of parameter space, where η is imaginary, no stable solutions exist. This region corresponds to the band gap. A simplified example is the Schrödinger equation for electrons in crystals,

$$\nabla^2\psi = -(\mathsf{E} - V)\psi, \tag{2.2}$$

where ψ is the wave function and V is the periodic crystal potential. The periodicity of the potential gives rise to the band gap between the valence and the conduction bands in semiconductors. All the semiconductor electronics exploit this feature. Another example is the wave equation describing light propagation in dielectric structures,

$$\nabla^2 \boldsymbol{E} = -\omega^2\mu\varepsilon\boldsymbol{E}. \tag{2.3}$$

Here, the electric field $\boldsymbol{E}$ plays the role of wave function for light, ω is the frequency, μ, the magnetic permeability and ε, the dielectric permittivity. If ε is spatially periodic, then light propagation is forbidden for some range of frequencies; this is the PBG.

Liquid crystals can spontaneously form periodic dielectric structures. They therefore have the potential to be self-assembled PBG materials.

3. Liquid crystals

In 1888, the Austrian botanist, Reinitzer observed two apparent melting points in cholesteryl benzoate. At temperatures between these two melting points, the material is a liquid with the optical properties of birefringent crystals. Reinitzer was subsequently credited with the discovery of liquid crystals. The traditional definition of liquid crystals is that they are liquids with some of the optical properties of solid crystals.

Solid crystals are characterized by the positional order of their constituent atoms or molecules, while the optical properties of liquid crystals are associated with the orientational order of their constituents. Both positional and orientational orders represent broken symmetry. In the case of solid crystals,

Table 1. Order and broken symmetry.

	positional order	orientational order	broken symmetry
solid crystal	yes	yes	discrete
plastic crystal	yes	no	discrete
liquid crystal	no	yes	continuous
isotropic fluid	no	no	none

the broken translational symmetry is discrete, while in the case of liquid crystals, the broken rotational symmetry is continuous. It is useful to consider both types of order in condensed matter, as illustrated in table 1.

This classification is of particular interest in light of Goldstone's theorem, which predicts the existence of gapless bosons, or Goldstone modes, in systems with broken continuous symmetry. Goldstone modes are low-energy excitations, with energy $\mathsf{E} \sim q^2$, where q is the wavenumber.

The symmetry-based definition of liquid crystals is that they are systems that possess orientational order, but not (complete) positional order. Consequences arising from such order are the anisotropy of physical properties and the presence of Goldstone modes. Anisotropy allows the coupling of external fields and the orientation of the constituent elements of the liquid crystal; Goldstone modes give rise to long length-scales and the possibility of periodic structures with periods comparable to the wavelength of light. They also make these materials 'soft', i.e. exceptionally responsive to external excitations.

The symmetry-based definition broadens the class of liquid crystals significantly. It is now recognized that many materials, in addition to the traditional low molecular weight liquid crystals and liquid crystal polymers, are liquid crystalline. The examples are chromonic liquid crystals, whose constituents are molecular aggregates found in many dyes and drugs, viruses and other biological materials, nanoparticle assemblies and mineral slurries and solid liquid crystal elastomers and gels. The emerging view today is that liquid crystals are an extremely broad class of materials, whose diverse members—some of which are only now being recognized as liquid crystals—are united by their symmetry-based softness and responsiveness.

For photonics applications, the relevant aspect is a periodic dielectric structure. An example of this is found in cholesteric liquid crystals. The simplest liquid crystal phase is the uniaxial nematic phase, where the symmetry axes of neighbouring constituents point, on the average, in the same direction. This direction is the nematic director, identified by the unit vector $\hat{\boldsymbol{n}}$. In the groundstate, the director is uniform in space. The energy cost of changing the direction of $\hat{\boldsymbol{n}}$ on long length-scales, however, is small; the energy density associated with twisting the director, so that its tip traces out a helix with period, p is

$$\varepsilon = \frac{1}{2} K_2 q^2, \tag{3.1}$$

where $q = 2\pi/p$, $K_2 \approx kT/l_0$ is an elastic constant and l_0, the length of the constituent molecule (deGennes 1972). The addition of chiral dopants or the chirality of the constituent nematic molecules themselves, can therefore readily induce a twisted helical structure and form the helical cholesteric phase, as shown in figure 1.

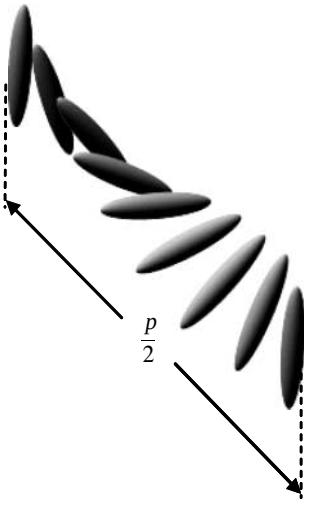

Figure 1. Structure of the helical cholesteric phase.

The connection between the periodic structure and the PBG follows from the wave equation in equation (2.3). Writing the electric field as a sum of monochromatic plane waves, and expanding the dielectric tensor in a Fourier series gives

$$n_j^2 \boldsymbol{E}_j \mathrm{e}^{i\boldsymbol{k}_j \cdot \boldsymbol{r}} = \sum \varepsilon_l \mathrm{e}^{i\boldsymbol{G}_l \cdot \boldsymbol{r}} \boldsymbol{E}_\mathrm{m} \mathrm{e}^{i\boldsymbol{k}_\mathrm{m} \cdot \boldsymbol{r}}, \tag{3.2}$$

where $k=2\pi n/\lambda_0$, n is the refractive index, λ_0 is the free space wavelength and $\boldsymbol{G}$ is a reciprocal lattice vector. It follows that $\boldsymbol{k}_\mathrm{j}=\boldsymbol{G}_\mathrm{l}+\boldsymbol{k}_\mathrm{m}$, i.e. that momentum is conserved. If the eigenvalue n^2 is negative, the index is imaginary. The corresponding mode is evanescent, there is total reflection and thus no propagation of light. A negative eigenvalue, $n^2<0$, is therefore a signature of the band gap.

4. Lasing

A simple laser consists of two mirrors, forming a Fabry–Perot cavity, typically enclosing an optically pumped active medium, which provides gain. A plane wave, with complex electric field amplitude, E_0, propagating in the active medium, will gain phase, will be both absorbed and amplified, and reflected by the mirror. After completing a similar return path, in the steady state, the amplitude must again be E_0, in other words

$$E_0 = E_0 \mathrm{e}^{(ik-\alpha+\gamma)L} r \mathrm{e}^{(ik-\alpha+\gamma)L} r, \tag{4.1}$$

where k is the wavenumber, α is the absorption coefficient, γ is the gain coefficient, L is the cavity length and r is the reflection coefficient of the mirrors. This gives the conditions for the cavity modes

$$kL = m\pi, \tag{4.2}$$

and for the threshold gain

$$\gamma_{\rm th} = \alpha - \frac{\ln r^2}{2L}. \tag{4.3}$$

The lasing threshold is therefore determined by the requirement that gain must be sufficient to overcome absorption and cavity losses.

An alternate approach to note is that, according to Fermi's golden rule, the emission rate Γ of an excited atom is given by

$$\Gamma = \Gamma_0 \rho(\omega), \tag{4.4}$$

where Γ_0 is a bare emission rate and $\rho(\omega) = {\rm d}k/{\rm d}\omega$, the density of states (DOS). For a given structure, the effective wavenumber k can be determined from the phase shift of transmitted light. For a Fabry–Perot cavity, this gives for the DOS

$$\rho = \frac{{\rm d}k}{{\rm d}\omega} = \frac{n}{c}\,\frac{1-r^4}{1-r^2\cos(2kL)+r^4}. \tag{4.5}$$

The maxima occur when $kL = m\pi$, with

$$\rho_{\rm max} = \frac{n}{c}\,\frac{1+r^2}{1-r^2} \simeq \frac{4\pi}{\omega L}\,Q, \tag{4.6}$$

where Q is the quality factor of the cavity. Thus, the DOS also provides a measure of photon confinement. Finally, we note that equation (4.2) for the threshold gain can be written in terms of $\rho_{\rm max}$ and for $r \approx 1$,

$$\gamma_{\rm th} \simeq \alpha + \frac{2n}{c\rho_{\rm max}}. \tag{4.7}$$

Since lasing first occurs at the wavelength where the threshold gain, $\gamma_{\rm th}$, is a minimum, the DOS determines both the lasing wavelength and the lasing threshold.

5. Mirrorless lasing in liquid crystals

In 1971, Kogelnik & Shank (1971) proposed that an external cavity was not required for lasing and that Bragg reflections in periodic structures could play a role similar to external mirrors. Such distributed feedback lasing was demonstrated, and it forms the basis of mirrorless lasing in liquid crystals today. Citing the work of Kogelnik & Shank, in 1973, Schnur & Goldberg proposed and obtained a US patent on a *tunable internal-feedback liquid crystal-dye laser* (Goldberg & Schnur 1973). However, there is no corresponding demonstration or evidence of lasing in the literature. Their basic idea is to use a fluorescent dye dissolved in the liquid crystal to provide gain, and to use the periodic structure of helical cholesterics to provide distributed feedback. To demonstrate lasing, in addition to light emission, line narrowing, directional emission, excited state lifetime reduction, threshold behaviour and coherence must also be demonstrated. Ilchishin *et al.* (1981) have demonstrated modification of the fluorescent emission, but the first unequivocal demonstration of lasing in polymer-stabilized cholesteric liquid crystals was by the group of Genack (Kopp *et al.* 1998) and, shortly thereafter, independently in pure cholesterics by us (Taheri *et al.* 1999).

The optics of cholesteric liquid crystals has been studied extensively (Belyakov 1992; deVries 1951). Light propagation in helical cholesterics is governed by the spatially periodic dielectric tensor, whose eigenvalues are $\varepsilon_{\parallel}$ and $\varepsilon_{\perp}$ for the electric field parallel and perpendicular to the director $\hat{\boldsymbol{n}}$. Useful quantities are the average dielectric constant

$$\bar{\varepsilon} = \frac{1}{2\varepsilon_0}(\varepsilon_{\parallel} + \varepsilon_{\perp}), \tag{5.1}$$

and the dielectric anisotropy

$$\delta = \frac{1}{2\varepsilon_0}(\varepsilon_{\parallel} - \varepsilon_{\perp}). \tag{5.2}$$

Solving Maxwell's equations in a helical cholesteric structure gives at once the secular equation for the eigenvalues

$$n^2 = \bar{\varepsilon} + \alpha^2 \pm \sqrt{\delta^2 + 4\bar{\varepsilon}\alpha^2}, \tag{5.3}$$

where $\alpha = \lambda_0/p$, and the optical eigenmodes

$$\boldsymbol{E} = E_0\left\{(\hat{\boldsymbol{x}} + i\hat{\boldsymbol{y}})\mathrm{e}^{i(((2\pi/\lambda_0)n-(2\pi/p))z-\omega t)} + \frac{\delta}{(n+\alpha)^2-\bar{\varepsilon}}(\hat{\boldsymbol{x}} - i\hat{\boldsymbol{y}})\mathrm{e}^{i(((2\pi/\lambda_0)n+(2\pi/p))z-\omega t)}\right\}, \tag{5.4}$$

where $\hat{\boldsymbol{x}}$ and $\hat{\boldsymbol{y}}$ are unit vectors in the x–y plane (plane of the director), and light propagates along the helix axis in the z-direction. The eigenvalue, n^2, in equation (5.2) has two branches, the branch corresponding to the negative radical is negative for $\sqrt{\varepsilon_{\parallel}} < \alpha < \sqrt{\varepsilon_{\perp}}$ and in this region, n is imaginary. This region therefore corresponds to the PBG, with the low-energy band edges at $\lambda_0 = pn_{\parallel} = p\sqrt{\varepsilon_{\parallel}}$ and the high-energy edge at $\lambda_0 = pn_{\perp} = p\sqrt{\varepsilon_{\perp}}$. Outside the band, the eigenmodes are combinations of left- and right-circularly polarized waves, propagating with different velocities. Inside the band, the mode corresponding to imaginary n is evanescent, with counter-propagating circularly polarized waves. At the band edges, $n=0$ and the modes are standing waves, with $\boldsymbol{E}$ parallel to the director at the low-energy edge, and perpendicular at the high-energy edge.

Ignoring finite size effects, the DOS is

$$\rho = \frac{\mathrm{d}k}{\mathrm{d}\omega} = \frac{\mathrm{d}}{\mathrm{d}\omega}\frac{2\pi n}{\lambda_0} = \frac{n}{c} - \frac{\lambda_0}{2nc}\frac{\mathrm{d}n^2}{\mathrm{d}\lambda_0}. \tag{5.5}$$

Since n^2 is a smoothly varying function of λ_0, as shown in figure 2, the DOS diverges at the band edges as $1/n$, where $n \to 0$. Photon emission by excited atoms and molecules and distributed feedback lasing is therefore expected at the band edges.

Light emission from samples in the helical cholesteric phase doped with laser dyes has been studied extensively. Figure 3 shows a typical emission spectrum (Palffy-Muhoray *et al.* 2000) from a blend of the commercial cholesteric mixture BLO61 and the nematic mixture E7, doped with 2 wt% of 4-dicyanomethylene-2-methyl-6-p-dimethylaminostyryl-4H-pyran (DCM) dye. The sample was 25 μm thick, between parallel glass plates, with the helix axis normal to the plane of the glass. The sample was pumped by 33 ps pulses at $\lambda = 355$ nm from a frequency tripled Nd: YAG laser. Since the sample thickness is finite, the DOS is large, but

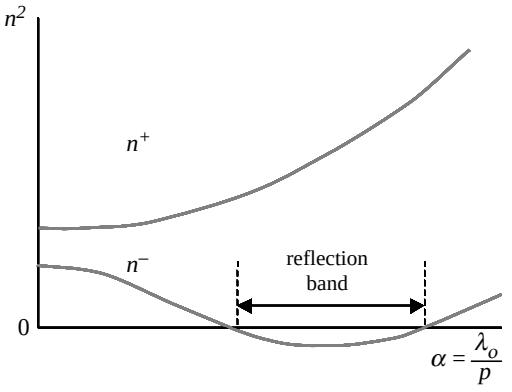

Figure 2. Eigenvalue n^2 versus wavelength/pitch.

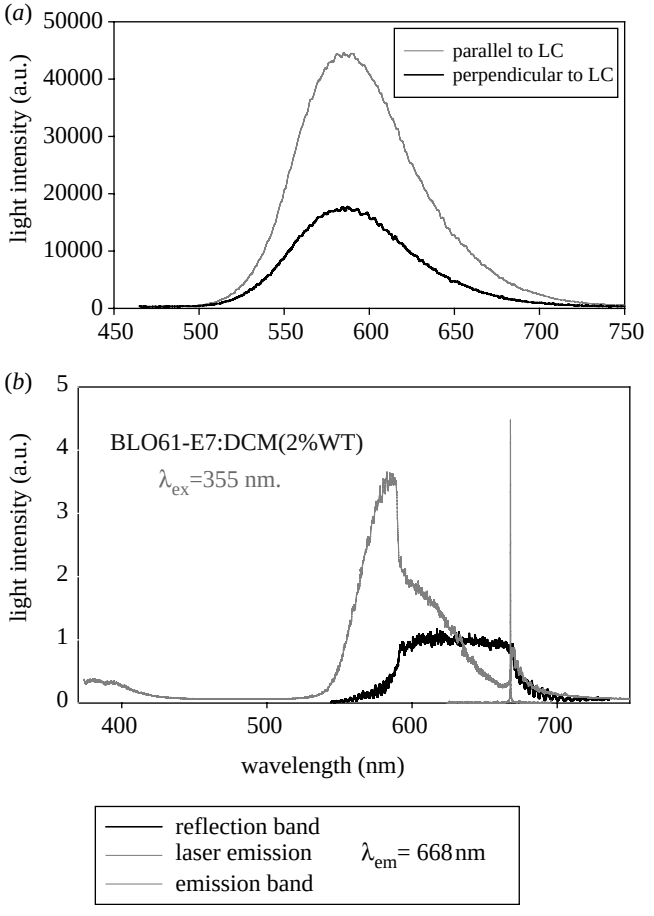

Figure 3. (*a*) Fluorescence spectrum of DCM dissolved in the nematic host E7. (*b*) Reflection band and modified fluorescence and laser emission from the dye doped BLO61-E7 mixture.

not infinite, at the band edges and is small, but not zero, inside the band for the evanescent mode (Cao 2005; Schmidtke & Stille 2003*a*). As shown in figure 3, below the lasing threshold, fluorescence from the dye is suppressed inside the band, and is enhanced at the band edges. Above the threshold, distributed feedback lasing occurs at the band edge. Since DCM aligns with the director, the transition moment of the dye is parallel with the electric field; the gain is consequently greater than at the high-energy edge, hence lasing first occurs here.

The laser emission is directional, coherent, typical line widths, full width at half maximum (FWHM), are approximately 2 Å, with a pump thresholds of approximately 10 nJ and light-to-light efficiency of approximately 20%. Mirrorless lasing has been demonstrated using two dyes with Förster transfer (Alvarez *et al.* 2001; Serak *et al.* 2002), as well as with no dye, where the liquid crystal itself is the active medium (Munoz *et al.* 2001). Lasing has been observed in free-standing helical cholesteric polymer films (Schmidtke *et al.* 2002; Shibaev *et al.* 2002). Finkelmann synthesized cholesteric elastomers, whose pitch could be varied by biaxial extension (Kim & Finkelmann 2001). These materials could be made to lase, and the lasing wavelength of these 'rubber lasers' could be tuned by stretching the samples (Finkelmann *et al.* 2001). Haase & Ozaki demonstrated mirrorless lasing in another periodic dielectric structure, ferroelectric liquid crystals (Ozaki *et al.* 2002). The lasing wavelength in these materials can be tuned by an applied electric field (Kasano *et al.* 2003; Ozaki *et al.* 2003*a*). Recently, electrically (Strangi *et al.* 2005) and mechanically (Chanishvili *et al.* 2004) tunable lasing in helical cholesterics was demonstrated. Since the pitch depends on temperature, the lasing wavelength can be tuned by changing the temperature. Interestingly, changing the temperature results not only in smooth variations of the wavelength, but also in abrupt jumps (Funamoto *et al.* 2003; Moreira *et al.* 2004). By the incorporation of photosensitive moieties, such as azo dyes, the cholesteric pitch can be varied by illumination. Tuning of the lasing wavelength by light has been demonstrated by a number of groups (Bobrovsky *et al.* 2003; Chanishvili *et al.* 2003; Ilchishin *et al.* 2004; Shibaev *et al.* 2005). Shibaev *et al.* (2004) have also demonstrated chemical tunability, where the lasing wavelength depends on pH. Two-photon lasing, where excitation is via two-photon absorption, and where lasing occurs at a shorter wavelength than that of the pump, has been demonstrated by Bunning (He *et al.* 2003) and Shirota *et al.* (2004). If two cholesteric films are put together so that the helical structure is not continuous across the interface, an allowed state associated with the defect appears in the band gap. Lasing can occur at this wavelength; such defect mode lasing has been demonstrated by a number of groups (Ozaki *et al.* 2003*b*; Schmidtke & Stille 2003*b*; Song *et al.* 2004). The dependence of lasing thresholds on sample thickness and dye concentration has also been studied recently (Cao *et al.* 2005).

In linear optics, most materials exhibit reciprocity—the transmittance is unchanged if the source and detector are interchanged. The optical response of chiral materials is non-local and a cholesteric need not be reciprocal (Potton 2004). In an interesting demonstration of broken reciprocity, Takezoe and co-workers (Hwang *et al.* 2005) have constructed an optical diode from two helical cholesteric samples with different reflection bands, separated by a birefringent nematic slab. The composite slab is non-reciprocal, and different laser lines are seen when pumped from opposite directions.

6. Blue phases

Helical cholesteric phases are periodic only in one dimension, hence photon confinement is not expected for light propagating perpendicular to the helix axis. Nonetheless, helical cholesterics lase at extremely low thresholds. Photon confinement in these systems is discussed in detail by Genack and co-workers (Kopp *et al.* 2001, 2003).

Unlike helical cholesterics, cholesteric blue phases (BPs) are periodic in three dimensions. They are self-assembled three-dimensional PBG materials, which offer the potential of more complete photon confinement. The structural elements of cholesteric BPs are double-twist cylinders, which may be assembled to form body centred cubic (BP I) and simple cubic (BP II) structures as shown in figure 4. The band structures of the BP I and BP II phases, based on this structural model, has been calculated (Hornreich & Shtrikman 1993). Lasing has been observed in dye-doped samples of BP II (Cao *et al.* 2002); this is the first demonstration of distributed feedback lasing in three-dimensional PBG materials. One interesting aspect here is the laser emission in three orthogonal directions, due to mode coupling by the reciprocal lattice vectors of the BP II structure.

The temperature range of BPs in conventional materials is very small, typically in the range of a few tenths of degree Celsius, making experiments difficult and rendering these materials not particularly useful for device applications. In order to exploit the potential of BPs, Kikuchi *et al.* (2002) has developed polymer-stabilized materials, where a polymer network stabilizes BP I over a temperature range of 66°C. Recently, he has realized samples with sufficiently large domains to be able to demonstrate lasing over a range of 36°C (Yokoyama *et al.* 2006). In a great advance in materials development, H. J. Coles, M. N. Pivnenko & A. D. Ford (2005, personal communication) have recently produced new bimesogen materials, which, when doped with chiral additives, exhibit the BP I phase over the temperature interval 16–60°C (Coles & Pivnenko 2005). This development further opens the door towards the use of cholesteric BP materials in photonics applications. Recently, lasing has also been demonstrated in these bimesogenic BP materials (H. J. Coles, M. N. Pivnenko & A. D. Ford 2005, personal communication).

In layered smectics with chiral molecules, in addition to the helical ferroelectric phase, helical twist grain boundary phases, in analogy with the Abrikosov flux lattice, have been predicted (Renn & Lubensky 1988) and observed (Goodby *et al.* 1989). In addition to these helical structures, smectic BPs have also been discovered (Li *et al.* 1997) and characterized (Grelet *et al.* 2002). Recently, materials with an iridescent isotropic smectic BP have been developed (Yamamoto *et al.* 2005) by mixing a chiral monomer with its twin. These materials exhibit a reflection band in the range of visible wavelengths that can be tuned by temperature and/or concentration, and show potential for applications as tunable three-dimensional PBG materials.

7. Random lasing

Photon localization is possible not only in periodic structures, but also in random dielectric media. Lasing in such structures, proposed by Letokhov (1968), is via photon localization by random multiple scattering and gain in an active medium.

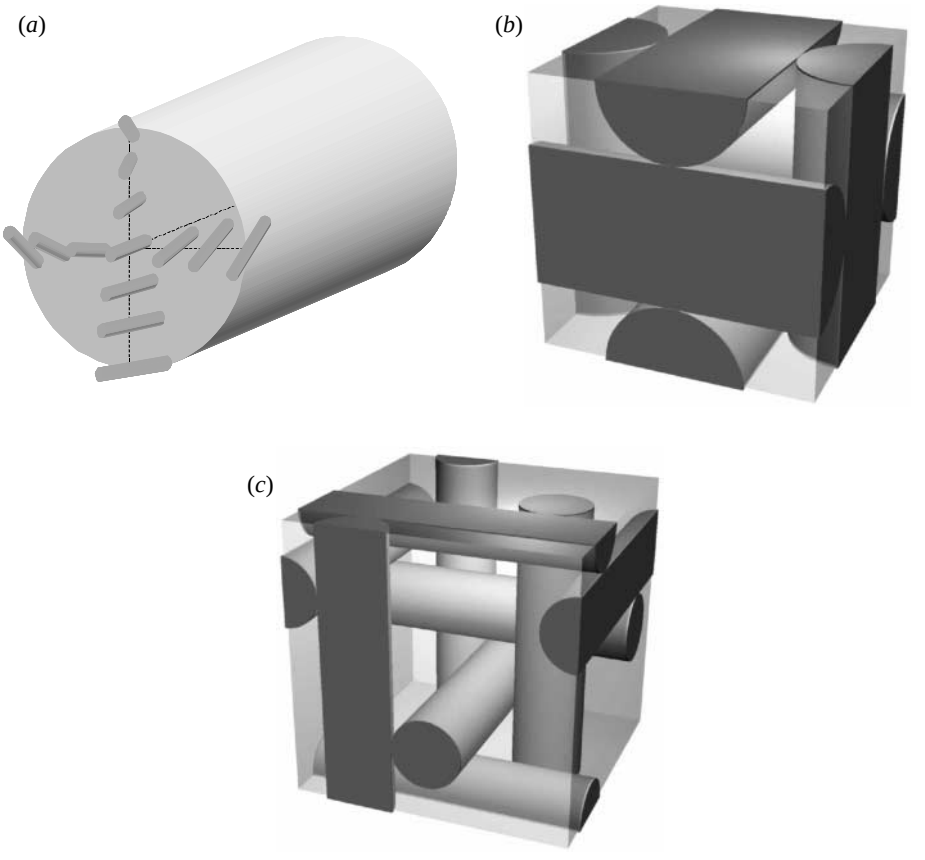

Figure 4. (*a*) Double-twist cylinder, (*b*) bcc structure of BP I and (*c*) sc structure of BP II.

Although line widths of random lasers are much greater than those of conventional lasers, both threshold behaviour and narrowing of the emission spectrum with pump intensity are exhibited. Random lasing using the liquid crystals has been demonstrated in systems composed of sintered glass infiltrated with a dye-doped liquid crystal (Wiersma & Cavalieri 2002), since the dielectric response of liquid crystals depends on temperature, these systems are temperature tunable. Recently, random lasing has been demonstrated in polymer-dispersed liquid crystals (Gottardo *et al.* 2004), where electric-field alignment of the liquid crystal changes the photon transport from three to two dimensions, and in a smectic liquid crystal with an electrically controllable focal conic structure (Morris *et al.* 2005). Random lasing by multidomain helical cholesterics has also been demonstrated (Moreira *et al.* submitted); strong scattering is demonstrated in these systems, since the scattering elements are PBG domains.

8. Conclusions

Owing to the fundamental reasons of symmetry, liquid crystals are soft materials with the possibility of periodically modulated groundstates and exceptional responsiveness to external excitations. Chiral liquid crystals can

form self-assembled PBG structures, which can be tuned by external fields. Such structures are realized in the helical cholesteric, the cholesteric blue, the helical ferroelectric and the smectic BPs. Owing to the periodic structure, light propagation is forbidden in the PBG. Since the DOS diverges at the band edges, mirrorless distributed feedback, with low threshold, occurs at the band edges. Mirrorless lasing in the liquid crystals can provide a sensitive probe of material structure.

These materials may find applications as light sources; specifically, as simple, small, low threshold, large area, flexible, tunable lasers, seed lasers or circularly polarized sources. They may be useful in display applications as switchable sources or as filters capable of modifying the spectrum and directionality of the emission of electroluminescent sources. In electro-optic and nonlinear optical applications, they may be used as optical amplifiers, electric or optical switches, fast light modulators and flexible mirrorless resonators. Owing to the sensitivity of liquid crystals to excitations, cholesteric lasers may be particularly useful as frequency encoded sensors for electric and magnetic fields, temperature and mechanical strain.

Perhaps, the greatest challenge in the field is to achieve continuous wave and current injected lasing. This requires further lowering of the lasing threshold. Understanding loss mechanisms and finding ways of reducing/eliminating losses is therefore of key importance. Significant developments in this area can be expected through new materials, such as banana and chromonic liquid crystals, stable BPs, orientationally ordered nanoparticle assemblies and structured liquid crystal elastomers and gels.

The field of photonics and lasing in liquid crystal materials has undergone dramatic growth since its relatively recent inception. Current interest in the field suggests that this growth is likely to continue, resulting in significant advances both in fundamental science and in device technology.

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References

Alvarez, E., He, M., Munoz, A. F., Palffy-Muhoray, P., Serak, S. V., Taheri, B. & Twieg, R. 2001 Mirrorless lasing and energy transfer in cholesteric liquid crystals doped with laser dyes. *Mol. Cryst. Liq. Cryst.* **369**, 75–82.

Belyakov, V. A. 1992 *Diffraction optics of complex-structured periodic Media*. Berlin, Germany: Springer.

Bobrovsky, A. Y., Boiko, N. I., Shibaev, V. P. & Wendorff, J. H. 2003 Cholesteric mixtures with photochemically tunable, circularly polarized fluorescence. *Adv. Mater.* **15**, 282–287. (doi:10.1002/adma.200390067)

Cao, W. 2005 *Fluorescence and lasing in liquid crystalline photonic band gap materials*. Chemical Physics: Ph.D., Kent State University.

Cao, W., Muñoz, A., Palffy-Muhoray, P. & Taheri, B. 2002 Lasing in a three-dimensional photonic crystal of the liquid crystal blue phase II. *Nat. Mater.* **1**, 111–113. (doi:10.1038/nmat727)

Cao, W., Palffy-Muhoray, P., Taheri, B., Marino, A. & Abbate, G. 2005 Lasing thresholds of cholesteric liquid crystals lasers. *Mol. Cryst. Liq. Cryst.* **429**, 101–110. (doi:10.1080/15421400590930782)

Chanishvili, A., Chilaya, G., Petriashvili, G., Barberi, R., Bartolino, R., Cipparrone, G., Mazzulla, A. & Oriol, L. 2003 Phototunable lasing in dye-doped cholesteric liquid crystals. *Appl. Phys. Lett.* **83**, 5353–5355. (doi:10.1063/1.1636818)

Chanishvili, A., Chilaya, G., Petriashvili, G., Barberi, R., Bartolino, R., Cipparrone, G. & Mazzulla, A. 2004 Lasing lasing in dye-doped cholesteric liquid crystals: two new tuning strategies. *Adv. Mater.* **16**, 791–795. (doi:10.1002/adma.200306542)

Coles, H. J. & Pivnenko, M. N. 2005 Liquid crystal 'blue phases' with a wide temperature range. *Nature* **436**, 997–1000. (doi:10.1038/nature03932)

deGennes, P. G. 1972 An analogy between superconductors and smectics A. *Solid State Commun.* **10**, 753–756. (doi:10.1016/0038-1098(72)90186-X)

deVries, H. 1951 Rotatory power and other optical properties of certain liquids crystals. *Acta Cryst.* **4**, 219. (doi:10.1107/S0365110X51000751)

Finkelmann, H., Kim, S. T., Muñoz, A., Palffy-Muhoray, P. & Taheri, B. 2001 Tunable mirrorless lasing in cholesteric liquid crystalline elastomers. *Adv. Mater.* **13**, 1069–1072. (doi:10.1002/1521-4095(200107)13:14<1069::AID-ADMA1069>3.0.CO;2-6)

Floquet, G. 1880 Sur lesequations diffétielles lineaires à coefficients periodiques. *CR Acad. Sci.* **91**, 880–882.

Funamoto, K., Ozaki, M. & Yoshino, K. 2003 Discontinuous shift of lasing wavelength with temperature in cholesteric liquid crystal. *Jpn. J. Appl. Phys.* **42**, L1523–L1525. (doi:10.1143/JJAP.42.L1523)

Goldberg, L. S & Schnur, J. M. 1973 Tunable internal feedback liquid crystal laser. US patent 3,771,065.

Goodby, J. W., Waugh, M. A., Stein, S. M., Chin, E., Pindak, R. & Patel, J. S. 1989 Characterization of a new helical smectic liquid crystal. *Nature* **337**, 449–452. (doi:10.1038/337449a0)

Gottardo, S., Cavalieri, S., Yaroshchuk, O. & Wiersma, D. S. 2004 Quasi-two-dimensional diffusive random laser action. *Phys. Rev. Lett.* **93**, 263 901–263 904. (doi:10.1103/PhysRevLett.93.263901)

Grelet, E., Pansu, B., Li, M.-H. & Nguyen, H. T. 2002 Faceted monodomains of liquid crystal smectic blue phases. *Phys. Rev. E* **65**, 050701–050704(R). (doi:10.1103/PhysRevE.65.050701)

He, G. S., Lin, T.-C., Hsiao, V. K. S., Cartwright, A. N., Prasad, P. N., Natarajan, L. V., Tondiglia, V. P., Jakubiak, R., Vaia, R. A. & Bunning, T. J. 2003 Tunable two-photon pumped lasing using a holographic polymer-dispersed liquid-crystal grating as a distributed feedback element. *Appl. Phys. Lett.* **83**, 2733–2735. (doi:10.1063/1.1615315)

Hornreich, R. M. & Shtrikman, S. 1993 Photonic bands in simple and body-centered-cubic cholesteric blue phase. *Phys. Rev. E* **47**, 2067–2072. (doi:10.1103/PhysRevE.47.2067)

Hwang, J., Song, M. H., Park, B., Nishimura, S., Toyooka, T., Wu, J. W., Takanishi, Y., Ishikawa, K. & Takezoe, H. 2005 Electro-tunable optical diode based on photonic bandgap liquid-crystal heterojunctions. *Nat. Mater.* **4**, 383–387. (doi:10.1038/nmat1377)

Ilchishin, I. P., Tikhonov, E. A., Tischenko, V. G. & Shpak, M. T. 1981 Generation of tunable radiation by impurity cholesteric liquid crystals. *JETP Lett.* **32**, 27–30.

Ilchishin, I. P., Yaroshchuk, O. V., Gryshchenko, S. V. & Shaydiuk, E. A. 2004 Influence of the light-induced molecular transformations on the helix pitch and lasing spectra of cholesteric liquid crystals. *Proc. SPIE* **5507**, 229–234. (doi:full_text)

John, S. 1987 Strong localization of photons in certain disordered dielectric superlattices. *Phys. Rev. Lett.* **58**, 2486–2489. (doi:10.1103/PhysRevLett.58.2486)

Kasano, M., Ozaki, M., Yoshino, K., Ganzke, D. & Haase, W. 2003 Electrically tunable waveguide laser based on ferroelectric liquid crystal. *Appl. Phys. Lett.* **82**, 4026–4028. (doi:10.1063/1.1580992)

Kikuchi, H., Yokota, M., Hisakado, Y., Yang, H. & Kajiyama, T. 2002 Polymer-stabilized liquid crystal blue phases. *Nat. Mater.* **1**, 64–68. (doi:10.1038/nmat712)

Kim, S. T. & Finkelmann, H. 2001 Cholesteric liquid single-crystal elastomers (LSCE) obtained by the anisotropic deswelling method. *Macromol. Rapid Commun.* **22**, 429–433. (doi:10.1002/1521-3927(20010301)22:6<429::AID-MARC429>3.0.CO;2-)

Kogelnik, H. & Shank, C. V. 1971 Stimulated emission in a periodic structure. *Appl. Phys. Lett.* **18**, 152–154. (doi:10.1063/1.1653605)

Kopp, V. I., Fan, B., Vithana, H. K. M. & Genack, A. Z. 1998 Low-threshold lasing at the edge of a photonic stop band in cholesteric liquid crystals. *Opt. Lett.* **23**, 1707–1709.

Kopp, V. I., Genack, A. Z. & Zhang, Z.-Q. 2001 Large coherence area thin-film photonic stop-band lasers. *Phys. Rev. Lett.* **86**, 1753–1756. (doi:10.1103/PhysRevLett.86.1753)

Kopp, V. I., Zhang, Z.-Q. & Genack, A. Z. 2003 Lasing in chiral photonic structures. *Prog. Quantum Electron.* **27**, 369. (doi:10.1016/S0079-6727(03)00003-X)

Letokhov, V. S. 1968 Quantum statistics of multi-mode radiation from an ensemble of atoms. *Sov. Phys. JETP* **26**, 835–840.

Li, M.-H., Laux, V., Nguyen, H. T., Sigaud, G., Barois, P. & Isaert, N. 1997 Blue phases and twist grain boundary phases (TGBA and TGBC) in a series of fluoro-substituted chiral tolane derivatives. *Liq. Cryst. Today* **23**, 389–408.

Moreira, M. F., Carvalho, I. C. S., Cao, W., Bailey, C., Taheri, B. & Palffy-Muhoray, P. 2004 Cholesteric liquid-crystal laser as an optic-fiber based temperature sensor. *Appl. Phys. Lett.* **85**, 2691–2693. (doi:10.1063/1.1781363)

Moreira, M. F., Gu, M., Lavrentovich, O. D., Taheri, B & Palffy-Muhoray, P. Submitted. Random lasing in multidomain cholesteric liquid crystals.

Morris, S. M., Ford, A. D., Pivnenko, M. N. & Coles, H. J. 2005 Electronic control of nonresonant random lasing from a dye-doped smectic A* liquid crystal scattering device. *Appl. Phys. Lett.* **86**, 141 103. (doi:10.1063/1.1885169)

Munoz, A., Palffy-Muhoray, P. & Taheri, B. 2001 Ultraviolet lasing in cholesteric liquid crystals. *Opt. Lett.* **26**, 804–806.

Ozaki, M., Kasano, M., Ganzke, D., Haase, W. & Yoshino, K. 2002 Mirrorless lasing in a dye-doped ferroelectric liquid crystal. *Adv. Mater.* **14**, 306–309. (doi:10.1002/1521-4095(20020219)14:4<306::AID-ADMA306>3.0.CO;2-1)

Ozaki, M., Kasano, M., Kitasho, T., Ganzke, D., Haase, W. & Yoshino, K. 2003*a* Electro-tunable liquid-crystal laser. *Adv. Mater.* **15**, 974–977. (doi:10.1002/adma.200304448)

Ozaki, M., Ozaki, R., Matsui, T. & Yoshino, K. 2003*b* Twist-defect-mode lasing in photopolymerized cholesteric liquid crystal. *Jpn. J. Appl. Phys.* **42**, L472–L475. (doi:10.1143/JJAP.42.L472)

Palffy-Muhoray, P., Munoz, A., Taheri, B. & Twieg, R. 2000 Lasing in cholesteric liquid crystals. *J. Soc. Inf. Display Symp. Dig. Tech. Pap.* **31**, 1170–1173. (doi:10.1889/1.1832874)

Potton, R. J. 2004 Reciprocity in optics. *Rep. Prog. Phys.* **67**, 717–754. (doi:10.1088/0034-4885/67/5/R03)

Renn, S. R. & Lubensky, T. C. 1988 Abrikosov dislocation lattice in a model of the cholesteric-to-smectic—a transition. *Phys. Rev. A* **38**, 2132–2147. (doi:10.1103/PhysRevA.38.2132)

Schmidtke, J. & Stille, W. 2003*a* Fluorescence of a dye-doped cholesteric liquid crystal film in the region of the stop band: theory and experiment. *Eur. Phys. J. B-Condensed Matter* **31**, 179–194. (doi:10.1140/epjb/e2003-00022-x)

Schmidtke, J. & Stille, W. 2003*b* Photonic defect modes in cholesteric liquid crystal films. *Eur. Phys. J. E-Soft Matter* **12**, 553–564. (doi:10.1140/epje/e2004-00027-2)

Schmidtke, J., Stille, W., Finkelmann, H. & Kim, S.-T. 2002 Laser emission in a dye doped cholesteric polymer network. *Adv. Mater.* **14**, 746. (doi:10.1002/1521-4095(20020517)14:10<746::AID-ADMA746>3.0.CO;2-5)

Serak, S. V., Arikainen, E. O., Gleeson, H. F., Grozhik, V. A., Guillou, J.-P. & Usova, N. A. 2002 Laser-induced concentric colour domains in a cholesteric liquid crystal mixture containing a nematic azobenzene dopant. *Liq. Cryst. Today* **29**, 19–26. (doi:10.1080/02678290110093778)

Shibaev, P. V., Tang, K., Genack, A. Z., Kopp, V. & Green, M. M. 2002 Lasing from a stiff chain polymeric lyotropic cholesteric liquid crystal. *Macromolecules* **35**, 3022–3025. (doi:10.1021/ma011738j)

Shibaev, P. V., Madsen, J. & Genack, A. Z. 2004 Lasing and narrowing of spontaneous emission from responsive cholesteric films. *Chem. Mater.* **16**, 1397–1399. (doi:10.1021/cm0305812)

Shibaev, P. V., Sanford, R. L., Chiappetta, D., Milner, V., Genack, A. & Bobrovsky, A. 2005 Light controllable tuning and switching of lasing in chiral liquid crystals. *Opt. Expr.* **13**, 2358–2363. (doi:10.1364/OPEX.13.002358)

Shirota, K., Sun, H.-B. & Kawata, S. 2004 Two-photon lasing of dye-doped photonic crystal lasers. *Appl. Phys. Lett.* **84**, 1632–1634. (doi:10.1063/1.1666996)

Song, M. H. *et al.* 2004 Effect of phase retardation on defect-mode lasing in polymeric cholesteric liquid crystals. *Adv. Mater.* **16**, 779–783. (doi:10.1002/adma.200306360)

Strangi, G., Barna, V., Caputo, R., Luca, A. D., Versace, C., Scaramuzza, N., Umeton, C., Bartolino, R. & Price, G. N. 2005 Color-tunable organic microcavity laser array using distributed feedback. *Phys. Rev. Lett.* **94**, 063903-1–063903-4. (doi:10.1103/PhysRevLett.94.063903)

Taheri, B., Palffy-Muhoray, P. & Kabir, H. 1999 Lasing in cholesteric band-gap materials. In *ALCOM Symp. Chiral materials and applications. February 18–19*, pp. 18–19. Cuyahoga Falls, OH.

Wiersma, D. S. & Cavalieri, S. 2002 Temperature-controlled random laser action in liquid crystal infiltrated systems. *Phys. Rev. E* **66**, 056612. (doi:10.1103/PhysRevE.66.056612)

Yablonovich, E. 1987 Inhibited spontaneous emission I solid-state physics and electronics. *Phys. Rev. Lett.* **58**, 2059–2062. (doi:10.1103/PhysRevLett.58.2059)

Yamamoto, J., Nishiyama, I., Inoue, M. & Yokoyama, H. 2005 Optical isotropy and iridescence in a smectic 'blue phase'. *Nature* **437**, 525–528. (doi:10.1038/nature04034)

Yokoyama, S., Mashiko, S., Kikuchi, H., Uchida, K. & Nagamura, T. 2006. Laser emission from a polymer-stabilized liquid-crystalline blue phase. *Adv. Mater* **18**, 48–51. (doi:10.1002/adma.200501355)

Discussion

D. Lacey (*Department of Chemistry, University of Hull, UK*). Would the properties of the chiral nematic material control whether the material is optically or electrically pumped?

P. Palffy-Muhoray. Yes, in essence, they would. The feasibility of electrical pumping is predicated on extremely low lasing thresholds. The lasing thresholds are determined by cavity and absorption losses; material properties play a key role in determining both of these.

J. R. Sambles (*School of Physics, University of Exeter, UK*). What are the opportunities and problems associated with current pumping of LC Lasers?

P. Palffy-Muhoray. The main challenges are the reduction of the lasing thresholds, and the efficient excitation of the gain medium. The fundamental limits on threshold reduction are not yet clearly understood. High-intensity light sources are required for photoexcitation of the gain medium; the higher the threshold, the greater the required intensity. Today, the excitation is via pumping by (current driven) pulsed lasers; as the thresholds are reduced, this will give way to flash lamps, CW lasers and eventually to LEDs and other electroluminescent sources.

J. R. Sambles And what are the key loss mechanisms?

P. Palffy-Muhoray. The competing loss mechanisms are cavity losses, which decrease with the thickness of the cholesteric sample, and absorption losses, which increase with thickness. Absorptive losses are due to linear absorption, both in the liquid crystal and the dye, and also due to dye–liquid crystal excimer formation, as evidenced by fluorescence quenching.

H. F. Gleeson (*School of Physics and Astronomy, University of Manchester, UK*). Do you see lasing in blue phases along directions other than the (100), e.g. (110) etc.? Possibly allowing angular turning?

P. Palffy-Muhoray. Laser emission has only been observed in three orthogonal directions; one parallel and two perpendicular to the cell windows. Presumably, other symmetry-allowed modes will also be excited at higher pump intensities, due to mode coupling via reciprocal lattice vectors, but we have not seen any evidence of this at the pump intensities that were used.

M. N. Pivnenko (*University of Cambridge, UK*). What limits the output energy in LC lasing?

P. Palffy-Muhoray. It is limited by the maximum attainable gain of the gain medium, and on the damage threshold of the material. We are unable to provide realistic estimates of either of these today, recent work has largely focused on low-energy behaviour near threshold.

J. R. Sambles. Would it be helpful to dispose with the dye doping and have intrinsically fluorescent liquid crystal materials? This should help improve the order parameter.

P. Palffy-Muhoray. This is an interesting question. The dye order parameter in general differs from that of the host liquid crystal, but may be higher as well as lower, depending on the molecular structure. Another relevant parameter is the angle between the absorption moment and the effective symmetry axis of the dye. We have, in fact, observed lasing in a pure liquid crystal, without any dye, where the fluorescent liquid crystal was the active medium as well as the distributed feedback host (Munoz *et al.* 2001). The band edge was tuned to the fluorescence peak in the near UV, where 'self-lasing' was observed; however, no comparison was made with dye-doped systems lasing at this frequency.