# Novel nonlinear optical phenomena in nematic liquid crystals

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Abstract | In normal materials, the nonlinear optical effects arise from nonlinearities in the polarisabilities of the constituent atoms or molecules. On the other hand the nonlinear optical effects in liquid crystals arise from totally different processes. Also they occur at relatively low laser intensities. In a laser field a liquid crystal exhibits many novel and interesting nonlinear optical effects. In addition we also find laser field induced effects that are peculiar to liquid crystals, like structural transformations, orientational transitions, modulated structures and phase transitions, to name a few. Here we dwell upon a few of these interesting and important nonlinear optical phenomena that exist in nematic liquid crystals.

## Dedication

The contributions of Prof. Alfred Saupe, to liquid crystal physics, are of immense significance and of lasting value. He had something new and original to say on almost every aspect of liquid crystalline state. His theoretical analysis of the nematic-isotropic transition, his great idea that nuclear magnetic resonance can be used to elucidate molecular structure, his conjectures on the defect structure of the blue phase of cholesteric liquid crystals, his discovery of a biaxial nematic phase in a lyotropic liquid crystal, development of the elastic theory of the smectic–C and biaxial nematic liquid crystals are only a few of his contributions that have become classics in liquid crystal physics. The author deems it an honour to dedicate this article to his memory.

## 1. Liquid crystals

A crystal is characterized by two types of order viz. orientational order and positional order. Its constituent molecules are not only positioned in a periodic way but their orientations are also well defined. The liquid state on the other hand is a random collection of randomly oriented molecules. Thus the familiar liquid state lacks both positional and orientational orders. Incidentally, normal liquids are also referred to as isotropic liquids. Liquid crystals are in a state in between these familiar crystalline and the liquid states. They are invariably made of highly shape anisotropic molecules. In the liquid crystalline state these anisotropic molecules are spatially organized with a well defined orientational order. In some liquid crystals the molecular aggregation may even have one or two dimensional positional or lattice order superposed on the orientational order[1]. Two characteristic features of all the liquid crystals are that they flow like liquids and are anisotropic like crystals, hence the name Liquid Crystals. In short, they not only exhibit double refraction like calcite crystals but they also flow like water. Liquid crystals come in two verities namely thremotropic and lyotropic liquid crystals. The phase transitions of thermotropic liquid crystals depend on temperature, while those of lyotropic liquid crystals depend on both temperature and concentration. Incidentally lyotropic liquid crystals were discovered long before the discovery of thermotropic liquid crystals. In this article we consider only thermotropic liquid crystals. On cooling, a thermotropic liquid crystal

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may either go through other liquid crystalline states or may directly crystallize. Similarly, on heating, it may either go through other liquid crystalline states or go over directly to the isotropic liquid state.

### 1.1. *Nematic liquid crystals*

Nematic liquid crystals are the simplest of the liquid crystals. They possess only orientational order. Depending upon the shape of molecules and the interaction between them, the medium as a whole can be either optically uniaxial[1] like calcite crystals or biaxial[1] like aragonite crystals. In this article we shall confine ourselves to uniaxial nematic liquid crystals. We have already stated that molecules that form liquid crystals are highly shape anisotropic. For example, the molecules could be either rod like or disk like in shape. If the molecules are rod like, then in a nematic liquid crystal they will all be nearly oriented in a particular direction. A schematic representation of such a liquid crystal is given in figure1.

In the case of disk like molecules, the disks will all be nearly oriented perpendicular to a particular direction. In both the cases the average molecular distribution will be cylindrically symmetric about this direction. This direction is called the director and it has been represented in the figure by the vector **n**. Thus, such nematic liquid crystals are uniaxially anisotropic about the director. This means that the diamagnetic susceptibility $\chi$ or the optical dielectric constant $\varepsilon$ varies with direction inside the medium, being cylindrically symmetric about the director. If $\chi_3$ and $\chi_1$ are respectively the diamagnetic susceptibilities parallel and perpendicular to the director then medium has a diamagnetic anisotropy of

$$\Delta\chi = \chi_3 - \chi_1$$

Similarly we can say that a nematic liquid crystal has a dielectric anisotropy of

$$\Delta\varepsilon = \varepsilon_3 - \varepsilon_1$$

Here $\varepsilon_3$ and $\varepsilon_1$ are respectively the optical dielectric constants parallel and perpendicular to the director. It should be remarked that $\Delta\chi$ or $\Delta\varepsilon$ will be positive or negative depending upon whether the nematic liquid crystals are made up of rod like or disk like molecules.

Even from the point of view of mechanical properties, nematic liquid crystals are interesting. The constituent molecules can easily slide past each other either along or perpendicular to the director. This results in the fluidity of the medium. The fluidity is not the same in all the directions. In general, in the case of rod like molecules, the viscosity is lower along the director than in the perpendicular direction. On the other hand, in the case of disk like molecules the viscosity is, in general, higher along the director than in the perpendicular direction. Further, a specimen of nematic liquid crystal with the director every where in the same direction has the lowest energy. Any distortion in the director field is associated with elastic energy. The energy associated with a particular type of distortion is again dependent on whether the nematic liquid crystal is made up of rod like or disk like molecules.

We consider throughout this article nematic liquid crystals made up of rod like molecules.

We have already stated that nematic liquid crystals are characterized by orientational order. The state of molecular aggregation in such a liquid crystal can be described by an order parameter. Saupe, defined the orientational order present in the medium in terms of the angle $\phi$ made by a rod like molecule with respect to the director. It is given by[1]

$$S = \langle (3\cos^2\phi - 1)/2 \rangle$$

The angular brackets represent the thermal average. This quantity properly describes the nematic and the isotropic states. A nematic liquid crystal, with all its rod like molecules perfectly aligned along the director has $\phi = 0$ everywhere and this leads to an orientational order $S = 1$. In the isotropic phase the angle $\phi$ can take all possible values in three dimensions resulting in $\langle\cos^2\phi\rangle$ being equal to $1/3$ or $S = 0$. The normal nematic

Figure 1: A typical nematic liquid crystal made of rod-like molecules.

liquid crystalline state has an order parameter between these two extreme values i.e. $0 < S < 1$. As the temperature is increased, a nematic liquid crystal undergoes a transition to the isotropic state at a precise temperature. At this temperature there is a sudden fall in the order parameter from a finite value to zero and the phase transition is first order in nature with a small latent heat. The essential features of this transition were first successfully explained by Maier and Saupe.[2] They pointed out that dispersion or van der Waals forces between anisotropic molecules play a dominant role in the formation of nematic liquid crystals. They predicted that the jump in the order parameter at this nematic-isotropic transition is equal to 0.44.

In this description of nematic liquid crystals, the orientational order is related to the microscopic arrangement of molecules in the nematic state. But in reality we must appeal to a measurable macroscopic property to describe the system. In other words it is important to know how this orientational order parameter $S$ is related to the bulk properties of nematic liquid crystals. In an important work, Saupe and Englert [3] showed that the order parameter $S$ is related to a measurable bulk property like the diamagnetic anisotropy $\Delta\chi$ of the nematic liquid crystal. The exact relationship is[1]

$$\Delta\chi = N(\Delta\chi_0)S$$

Here $N$ is the number of molecules per unit volume and $\Delta\chi_0$ can be obtained from a measurement of the diamagnetic susceptibility in the crystalline state of the liquid crystal, in which the molecular orientations are in principle known. In fact $S$ is also proportional to the optical dielectric anisotropy $\Delta\varepsilon$ of the nematic liquid crystal.

## 2. Nonlinear optical phenomena

Liquid crystals are "soft" materials and are extremely sensitive to external electric and magnetic fields. The electric field of the laser results in the nonlinear optical effects that are seen in these systems. At a gross level, we can identify three important processes that can take place in a nematic liquid crystal in a laser field. They are:

1. Mechanical orientational torque on the director
2. Change in the order parameter
3. Heating due to laser absorption

These processes can either individually or collectively operate leading to many different situations. In each case, we find very many new optical effects, some of which do not even have their counterparts in the familiar nonlinear optics of crystals. Even in situations where we encounter the familiar nonlinear optical effects, the underlying mechanisms are often very different. Further, we can have additional effects if a magnetic field is also present. In this article we highlight a few of these unusual nonlinear optical phenomena found in nematic liquid crystals.

### 2.1. *Orientational transitions*

#### 2.1.1. *Free nematic liquid crystals*

As already stated the usual nematic liquid crystal has uniaxial symmetry. With rod like molecules, the medium will have positive dielectric and diamagnetic anisotropies. This means that dielectric constant or the diamagnetic susceptibility parallel to the director is more than that perpendicular to it. The associated energy in an electric or magnetic field will depend on the direction of the field with respect to the director. The energy will be the least when the director is along the external field. As a consequence, an external electric or magnetic field exerts a torque on the director when it is acting at an angle to it. The director gets oriented along the applied field.

Let us consider a simple geometry where the applied magnetic field $\mathbf{H}$ and the electric field $\boldsymbol{\varepsilon}$ of the linearly polarized laser field are at right angles to each other. This situation is depicted in figure 2.

The magnetic free energy density is given by[1]

$$F_m = F_m^\circ - (1/2)H^2(\Delta\chi)\sin^2\theta$$

The optical free energy density is given by[4]

$$F_e = F_e^\circ - I\sqrt{[(\varepsilon_3\varepsilon_1)/(\varepsilon_1 + \Delta\varepsilon\sin^2\theta)]}$$
$$I = (|\boldsymbol{\varepsilon}|^2/8\pi c)\sqrt{(\varepsilon_1\varepsilon_3)}$$
$$c = \text{velocity of light}$$

Figure 2: A nematic liquid crystal in the field of a laser and a crossed magnetic field.

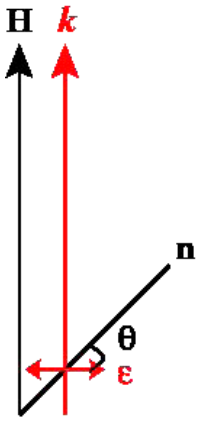
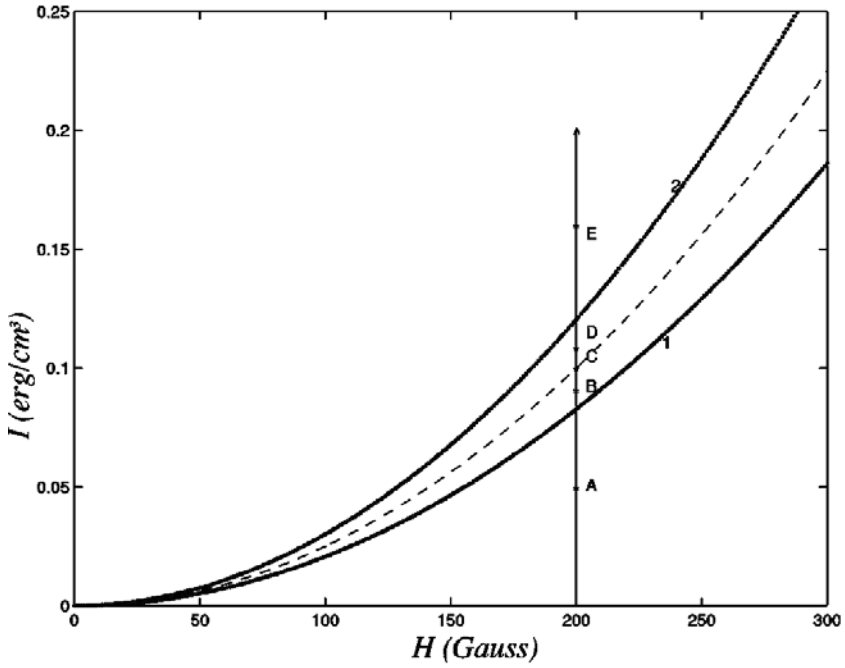


Figure 3: Phase diagram for the orientational transitions in a nematic liquid crystal.




Here $\theta$ is the angle made by the director **n** with respect to the electric field of the laser field; $\Delta\chi$ and $\Delta\varepsilon$ are respectively the diamagnetic and optical dielectric anisotropies and both are taken to be positive quantities. The quantity $I$ is a measure of the laser intensity. The constant terms $F_m^\circ$ and $F_e^\circ$, depend only on the material parameters and the field strengths. Since $F_m$ or $F_e$ is a function of $\theta$ we conclude that the field exerts a torque on the director. The director will take up an orientation that minimizes the total energy.

If either the laser or the magnetic field alone is present, then it is easy to show that the director will orient along the field direction. On the other hand, when both the laser and the magnetic fields are acting we get a very interesting result. In this case the free energy density is

$$F = F_e + F_m$$

Generally, the optical dielectric anisotropy will not be as strong as the static dielectric anisotropy. In such a case we can simplify F to get:

$$F = F_0 - (1/2)[H^2(\Delta\chi) - I\Delta\varepsilon(\sqrt{\varepsilon_3}/\varepsilon_1)]\sin^2\theta$$

We see that, by varying the strength of the magnetic field $H$ and or the intensity $I$ of the laser, the sign of the second term can be changed. When laser field term is larger than the magnetic field term, the second term becomes positive. Then $\theta = 0^\circ$ state is of lowest energy. This means the director will align itself along the electric field of the laser. On the other hand, when the magnetic field term is larger than the laser field term then $\theta = 90^\circ$ state is of lowest energy and the director will align itself along the magnetic field. This transition from one orientation to the other is of second order and takes place at

$$(H^2/I) = (\Delta\varepsilon/\Delta\chi)(\sqrt{\varepsilon_3}/\varepsilon_1) = \text{Constant}$$

It is easy to see that, $I$ verses $H$ represents a parabola. In an $I$–$H$ diagram, above this parabolic curve we find $\theta = 0^\circ$ to be the state of lowest energy and below this curve we find $\theta = 90^\circ$ state to be of the lowest energy. The parabolic curve represents a second order phase boundary between these two orientational states.

When the optical dielectric anisotropy is not small, then we have to use the full expression for $F_e$. We can work out the allowed director orientation, by minimizing the net energy. In such a case we get a very interesting phase diagram[4] . This is shown in the figure 3.

In the region A the director will be along the magnetic field ($\theta = 90^\circ$). In the region $E$ the director will be along the electric field of the laser ($\theta = 0^\circ$). The dashed line C represents a first order transition between these two states. Interestingly, in the region

B the director can be along the electric field but it will be a metastable state. Similarly, in the region D the director can be along the magnetic field but it again will be a metastable state. Hence as we increase the laser intensity at a given magnetic field, the director will be along the magnetic field not only up to the first order boundary line C but also in the region D. The region D is akin to super heating (or cooling) seen in usual first order transitions. The director switches over to the direction of the electric field of the laser, at the boundary line 2 between the regions D and E. In the same way, as we lower the laser intensity from the region E, the director will be along the electric field not only up to the first order phase boundary C but it will continue to be in the same state in the region B also. It will switch over to the direction of the magnetic field, at the boundary line 1 between B and A.

2.1.2. *Confined nematic liquid crystals*

We considered above, a free nematic liquid crystal without bounding surfaces. It is possible to confine a nematic liquid crystal between thin plane glass cover slips, with a spacer of definite thickness. The bounding surfaces can be so treated, mechanically or chemically, that the director is anchored perpendicular or parallel to the surface. We shall consider the case where the director is anchored perpendicular to the bounding surfaces. Then, throughout the volume of the sample the director will be in a direction perpendicular to the two confining plane walls.

We apply a magnetic field **H** parallel to the walls i.e. perpendicular to the director as shown in figure 4(a).

The free energy density in this case becomes[1]

$$F = F_0 + (1/2)K(\partial\theta/\partial z)^2 - (1/2)H^2(\Delta\chi)\sin^2\theta$$

Where the z-direction is normal to the bounding plane surfaces and $\theta$ is the tilt of the director away from this direction in the presence of the magnetic field. This tilt varies as we go from one boundary to the other. The second term expresses the energy involved in the distortion of the director. It is in the nature of deformation energy and hence, K is called the elastic constant of the nematic liquid crystal. The third term is the energy density due to the magnetic torque on the director.

We assume strong anchoring at the boundaries. This means, the external field does not affect the orientation at the walls.

$$\theta = 0 \text{ at } z = 0 \text{ and } z = d.$$

Here $d$ is the sample thickness.

We express the spatial variation of $\theta$ as a Fourier series. If $\theta$ is small, then the fundamental harmonic is enough for our discussions. It is given by

$$\theta = \theta_0 \sin(\pi z/d)$$

In this approximation we calculate the total energy by integrating $F$ over the sample thickness, to get

$$G = G_0 + (1/4)[K(\pi/d)^2 - H^2(\Delta\chi)]\theta_0^2$$
$$\Delta G = G - G_0 = (1/4)[K(\pi/d)^2 - H^2(\Delta\chi)]\theta_0^2$$

Here $G_0$ is the total energy of the undistorted nematic liquid crystal. We find that the presence of the distortion $\theta$, leads to a positive $\Delta$G, as long as the term inside square brackets is positive. Hence, the distortion, described by $\theta$ is not allowed energetically. The distortion will spontaneously set in, only when this term becomes negative. This happens when the magnetic field strength exceeds a threshold (critical) value

$$H_c = (\pi/d)\sqrt{(K/\Delta\chi)}$$

This process, by virtue of which, a uniformly oriented nematic liquid crystal undergoes a transition to a distorted state, above a threshold field, is called the Freedericksz Transition. Figure 4(b) depicts this situation.

Analysis of the distortion, leads to the fact that it is a second order transition. This field induced distortion can be detected optically.

If the diamagnetic anisotropy $\Delta\chi$ is known, then we can determine the elastic constant K, from a measurement of the threshold field $H_c$ and the sample thickness $d$. It is found that K is of the order of $10^{-6}$ dyne. This is pretty small and that is the reason why nematic liquid crystals are often referred to as soft materials. Earlier measurements of elastic constants of nematic liquid crystals by Zvetkov using this method were beset with errors. Saupe used this method and got very reliable values. He undertook this investigation, to confirm his theory of the nematic-isotropic transition, which led to definitive relationships between elastic constants and the orientational order parameter $S$. The theory also gave the relative magnitudes of the elastic constants[1].

Instead of a magnetic field, we can use a laser field to induce a similar director distortion. The same type of distortion is seen, if a linearly polarized laser beam is propagating perpendicular to the bounding surfaces. This situation is shown in figures 4(c) and 4(d).

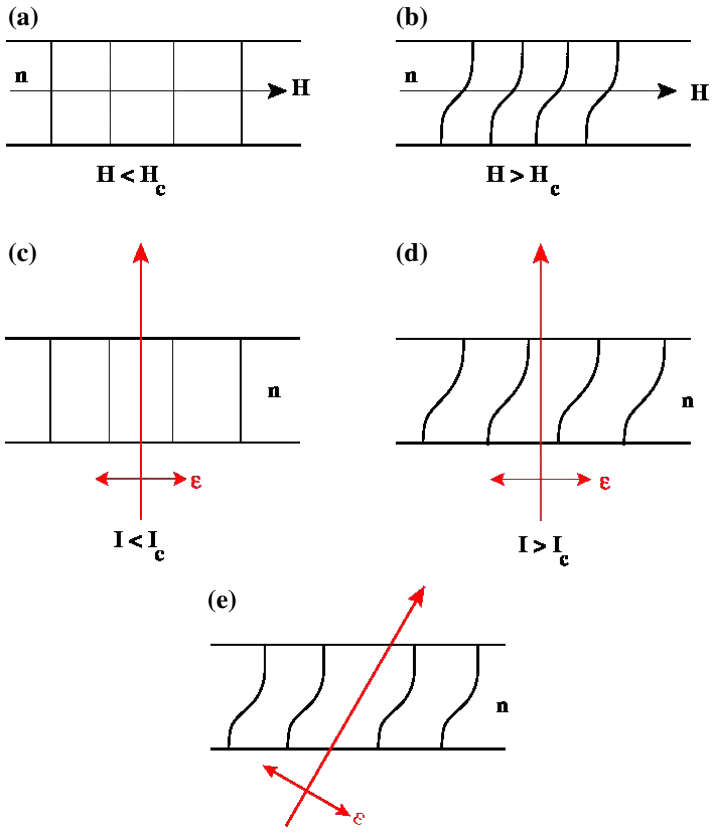



Figure 4: (a) Aligned nematic liquid crystal in a magnetic field. (b) Distortion in a nematic liquid crystal above a critical magnetic field. (c) A nematic in a laser field below the critical intensity. (d) A nematic liquid crystal in a laser field above the critical intensity. (e) A nematic liquid crystal in a laser field at oblique incidence.

We get a director distortion, in the plane defined by the electric field of the laser and the direction of laser propagation. We find that the critical laser intensity is given by

$$I_c = (\pi/d)^2[(\varepsilon_1/\sqrt{\varepsilon_3})K/\Delta\varepsilon]$$

This phenomenon is also referred to as the Optical Freedericksz Transition. This is a nonlinear optical effect due to the torque exerted by the laser field on the director. It has been observed experimentally, even with low power lasers. The advantage of this method over the magnetic field method is that it is easier to measure the optical dielectric constants and their anisotropies than diamagnetic susceptibilities and their anisotropies. Also, the laser beam itself can be used to detect the field induced distorted state. Further, for high optical anisotropy the laser induced Fredericks Transition can even become first order[5,6].

When the external electric or magnetic field is not perpendicular to the director, there will be no threshold field. Director distortion sets in the moment the field is turned on. This happens naturally in the case of the laser beam at oblique incidence, with the electric vector in the plane of incidence. This geometry is shown in figure 4(e).

The effective change in the refractive index due to director distortion is given by[5]

$$\langle \Delta\mu \rangle = \beta I$$

$$\beta = (\sqrt{\varepsilon_1}/\varepsilon_3^{3/2}/12Kc)(\Delta\varepsilon^2 d^2 \sin^2\psi\cos^2\psi)$$

Here $\psi$ is the internal angle of incidence of the laser beam. This process is akin to the familiar Optical Kerr Effect. The change in refractive index is linearly dependent on laser intensity. Interestingly, the nonlinear coefficient $\beta$ depends on sample thickness. For a 100 micron thick sample, at $\psi = 15°$, we find it to be about $10^{-5}$ cm$^2/W$. Thus it is several orders larger than that found in even highly nonlinear crystals. For an intensity of $2.5 \times 10^2 W/\text{cm}^2$, we get change in refractive index to be of the order of $2.5 \times 10^{-3}$ !

### 2.2. *A new nonlinear process*

A nematic liquid crystal appears clear and transparent, only when it is very thin and its thickness is less than about 100 microns. However, very thick samples or samples in bulk, will be turbid in appearance like milk. In the early days of liquid crystal research, it was thought that the turbidity of the liquid crystal arises, as in milk, from the heterogeneity of the medium. It was postulated that the medium is in general made up of randomly oriented blobs of nematic liquid crystals. This is referred to as the swarm model of nematic liquid crystals. Since the blobs are optically anisotropic, the medium will strongly scatter light. Later experiments, completely disproved this swarm model. It was established that the thermal fluctuations, in the orientation of the director, are responsible for the turbidity of nematic liquid crystals. It was noticed long ago in 1918 by Moll and Ornstein that the optical transmission by a nematic liquid crystal, increased when it was kept in a magnetic field. That this is due to the suppression of director fluctuations by the magnetic field was pointed out by de Gennes[1], who also worked out the enhancement in the dielectric anisotropy due the external magnetic field. The same effect can be expected, when a laser beam is passing through the liquid crystal. When a linearly polarized beam is passing through a nematic liquid crystal, with its electric vector parallel to the director, then the electric field of the laser beam suppresses the thermal fluctuations in the director, leading to an increase in the dielectric constant along the director[7]. The

corresponding increase in the refractive index along the director is given by:

$$\Delta\mu = \eta\sqrt{I}$$
$$\eta = (kT/\mu)\sqrt{\{[\Delta\varepsilon/4\pi K]^3/4c\}}$$

Here $k$ is the Boltzmann constant, $K$ is the elastic constant involved in director distortions, $\mu$ is the refractive index along the director without thermal fluctuations and T is the sample temperature.

We notice that the increase in refractive index varies as the square root of the laser intensity and is unlike the familiar nonlinear optical phenomenon where the change in refractive index is proportional to the laser intensity. Estimates of the effect indicate that the change in refractive index $\Delta\mu$ is of the order $10^{-4}$ for a laser intensity of $10^5$ kW/m$^2$. This is three orders of magnitude more than what we get for normal Optical Kerr Effect in isotropic liquids. It is therefore a measurable and an important nonlinear optical effect.

We have stated in the previous section that the laser field changes the orientation of the director. This by itself can lead to a change in the refractive index and it is a well studied phenomenon[5,6]. We have already discussed one such effect. If the director is already strongly oriented in a particular direction, due to anchoring at the walls of the sample, then the laser field reorients the director in the bulk of the sample. We have stated that this leads to a change in the local refractive index, as seen by the laser field and the effective change in refractive index varies linearly with laser intensity, like in the familiar nonlinear optics. In this sense, what we have discussed in this section, represents a new nonlinear optical process in nematic liquid crystals. This process is operative even in other liquid crystals as well and leads to many new effects[7].

It must be noted that the laser induced increase in the refractive index along the director is equivalent to an increase in the orientational order parameter of the nematic liquid crystal. There are many interesting consequences of this process[7]. We will consider here only two of them.

2.2.1. *Light scattering*

We have already stated that the turbidity of nematics is due to the thermal fluctuations in the director. The scattering cross- section per unit volume is given by[1]

$$\Sigma = (\Delta\varepsilon\pi/\lambda^2)^2\{kT/[Kq^2]\}$$

Here $q$ is the wave vector of the director fluctuation. When a linearly polarized laser beam is propagating through a nematic liquid crystal, with its electric vector along the director then we get[7]

$$\Sigma = (\Delta\varepsilon\pi/\lambda^2)^2\{kT/[Kq^2 + (\Delta\varepsilon I/8\pi c)]\}$$

Thus the laser field reduces the scattering of light. This leads to an increase in the transmitted laser intensity. The turbidity of the nematic liquid crystal decreases with increase in laser intensity.

2.2.2. *Modulated nematic liquid crystals*

When a laser beam is made to undergo a mirror reflection at a surface then a standing wave will be set up with its nodes and antinodes. The intensity at the antinode will be four times the laser intensity and there will be no intensity at the nodes. Thus we can generate a spatially periodically varying electric field. We consider the standing wave due to a linearly polarized laser field.

We have already seen that when the electric field of the laser field is along the director, then it suppresses the director fluctuations leading to an increase in the orientational order parameter. Now consider a nematic liquid crystal with its director along the field direction of the standing wave. The laser suppression of the director fluctuations will be a maximum at an antinode and it will be absent at a node. Thus we get a nematic liquid crystal with a periodically varying order parameter. The refractive index along the director will be a maximum at an antinode and it will be a minimum at a node. The structure will be periodic perpendicular to the director. The period is equal to half the wavelength of the laser light. Thus we get a spatially modulated nematic liquid crystal using a standing wave field[7]. This structure is schematically depicted in figure 5. There is no simple way of producing such modulated nematic liquid crystals.

### 2.3. *Absorbing liquid crystals*

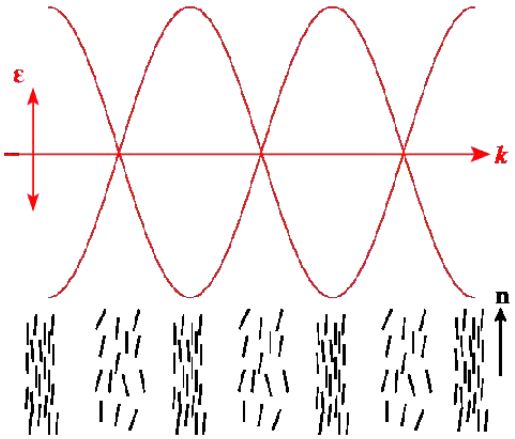
We can dope a transparent medium with molecules that absorb the laser light. As a result of this the intensity of the laser beam falls as it propagates through the sample. The law governing the intensity reduction in a medium of optical absorption coefficient $\alpha$ is

$$I(z) = I(0)\exp(-\alpha z)$$

In the simple model of laser heating, the intensity lost by the laser appears as heat in the medium with a consequent rise in temperature. This results in changes in its properties including its refractive index. In steady state, the rise in temperature due to laser absorption is given by[5]

$$\delta T = (\alpha d^2/\kappa)I$$

Figure 5: A nematic liquid crystal in the standing wave of a linearly polarized laser.




Here $d$ is the sample thickness, $\kappa$ is the thermal conductivity and I ( W/cm$^2$ ) is the laser intensity. Since, $\kappa \sim 10^{-3}$ W/°C we find that in a 100 micrometer thick sample with an optical absorption coefficient $\alpha \sim 0.1$ /cm, the temperature change is about 10 K for a laser intensity of 1000 W/cm$^2$ and the attenuation of the laser intensity in the medium is about 0.1% which is negligible. For all practical purposes, the optics of the medium is literally like that of a transparent medium but at a higher temperature. It is in this approximation that we consider the effects of laser absorption. The rise in temperature is proportional to the laser intensity and the change in refractive index is proportional to the change in temperature. Thus the change in refractive index will be proportional to the laser intensity.

$$\Delta\mu = \eta' I$$

This nonlinear effect is called thermal indexing. The constant $\eta'$ is the nonlinear coefficient. It can be positive or negative.

2.3.1. *Giant nonlinear coefficient*

The same process operates even in a liquid crystal. Increase in the temperature of a nematic liquid crystal decreases its orientational order parameter. In a nematic liquid crystal, absorption of a linearly polarized laser with its electric vector parallel to the director leads to a heating of the sample. As a consequence the order parameter $S$ will decrease and as a result there will be a decrease in the refractive index parallel to the director. Thus the nonlinear optical coefficient $\eta'$ will be negative[5].

If the laser is polarized perpendicular to the director, then a torque will act on the director which will align it along the laser field. However, in view of what has been stated in the first section, we can have a magnetic field strong enough to keep the director perpendicular to the electric field of the laser. Even in this case, we find heating due to laser absorption. Again this results in a decrease in the order parameter $S$. However, for this polarization there will be in an increase of refractive index. Thus the nonlinear optical coefficient $\eta'$ will be positive in this case.

As already stated, for a 100 micron thick sample at a laser intensity of 1000 W/cm$^2$, we get an increase in temperature of about 10 K. This leads to a change in the refractive index of about $10^{-2}$ which is quite large. If the nematic liquid crystal is at a temperature close to the nematic-isotropic phase transition, then even a slight increase in temperature will lead to a very large change in refractive index. The nonlinear coefficient will be very large. Thus we get giant nonlinearities near the phase transition, due to thermal indexing.

2.3.2. *Beam modulation*

In a proper discussion of the effect of the laser field, we have to also include the fact that the electric field of the laser suppresses the director fluctuations. We consider the case of the laser field being along the director. The decrease in refractive index due to laser heating is proportional to the laser intensity. It is given by:

$$\Delta\mu = -\eta' I$$

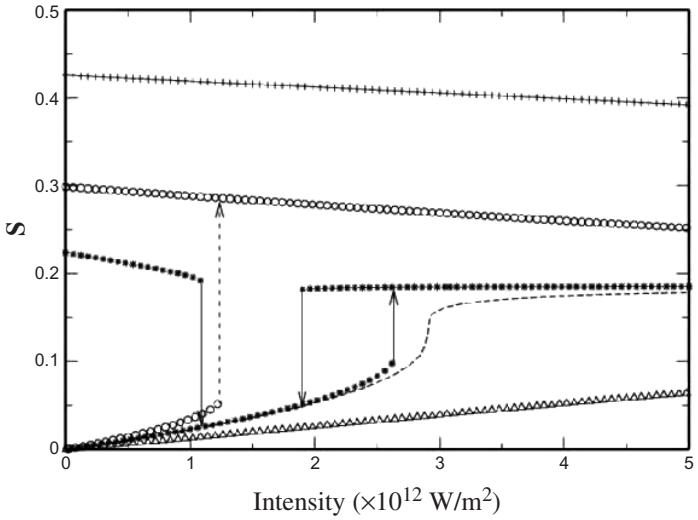
We have already stated, that the electric field of the laser suppresses the director fluctuations leading to an increase in refractive index $\Delta\mu = \eta\sqrt{I}$. Therefore due to these two processes the net change in refractive index parallel to the director is

$$\Delta\mu = -\eta' I + \eta\sqrt{I}$$

Calculations indicate[8] that, generally, $\eta$ is greater than $\eta'$. Hence, at intensities lower than $(\eta/\eta')^2$ the second term dominates and at higher laser intensities the first one becomes important. Thus the nonlinear optical coefficient changes sign as the laser intensity is increased.

This change in the sign of the nonlinear coefficient leads to beam modulation inside the sample[8]. We consider a parallel beam of laser with a Gaussian intensity profile. It will have maximum intensity at the centre of the beam and the intensity tapers off gradually as we move outward. When the nonlinear coefficient is positive then the local change in refractive index is not proportional to the local laser intensity. Yet, the medium will have a higher refractive index at the centre of the beam

Figure 6: Phase transition in an absorbing nematic liquid crystal in a laser field.




relative to its edges. Therefore, the medium acts as a convex lens and the beam will converge. On the other hand, when the nonlinear coefficient is negative the beam diverges.

In view of the above effect, in thin enough samples of nematic liquid crystals, at intensities less than $(\eta/\eta')^2$ the beam converges on emergence from the sample. The beam diverges on emergence at higher intensities.

If the sample is thick enough we get another interesting effect. When at the entry point the peak intensity is less than $(\eta/\eta')^2$ the beam converges on entrance. As it travels down the sample we find a continuous increase in beam intensity. At some point the intensity will exceed $(\eta/\eta')^2$ resulting in a negative nonlinear coefficient. From then on, the beam diverges till the intensity again falls below $(\eta/\eta')^2$ after which it starts converging. Thus, we end up with a modulated beam inside the sample. It repeatedly converges and diverges as it travels down the sample.

2.3.3. *Phase transitions*

Heating results in a rise in the temperature of the liquid crystal. As the temperature increases at some higher temperature the liquid crystal undergoes a phase transition. In normal nematic liquid crystals this phase transition will be to the isotropic phase. An external electric field alters the order parameter of a nematic liquid crystal and hence it affects this phase transition as well. This problem has been considered by Lelidis and Durand[9]. If the liquid crystal is optically absorbing, then the laser beam will heat the sample, leading to a change in the orientational order parameter $S$. Further, the electric field of the laser beam will also influence the order parameter. Hence, laser field induced phase transition will be quite different from the phase transition induced by mere heating or the electric field alone. We discuss this problem in this section.

Absorption of the laser in a nematic liquid crystal is characterized by not only the average absorption coefficient $\alpha$ but also by the anisotropic absorption coefficient

$$\Delta\alpha = \alpha_3 - \alpha_1$$

Here $\alpha_3$ and $\alpha_1$ are respectively the absorption coefficients parallel and perpendicular to the director. We have to consider the heating of the medium not only due to $\alpha$ but also due to $(\Delta\alpha)$. Then the rise in temperature is given by[10]

$$\delta T = (\zeta + 2\zeta' S/3)I$$
$$\zeta = (\alpha/\kappa)(d/\pi)^2 \text{ and } \zeta' = (\Delta\alpha/\kappa)(d/\pi)^2$$

Here $d$ is the sample thickness. The free energy density for the nematic-isotropic phase transition in a laser field is given by[10]

$$F_{NI} = (A/2)[T - (T* - \zeta I)]S^2 - (1/3) \times [B - A\zeta' I]S^3 + (D/4)S^4 - (\Delta\varepsilon/c)IS$$

Here A, B and D are constants and T* is the temperature up to which the isotropic state can be super cooled, in the absence of the laser field. The first two terms contain the effects due to laser heating arising from laser absorption. Last term represents the influence of the electric field of the laser. By minimizing $F_{NI}$ we can get the order parameter $S$ at different laser intensities $I$. From this we can work out the phase transitions that are possible. We show in figure 6 one of the results obtained for a certain set of reasonable parameters[10].

We briefly summarize the interesting results. As the system is absorbing we find, as is to be expected, that the order parameter $S$ in the nematic phase decreases with increase in laser intensity. This process is described by the (+++) line. We see that, at any finite intensity, the isotropic phase has a small but non vanishing value for $S$. This is due to the laser electric field induced orientational order in the nematic phase. Such an isotropic phase with a weak nematic order is also called a paranematic phase. As we increase the intensity, in the paranematic phase, the orientational order increases and at a certain intensity the system undergoes a transition to the usual nematic state. At this intensity there

will be a finite jump in the order parameter *S*. After such a transition, if we lower the intensity from the nematic state we get stuck in the nematic phase without getting back into the paranematic phase. The curve (ooo) represents this process. There is also an interesting phase transition indicated by the curve (***). Here if we start from the nematic phase, then on increase of laser intensity there is a jump to the paranematic state characterized by a much lower order parameter. The paranematic state on further increase of laser intensity goes back to the nematic state. This is laser induced reentrance of the nematic phase. Interestingly, on decrease of laser intensity, the nematic goes to the paranematic phase but remains in that state without a transition back to the nematic phase we started with. Thus there is reentrance only on the increase of intensity but not on its decrease.

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