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# Non-linear optics in liquid crystals: basic ideas and perspectives

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In this paper the 'unconventional' optical non-linearities of liquid crystals are considered. The basic ideas for the interpretation of collective reorientation and of other phenomena of thermal origin are reviewed. After reporting the main results of this approach, the issues which are the major challenges for scentists in this field are recalled.



## 1. Introduction

The study of non-linear optics in liquid crystals has been one of the most exciting subjects of investigation concerning the physics of liquid crystals from the very beginning of the 1980s.

As a matter of fact, these studies started about 20 years ago with the investigation of liquid crystalline materials in the isotropic state, by looking at the enhancement of the non-linear optical response near the phase transition temperature between the isotropic and the mesomorphic state due to the increased molecular correlation [1]. The initial approach was characterized by considering the usual mechanisms leading to a non-linear optical behaviour in condensed matter (perturbations of electronic polarizabilities, molecular orientation, electrostriction, etc.). Therefore, a strong limitation on such studies was presented by the short interaction lengths available in liquid crystal samples due to the difficulty of preparing homogeneous samples thicker than 100 μm or (at most) 200 μm. The short interaction length gave rise to very weak signals from non-linear effects. For this reason there are very few papers published which report the observation of non-linear phenomena in the mesomorphic phase [2] before 1980, and most of the work done on this subject on liquid crystalline materials was concerned with the isotropic phase.

The interest in non-linear optics of liquid crystals grew after the discovery of the giant optical non-linearities [3] (GON) due to collective reorientation in nematic liquid crystals, which are several orders of magnitude bigger than in other liquids, thus allowing the observation of non-linear optical phenomena with light beams of moderate intensity. The discovery of GON showed that an optical field acts on the liquid crystal director in a way very similar to a low frequency electric field, and so the strong non-linearity is due to the possibility of getting a light-induced change of the refractive index of the same order of magnitude as the optical anisotropy of the liquid crystal, i.e. the non-linear refractive index can be as high as $\delta n = 0{\cdot}1$–$0{\cdot}2$.

Several reasons made these effects attractive to study: perculiar features are present in liquid crystals underlining new aspects of non-linear phenomena; the strong non-linear response suggests the use of these materials as media where non-linear optical effects can be easily studied with great simplification of the investigation techniques; non-linear optics can be a powerful tool to measure physical parameters of liquid crystals; new optical devices can be designed based on this non-linear behaviour.

The 'explosion' of work on GON at the beginning of the '80s drew attention to a series of other phenomena which are peculiar to liquid crystals: thermal indexing, photo-induced molecular conformations, thermal hydrodynamical effects, optically induced phase transitions, thermal reorientation, etc. All of them can lead to a strong non-linear optical response and are often mixed with collective optical reorientation making this field of investigation very wide and increasingly more attractive for an ever growing number of scientists.

The question also raised: is this field 'real' non-linear optics? This question finds its motivation in the fact that 'traditional' non-linear optics is that where the polarization of the medium can be expressed by a series expansion of the optical field, while generally this approach is not useful or is even impossible when dealing with the above mentioned collective phenomena. Moreover, this point of view considers that since some phenomena, e.g. opto-thermal effects or photo-transformations, give rise to structural changes, and obviously lead to a non-linear response, this has nothing to do with 'real' non-linear optics. However, I would consider this point of view not to be correct. First of all I believe that a definition of non-linear optics cannot be linked to the 'traditional' approach, and on the contrary, one deals with 'real' non-linear optics any time there is a possible energy exchange between waves of different frequency or wavevector. The widely studied two and four wave mixing process in liquid crystals demonstrates that this energy exchange can be induced by the phenomena mentioned above. So no matter which phenomenon is exploited to get this result, we have a non-linear optical effect when the enegy redistribution occurs, which is impossible in the frame of linear optics, when the medium response is just proportional to the optical field. Moreover, some of the effects which trivially lead to a non-linear response are not trivial at all to study and have very peculiar and interesting consequences.

## 2. Basic concepts about optical reorientation

It seems amazing that an optical field oscillating at a frequency of $10^{14}$–$10^{15}$ Hz can induce a collective molecular reorientation of a viscous nematic material with a very slow response time (seconds). However, it is trivial to understand it in a very classical way. The optical field $\mathbf{E}=\mathbf{E}_0\cos\omega t$ induces a dipole moment $\mathbf{p}=\alpha\mathbf{E}=\mathbf{p}_0\cos\omega t$ proportional to and in phase with it, so that the induced instantaneous torque on the molecule is $\tau(t)=(\mathbf{p}_0\times\mathbf{E}_0)\cos^2\omega t$. Averaging over a time that is long compared with the characteristic period of molecular motion, one gets $\tau(t)=1/2(\mathbf{p}_0\times\mathbf{E}_0)$, showing that the only difference with respect to the effect of a low frequency field is the factor 1/2.



### 2.1. *Planar reorientation*

In order to explain in a more rigorous way the phenomena related to optical reorientation in nematic liquid crystals, one usually deals with the case of in-plane deformations of the director field [4]. This geometrical configuration has been intensively studied: it requires a linearly polarized light field, a sample without any twist deformation and, moreover, optical field and director must lie in the plane of incidence of the radiation. Nevertheless, under these restrictions several different situations may occur leading to either threshold or non-threshold effects.

In general we should expect a threshold behaviour for the onset of the director reorientation if the incoming light polarization corresponds to a pure ordinary wave in the undistorted nematic. On the contrary, a non-threshold effect should be expected when an extraordinary wave is travelling in the medium.

Initially the nematic liquid crystal may have a homeotropic, planar, hybrid alignment or any other uniform deformation in the plane of incidence of the linearly polarized radiation field. In order to know the actual director orientation in the presence of the optical field, one has to find the equilibrium condition of the system in the frame of continuum theory by working out the free energy and applying to it the variational calculus. The main problem arises from the need to include in the free energy density the interaction of the medium with the electromagnetic field which, in turn, must satisfy Maxwell's equations in an inhomogeneous and anisotropic medium. In this case the director is completely detemined by the tilt angle $\theta$ with respect to the $z$ axis (normal to the boundaries). As a matter of fact, an optical wave brings both an electric and a magnetic field which can contribute to the total energy of the system. Anyway it is possible to state that the orientational contribution of the magnetic field can be disregarded.

The usual minimization of the free energy density which includes the interaction with the optical field gives an equation which must be solved taking into account that the optical field itself is dependent on $\theta$. The Geometrical Optical Approximation is usually assumed when changes of the director orientation occur over distances much longer than the light wavelength. The differential equation for $\theta$ becomes:

$$(K_1\sin^2\theta+K_3\cos^2\theta)\left(\frac{\mathrm{d}^2\theta}{\mathrm{d}z^2}\right)-(K_3-K_1)\sin\theta\cos\theta\left(\frac{\mathrm{d}\theta}{\mathrm{d}z}\right)^2+\frac{\Delta\varepsilon}{16\pi}h(\theta)=0 \quad (1)$$

where $h(\theta)$ is defined as:

$$h(\theta)=\left[\frac{g_0^2g-s^2(\varepsilon_\perp\varepsilon_\|)-\Delta\varepsilon^2\sin^2\theta\cos^2\theta g-2s\Delta\varepsilon(\varepsilon_\perp\varepsilon_\|)^{1/2}\sin\theta\cos\theta g^{1/2}}{g_0^2g^{1/2}}A^2\sin 2\theta-\left[\frac{s(\varepsilon_\perp\varepsilon_\|)^{1/2}+\Delta\varepsilon\sin\theta\cos\theta g^{1/2}}{g_0}\right]2A^2\cos 2\theta.\right] \quad (2)$$

Also, $g=\varepsilon_\perp+\Delta\varepsilon\cos^2\theta-s^2$ and $g_0=\varepsilon_\perp+\Delta\varepsilon\cos^2\theta=\varepsilon_\|-\Delta\varepsilon\sin^2\theta$, $A$ is proportional to the optical field and the other quantities have the usual meaning. In the case of normal incidence ($s=\sin\beta_0=0$) it is possible to find analytical solutions for particular cases. When $s=0$, defining the elastic anisotropy $K=1-K_1/K_3$ and using the light intensity instead of $A^2$, the equation can be integrated to obtain:

$$(1-K\sin^2\theta)\left(\frac{\mathrm{d}\theta}{\mathrm{d}z}\right)^2+f(\theta,I)=c^2 \quad (3)$$

where

$$f(\theta,I)=I\frac{2}{cK_3}\frac{(\varepsilon_\|\varepsilon_\perp)^{1/2}}{(\varepsilon_\perp+\Delta\varepsilon\cos^2\theta)^{1/2}}. \quad (4)$$

Here $c^2$ is an integration constant fixed by the boundary conditions and can be determined by integration of equation (3) taking into account the tilt angles at the boundaries ($\theta_1$ and $\theta_2$).

The director distribution $\theta(z)$ can be calculated from the integral equation:

$$\int_{\theta_1}^{\theta(z)} \left[ \frac{1 - K \sin^2 \theta'}{c^2 - f(\theta', I)} \right]^{1/2} \mathrm{d}\theta' = z + d/2 \qquad (5)$$

placing the $z = 0$ plane in the centre of the cell. A numerical solution is usually necessary to get $\theta(z)$ at each impinging intensity $I$.

The knowledge of $\theta(z)$ for each value of the impinging light intensity allows one to describe the non-linear optical effects which take place:

$$\delta\varepsilon[\theta(z, I)] = \frac{\Delta\varepsilon}{2} \left[ \varepsilon^2 - \left( \frac{\Delta\varepsilon}{2} \right)^2 \right] \times \frac{\cos 2\theta(z, 0) - \cos 2\theta(z, I)}{\left[ \varepsilon + \frac{\Delta\varepsilon}{2} \cos 2\theta(z, 0) \right] \left[ \varepsilon + \frac{\Delta\varepsilon}{2} \cos 2\theta(z, I) \right]} \qquad (6)$$

where $\theta(z, 0)$ is the zero intensity tilt angle distribution and $\theta(z, I)$ is that under the intensity $I$. Usually an optical investigation deals with the non-linear response averaged along the cell thickness:

$$\langle \delta\varepsilon \rangle = \frac{1}{d} \int_{-d/2}^{d/2} \delta\varepsilon(z, I)\, \mathrm{d}z. \qquad (7)$$

Another important quantity is the non-linear induced phase shift:

$$\psi_{\mathrm{NL}} = \frac{2\pi}{\lambda} \int_{-d/2}^{d/2} \delta n \, \mathrm{d}z = \frac{2\pi}{\lambda} \langle \delta n \rangle d \qquad (8)$$

where the non-linear refractive index is:

$$\delta n = \frac{\delta\varepsilon}{2 n^0_{\mathrm{eff}}}. \qquad (9)$$

It is worth remarking that the above expressions for $\delta n$ and $\delta\varepsilon$ include the whole effect of the field. A series expansion of them shows a square dependence on the field (linear in the intensity), but it is a good approximation only for weak fields when distortions are small. In order to explain the behaviour under strong light intensities it is necessary to use the complete expressions.

Therefore the behaviour of the non-linear dielectric permittivity $\langle \delta\varepsilon \rangle$ versus the intensity $I$ shows a linear dependence (square on the field) at low values of $I$, while a saturation occurs for high intensities, because the molecules become completely oriented along the field direction. It is interesting to underline that the maximum light power necessary to reach saturation is of the order of $10^4\ \mathrm{W\,cm^{-2}}$, which is easily obtained with a low power laser source slightly focused on the sample.

Analytical expressions of the non-linear permittivity and of the non-linear refractive index, which show more clearly the physical parameters important in determining the non-linear behaviour, can be worked out in the case of small angular distortions, i.e. under the approximation of weak reorientation under moderate light intensity. Generally there is found a square dependence on the optical anisotropy $\Delta\varepsilon$, which is responsible for the giant non-linear response. Even for low reorientation one may have $\langle \delta n \rangle = 0{\cdot}005$ which is huge compared with highly non-linear liquids where $\langle \delta n \rangle$ may reach the value of $10^{-6}$.

### 2.2. *Self-focusing and self-phase modulation*

The non-linear director reorientation gives rise to effects such as light self-focusing and self-phase modulation easily observable in nematic liquid crystals under different geometrical configurations and alignments. For instance we may consider a planar aligned cell and the impinging light beam linearly polarized in order to travel as a pure extraordinary wave into the sample with an incidence angle different from zero. One can focus the light before the liquid crystal cell, in order to have a diverging beam on the sample. Under these conditions, in the far field after the sample a remarkable decrease of the beam diameter is observed as the light intensity is increased. This fact is due to the self-focusing effect occurring inside the liquid crystal. It is due to the Gaussian intensity distribution of the beam and to the intensity dependent refractive index caused by director reorientation:

$$n = n^0 + \delta n(I). \qquad (10)$$

As a consequence, the light speed is different across the wave front and the medium acts as a positive or negative (depending on the sign of $\delta n$) non-linear lens. The extraordinary wave has a refractive index sensitive to the director orientation through $\theta$, while the ordinary wave sees always a refractive index equal to $n_\perp$ and it is not then affected by director reorientation. For this reason one is able to observe non-linear phenomena of this kind only with the extraordinary wave: the experimental data say that $n$ is changing with the intensity, so then $\theta$ too is changing. In a self-focusing experiment one can directly measure the inverse focal length of the non-linear film which can be easily related to the beam diameter measured in the far field. It can be shown that the inverse focal length of the non-linear lens can be related to the non-linear response of the liquid crystal through:

$$n_2 = \varepsilon_{\mathrm{eff}}(0)\frac{\pi}{2}\left(\frac{\zeta^4}{d}\right)\left(\frac{f^{-1}}{P}\right) \tag{11}$$

where $n = n^0 + n_2 I$ ($I$ being the intensity) and $P$ is the optical power of the beam. Therefore a measurement of $f^{-1}/P$ enables us to work out $n_2$ when the geometrical parameters $\zeta$ (beam waist on the sample) and the sample thickness $d$ are known.

By a slight change in the experimental geometry, one can observe the effect of self-phase modulation. It is just necessary to move the sample on to the focal spot of the light beam to see a strong wavefront distortion: after the sample an amazing pattern of rings appears. The Gaussian laser beam traveling in the $z$ direction produces a transverse distribution of the non-linear refractive index leading to an induced phase shift having a Gaussian distribution. That is, the wavefront experiences a phase shift dependent on the transverse coordinate. In this way, the maximum phase shift $\delta\psi_0$ is easily related to the total number of rings:

$$N = \frac{\delta\psi_0}{2\pi}. \tag{12}$$

This result comes from a qualitative description often used to work out the maximum non-linear phase shift introduced by a nematic sample and can be considered a good approximation when $N$ is large.

A more rigorous treatment is necessary to explain the details of the wavefront intensity profile. This would require the use of Fraunhofer diffraction theory, by considering the laser spot on the sample as a circular diffracting aperture which also introduces a transverse phase factor on the field given by the non-linear response of the medium. Following this procedure it is possible to work out the far field intensity distribution after the sample as a function of the diffraction angle $\alpha$:

$$\mathfrak{J}(\alpha) = \left(\frac{2\pi}{D}\right)^2 I_0 \left| \int_0^a J_0(k\rho\alpha)\rho \exp\left(-\frac{\rho}{\omega}\right)^2 \times \exp\left(-\mathrm{i}\frac{k\rho^2}{2R}\right) \exp(-\mathrm{i}\psi_{\mathrm{NL}}(\rho))\,\mathrm{d}\rho \right|^2 \tag{13}$$

where $R$ represents the wavefront curvature radius, $D$ is the distance of the observation plane from the sample, $k$ is the light wavevector and $\psi_{\mathrm{NL}}$ is the non-linear phase shift.

### 2.3. *Non-planar reorientation*

The qualitative difference between optical and dielectric reorientation has been pointed out by the new effects which have been discovered for non-planar director reorientation [5] ($\mathbf{n} = \sin\theta\cos\phi\boldsymbol{x} + \sin\theta\sin\phi\boldsymbol{y} + \cos\theta\boldsymbol{z}$) induced by light with circular and elliptical polarization. These effects have no counterpart in the static case and in particular a very important achievement has been the statement of the Angular Momentum Conservation Law:

$$\frac{\mathrm{d}}{\mathrm{d}z}\left[(K_2\sin^2\theta + K_3\cos^2\theta)\sin^2\theta\frac{\mathrm{d}\phi}{\mathrm{d}z}\right] + \frac{\mathrm{d}L_z}{\mathrm{d}z} = 0 \tag{14}$$

where $L_z$ is the light angular momentum per unit area and unit time: $L_z = (I/\omega)\sin 2\chi$ and $\chi$ is the ellipticity angle. This means that an exchange of angular momentum takes place between the light and the medium any time that $\mathrm{d}\phi/\mathrm{d}z \neq 0$, i.e. the geometry of the interaction between the liquid crystal molecules and the optical field induces a change of the azimuthal angle variation through the cell thickness. In this way light not only induces a distortion of the structure, but can also transfer to the medium part of its angular momentum. Therefore under non-stationary conditions, the light can induce a precession of the molecular director around the $z$ axis and different dynamic regimes take place: distorted equilibrium state, persistent oscillations, precession–nutation regime.

## 3. Other collective phenomena leading to non-linear response

In studying non-linear optical reorientation, it has often been found impossible to neglect thermal effects due to light absorption caused either by the liquid crystal itself or by small impurities present in the medium giving rise to a non-linear behaviour because of induced temperature changes. As a consequence the physical properties of the liquid crystal may change, thus affecting the propagation of the light beam itself. The strongest effect is due to the induced variation of the refractive indices.

This effect is often referred to as *thermal indexing* and is due to the dependence of the refractive indices of the liquid crystal on the temperature $T$. Other physical parameters are also affected by temperature variations (e.g. elastic constants), thus producing changes in the director orientation and as a consequence a non-linear optical response. Indeed, it is a peculiarity of liquid crystals that we have this interdependence of thermal response and orientation, and for this reason it is often difficult to separate thermal from orientational effects.

It is well known that for a nematic liquid crystal $\mathrm{d}n_\perp/\mathrm{d}T > 0$ and $\mathrm{d}n_\parallel/\mathrm{d}T < 0$. It is remarkable then that thermal indexing also gives rise to a non-linear behaviour in the ordinary wave and with a different sign with respect to that of the extraordinary wave. For an ordinary wave we have always a self focusing medium ($\delta n > 0$), while for the extraordinary wave the strength and sign of the non-linearity depends on the director orientation in the sample. This is a very peculiar property of liquid crystals [4].

In general it is possible to write:

$$\frac{dn_{\mathrm{eff}}}{dT} = \xi(\theta, \beta, T)\frac{n_{\perp}}{n_{\mathrm{eff}}}\frac{dn_{\perp}}{dT}. \tag{15}$$

Through the function $\xi$, this equation points out the dependence of the thermal non-linearities on the director orientation and on the incidence angle $\beta$ for an extraordinary wave, where $n_{\mathrm{eff}} = (\varepsilon_{\mathrm{eff}})^{1/2}$.

It is easy to understand now why, depending on the experimental geometries, thermal effects can be connected with optical reorientation since the director reorientation affects the local tilt angle, thus changing the thermal gradient $dn_{\mathrm{eff}}/dT$ (even its sign).

The geometry of the experiment and the thermal conductivity $\kappa$ and the absorption coefficient $\alpha$ of the liquid crystal cell determine the actual relationship between light intensity and temperature rise. A simple approximation gives the maximum temperature rise as:

$$\delta T_{\mathrm{M}} = \left(\frac{d}{\pi}\right)^2 \frac{\alpha}{\kappa} I. \tag{16}$$

The main feature shown in this equation is the linear dependence of the temperature rise on the light intensity and the square dependence on the cell thickness. This expression may be used to evaluate the expected temperature rise for a particular light intensity. Choosing $\alpha = 0{\cdot}2\,\mathrm{cm}^{-1}$, with $I = 10^4\,\mathrm{W\,cm}^{-2}$, a sample thickness $d = 50\,\mu\mathrm{m}$, and a typical value for the conductivity $\kappa = 10^{-3}\,\mathrm{W\,K}^{-1}\,\mathrm{cm}^{-1}$, we get $\delta T \approx 5\,\mathrm{K}$. Since the non-linear refractive index can be written as:

$$\delta n = \frac{dn}{dT}\delta T \tag{17}$$

and considering an index gradient $\approx 10^{-3}\,\mathrm{K}$, we obtain a non-linear refractive index $\delta n \approx 5 \times 10^{-3}$, which is comparable to that due to optical reorientation!

It is interesting to remark that it is possible to modify the sample thickness $d$ and the absorption coefficient $\alpha$ to change the induced temperature rise by several orders of magnitude. One of the most common ways to get a remarkable temperature rise with very low intensity is to dope the liquid crystal with highly absorbing dyes. However recent studies have shown that several different phenomena may occur in dye-doped liquid crystals, and therefore one should be very careful in using this technique with the unique purpose of enhancing thermal indexing.

The knowledge of the temperature rise is necessary in order to work out the corresponding non-linear refractive index, which for the extraordinary wave may be written as:

$$\delta n_T = \xi(\theta, \beta, T)\frac{n_{\perp}}{n_{\mathrm{eff}}}\frac{dn_{\perp}}{dT}\delta T \tag{18}$$

with the associated non-linear phase shift:

$$\psi_T = \frac{2\pi}{\lambda}\int_{-d/2}^{d/2}\left\{\xi(\theta, \beta, T)\frac{n_{\perp}}{n_{\mathrm{eff}}}\frac{dn_{\perp}}{dT}\delta T\right\}dz. \tag{19}$$

An increase of temperature of only 1 K, may give $\psi_T \approx -\pi$. Indeed, higher temperatures can be easily reached just by increasing the impinging light intensity or enhancing the absorption by dye dopants, or by using a working temperature near the phase transition to the isotropic state. It is then evident that the thermally induced optical phase shift can be comparable to the one due to optical reorientation. This fact often makes it difficult to separate the two effects.

Other light induced effects of thermal origin, different from thermal indexing, may occur in liquid crystals [4]. One is *thermal reorientation*, i.e. the director reorientation which may occur when some material parameter changes because of a light induced temperature variation. It may occur in distorted nematics because of the temperature dependence of the Frank elastic constants, in cholesteric liquid crystals due to the temperature dependence of the helix pitch and in ferroelectric liquid crystals where the induced tilt angle is strongly temperature dependent.

Other interesting phenomena can be due to *thermal hydrodynamical effects* such as light absorption induced flow and optically induced convection motion.

*Light induced phase transitions* from the nematic to the isotropic state represent other phenomena leading to strong non-linear optical behaviour. Two main features are remarkable in these processes: the large jump between the two refractive indices of the liquid crystal, and that of the isotropic state and the jump in the propagation properties between anisotropic and isotropic media. The most important optical effect is generally a variation of the light transmittivity between the two states, a variation which can be enhanced by proper experimental conditions.

Another class of phenomena leading to strong non-linear behaviour is due to *photo-induced changes of the molecular conformation*. These photostimulated variations in shape and structure of molecules act as light induced impurities which affect the intermolecular energy and, as a consequence, the order parameter in the vicinity of the impurity. In this way a large change of the refractive index may be induced which is of the same order of magnitude as that due to optical reorientation. The study of these phenomena is becoming increasingly important for the recording of optical information.

## 4. Main issues of present and future investigations

In the following I will mention, in my opinion, the most interesting and promising subjects of investigation concerning the non-linear optical properties of liquid crystals [6].

### 4.1. *Photorefractive effects*

The photorefractive effect arises from optically generated charge carriers which migrate under spatially varying patterns of illumination. The migration due to diffusion and the action of a d.c. bias field produces a space-charge separation which gives rise to a strong space-charge field. The consequent strong modulation of the refractive index makes photorefractive materials very efficient for non-linear wave mixing processes. For this reason they can be employed in many applications: information storage, image processing and optical neural networks. The effect has also recently been demonstrated in nematic liquid crystals doped by dyes or fullerene. However, again liquid crystals have shown their amazing features since besides the 'conventional' diffusive photorefractive effect due to induced photoconductivity by impurities, a peculiar photorefractive phenomenon due to Helfrich's effect has been demonstrated. This is at present the strongest of the non-linear effects known in liquid crystals leading to director reorientation with a modulation of the LC refractive index three orders of magnitude higher than in GON!

### 4.2. *Dye enhanced reorientation*

In nematic liquid crystals doped by anthraquinone dyes, the threshold for the optical Fréedericksz transition can be reduced by two orders of magnitude allowing *sub-milliwatt* light power to induce the effect. The experimental observations demonstrate that this phenomenon is not due to thermal effects. It seems that the excited dye molecules introduce an additional torque on the molecules which gives rise to an additional contribution to molecular reorientation. It has also been found that the effect can be *positive*, enhancing the reorientation effect towards the light polarization direction, or *negative*, stabilizing the director orientation against the light effect. The nature of the phenomenon is not yet understood and may be interpreted as due to a new type of molecular interaction.

### 4.3. *Thermal reorientation in ferroelectrics*

Very little work has been done up to now on the non-linear optical properties of liquid crystals in phases different from nematics. Among them, and very interesting and promising for possible applications, are the experiments performed on ferroelectric liquid crystal materials both in the SmC* and SmA* phases. The starting point is the strong temperature dependence of the tilt angle in these phases. In the case of electroclinic effect, this property gives rise to an original coupling between the orientational and thermal properties of the liquid crystal. These studies have shown the occurrence of unexpected reorientational phenomena probably due to light induced flow, with consequent director reorientation.

### 4.4. *Non-linear optics in composite liquid crystalline materials*

The investigation of non-linear optical properties in materials containing liquid crystals and polymer has been limited in the past few years to polymer dispersed liquid crystals (PDLC) revealing several peculiar features such as the self-transparency effect, all optical bistability and voltage controlled self-diffraction. The examples mentioned point out that the amazing non-linear properties of liquid crystals combined with the particular structure of a composite material may give rise to very original properties which may suggest new applications. It is believed that other materials of this kind such as filled nematics or liquid crystal dispersed polymers deserve investigations in this direction which have not been performed as yet.

### 4.5. *Pattern formation*

Complex spatial structures may arise in the transverse profile of the laser radiation due to non-linear optical interaction. These effects become easy to observe using liquid crystals because of the giant non-linear response, and are usually obtained under a single mirror feedback configuration or in Fabry–Perot cavities. First of all, these patterns are amazing since a redistribution of the light wavefront occurs in regular geometrical structures, but they are also very interesting for the fundamental investigation of spatio-temporal instabilities. From the point of view of applications, these patterns are very attractive for optical information processing. Even if these effects are not peculiar to liquid crystals, they are easier to produce and study in these materials and this can make applications really feasible.

### 4.6. *Fibre optics applications*

In the last few years there has been a growing interest in studying light propagation in guided structures with liquid crystals as the core or cladding materials. One of the most addressed issues concerns the possibility of using the strong non-linear response of liquid crystals to achieve an all optical switching between different guided modes. The possibility of having active optical connections between fibres has also been considered. The published papers represent only the starting point of this research subject which may find great development in the near future if it is accompanied by a parallel development of the technology needed to make the envisaged applications feasible.

### 4.7. *Light induced anchoring and optical recording*

This subject is not strictly related to non-linear optics since it is more connected to photochemistry and phototransformation. However, it is often linked to the existence of a strongly non-linear material. At present much effort is devoted in this direction since liquid crystalline materials may play a role which is not negligible in the development of new technology for optical storage. The investigations in this field follow different paths, e.g. photoisomerization processes in liquid crystal polymers, light induced surface orientation, surface mediated photopolymerization, change of anchoring conditions by light action in the liquid crystal bulk and photoalignment of Langmuir–Blodgett films. Some of these techniques are among the most sensitive existing today, since they require vey low energy density for the recording of optical information.

## References

[1] Wong, G. K. L., and Shen, Y. R., 1974, *Phys. Rev. Lett.*, **32,** 527; Narashima Rao, D. V. G. L., and Jayaraman, S., 1974, *Phys. Rev. A*, **10,** 2457; Prost, J., and Lalanne, J. R., 1973, *Phys. Rev. A*, **8,** 2090; Lalanne, J. R., 1975, *Phys. Lett.*, **51A,** 74; Wong, G. K. L., and Shen, Y. R., 1974, *Phys. Rev. A*, **10,** 1277; Hanson, E. G., Shen, Y. R., and Wong, G. K. L., 1976, *Phys. Rev. A*, **14,** 1281.

[2] Shelton, J. W., and Shen, Y. R., 1972, *Phys. Rev. A*, **5,** 1867; Saha, S. K., and Wong, G. W., 1979, *Appl. Phys. Lett.*, **34,** 423; Saha, S. K., and Wong, G. W., 1979, *Opt. Commun.*, **30,** 119.

[3] Herman, R. M., and Serinko, R. J., 1979, *Phys. Rev. A*, **19,** 1757; Zel'dovich, B. Ya., and Tabiryan, N. V., 1979, *JETP Lett.*, **30,** 479; Zel'dovich, B. Ya., Pilipetskii, N. F., Sukhov, A. V., and Tabiryan, N. V., 1980, *JETP Lett.*, **31,** 264; Khoo, I. C., and Zhuang, S. L., 1980, *Appl. Phys. Lett.*, **37,** 3; Zolot'ko, A. S., Kitaeva, V. F., Kroo, N., Sobolev, I. N., and Csillag, L., 1980, *JETP Lett.*, **31,** 264.

[4] Simoni, F., 1997, *Nonlinear Optical Properties of Liquid Crystals and PDLC* (Singapore: World Scientific); Khoo, I. C., and Simoni, F. (editors), 1991, *Physics of Liquid Crystalline Materials* (Philadelphia: Gordon and Breach).

[5] Santamato, E., Daino, B., Romagnoli, M., Settembre, M., and Shen, Y. R., 1986, *Phys. Rev. Lett.*, **57,** 2423; Santamato, E., Abbate, G., Maddalena, P., and Shen, Y. R., 1987, *Phys. Rev. A*, **36,** 2389.

[6] References to all the subjects mentioned can be found in the *Proceedings of the* 6*th International Topical Meeting on Optics of Liquid Crystals* '95, edited by M. Warenghem, 1996, *Mol. Cryst. liq. Cryst.*, **282,** 1–472.