

# Chapter 6

## The effects of a laser field on phase transitions in liquid crystals

### 6.1 Introduction

In the previous chapter, we considered the effect of laser absorption in cholesteric liquid crystals. Generally, for observing nonlinear effects due to electronic processes, often one needs to employ very high laser intensities. On such occasions, the electric field of the laser beam also plays a vital role. This is all the more important near a phase transition. We address to such situations in the present chapter. Here, we have studied the effects of light absorption as well as that of electric field of laser beam on phase transitions in liquid crystals. Phase transitions in liquid crystals, in an external electric or magnetic field is a well studied area of research. The effects of an external electric field on phase transitions from one liquid crystal phase to another or from a liquid crystal to an isotropic phase have been considered in detail in the literature [1–7]. Similar effects can also be expected when a laser beam is propagating through the liquid crystal. One main advantage in the latter case is the absence of ionic currents which invariably accompany static electric fields. Such ionic currents lead to many complex hydrodynamical motions. But, to get a reasonable electric field effect with a laser beam, one will have to employ high power lasers since the dielectric anisotropy of the medium is rather low at optical frequencies. An additional feature is possible if the medium is absorbing. Absorption of the laser beam leads to heat generation which increases the temperature of the medium. It is known that in liquid crystals the absorption

is anisotropic. Generally, the light beam polarized along the director is absorbed more than the one which is polarized perpendicular to the director. Phase transitions in nematic liquid crystals due to light absorption induced heating have been discussed in the literature [8,9]. In some situations this leads to self transparency [10] and optical bistabilities [11]. Even if the absorption is low the temperature raise can be considerable if the intensities are high enough. We have considered here the molecular alignment due to the electric field of the laser beam, as well as the heating of medium due to the absorption. These two effects together can lead to some new and interesting phase transitions in liquid crystals, which appear to have not been stressed in literature. We discuss in this chapter the laser field induced phase transitions, by generalizing the Landau-deGennes (LdG) theory [12].

In the nematic phase of a liquid crystal, molecules have only orientational order and no positional order. Nematic to isotropic (N-I) transition in an electric field was first observed by Hanus [13]. Later on, Lelidis and Durand [6] also studied it theoretically and experimentally. In the field free case, it is a weakly first order phase transition with the nematic phase going to an isotropic phase at a definite temperature. In the presence of an electric field, however, there is a paranematic phase in the place of the isotropic phase because of the field induced molecular ordering. In materials with positive dielectric anisotropy, the paranematic phase is exactly like a nematic phase but with a much smaller orientational order. Thus, both the nematic and the paranematic phases have the same symmetry. Lelidis and Durand showed that this phase transition is analogous to a liquid-gas phase transition in the temperature-pressure plane. Here, we consider the N-I phase transition in the presence of a linearly polarized laser beam with its electric vector parallel to the director. When there is no laser beam absorption, we find that a paranematic phase can go to a nematic phase *via* a first order transition on increase of intensity. While on decrease of intensity, it remains in the nematic phase. This is similar to a “monotropic transition” found in some liquid crystals where a particular phase transition is seen on cooling, but it is absent on heating the sample. In the presence of laser absorption, as expected, the order parameter in nematic phase decreases with increase in laser intensity. Interestingly, in this case we find in addition

to the previous one way transition, a one way transition from a nematic to a paranematic phase on increase of intensity. At some initial sample temperatures we get, laser induced nematic→paranematic→nematic transition while at some other initial sample temperatures we get a paranematic→nematic→paranematic transition on increase of intensity. In other words, we find a one way reentrant nematic or a reentrant paranematic phase *via* a paranematic or a nematic, respectively [14].

Smectic A liquid crystals exhibit both orientational and one dimensional positional order. The molecules are stacked in layers which are perpendicular to the direction of preferred molecular alignment. Smectic A to nematic (A-N) transition is often second order. As was first shown by de Gennes, it can become first order due to coupling between layering order and orientational order [12]. The effect of an external electric field on this phase transition has been considered by Lelidis and Durand [15]. They predict a field induced “parasmectic” and a “nonspontaneous nematic” phase. In a parasmectic phase, there is a very small layering order induced because of strong electric field under certain conditions. A material which undergoes an isotropic to smectic A transition, in the absence of field, can have a nematic phase when a field is applied. This is termed as a nonspontaneous nematic phase which disappears above a critical point. We consider here the effect of a laser field on this transition. In the absence of absorption we can expect the laser field effects to be identical to that of an external static electric field. On the other hand, laser light absorption and absorption anisotropy of the medium not only increases the temperature of the system but also leads to a coupling between orientational and layering orders. As a result of this effect we find a tricritical point at which a second order A-N transition becomes first order. Another interesting feature is that if an A-N transition is first order due to deGennes coupling then, at a particular laser intensity it becomes second order. Above this intensity it becomes a first order transition [14].

The A-N transition can also become first order due to a coupling of thermal fluctuations in the nematic director to the smectic order parameter [16, 17]. In fact, director fluctuations get suppressed in an external electric field [18, 19] or in the field of an appropriately polarized

laser beam [20]. Hence, we have also briefly described the effect of a laser beam on the A-N transition in the presence of director fluctuations.

In a smectic C phase, the molecules are stacked in layers as in smectic A but they are tilted uniformly in a particular direction with respect to the layer normal. Smectic C to smectic A (C-A) transition is generally second order [12]. It has been studied in various systems by Huang and Lien [21]. Effect of external fields on C-A transition has been considered by Sakamoto *et al.* [2] and Pikin [5]. They have predicted a change in the transition temperature under the influence of the field. We find that second order C-A transition becomes a first order transition in the presence of a laser beam. In a particular geometry there can be an elevation or depression of the C-A transition temperature depending upon the relative strengths of absorption coefficient and dielectric anisotropy [14].

In a cholesteric liquid crystal the constituent molecules lack mirror symmetry and the structure has a spontaneous twist. The same structure is obtained when a nematic is doped with chiral molecules. A light beam linearly polarized along the director and propagating along the twist axis, is a permitted mode for wavelengths much smaller than the pitch of the structure. This is often referred to as the Mauguin regime [12]. For cholesterics in this geometry when the laser intensity is increased, it exhibits a transition to the isotropic phase. At very high intensities there is a distinct possibility of a gradual transformation of the isotropic phase to a parcholesteric phase, which is like a cholesteric but with a smaller orientational order.

## 6.2 Nematic - isotropic transition

In the case of any first order transition, supercooling of the high temperature phase is possible down to a temperature  $T^*$  and superheating of the low temperature phase is possible up to a temperature  $T^{**}$ . In between these two temperatures one can get both the high and low temperature phases. Of these one is metastable and the other is stable. In the case of N-I transition also a similar behavior is found. The free energy has two minima between  $T^*$  and  $T^{**}$ . One corresponds to the nematic state and the other corresponds to the isotropic state.

The N-I transition temperature ( $T_{NI}$ ) is in between  $T^*$  and  $T^{**}$ . This is the temperature at which the free energies of the isotropic and the nematic phases are equal. We concentrate on the temperature range between  $T^*$  and  $T^{**}$ . Since, in practice this region is very small (about 1 to 2°C), we have to consider only a weak absorption. Further, we consider the situation when there is no torque, due to the electric field of the laser beam, on the director. This is possible when the laser beam is linearly polarized with its electric vector along the director. Landau-deGennes free energy density for N-I transition in such a geometry which is depicted in Fig. 6.1, is given by:

$$F = \frac{A_1}{2}(T - T^*)S^2 - \frac{B_1}{3}S^3 + \frac{C_1}{4}S^4 - \frac{\epsilon_a}{c}IS \quad (6.1)$$

Here,  $\epsilon_a$  is the optical dielectric anisotropy and  $I$  is the laser intensity,  $S$  is nematic orientational order parameter,  $c$  is velocity of light,  $T$  is the temperature and  $A_1, B_1, C_1$  are positive constants.

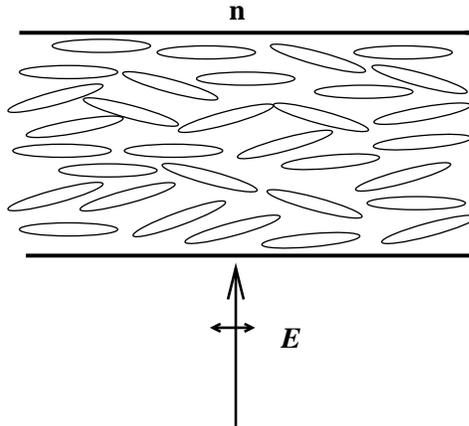


Figure 6.1: The geometry for (N-I) phase transition. The molecules have positive dielectric anisotropy.

One important fact to be noted is that, in view of the last term in Eqn. (6.1) a true isotropic phase does not exist in the presence of a laser beam. The electric field of the laser beam always leads to some molecular alignment *i.e.* non vanishing  $S$  in the isotropic phase. Such a state is referred to as a paranematic phase. It is exactly like a nematic but with a weak orientational order induced by the electric field. The equilibrium condition  $(\partial F/\partial S) = 0$

gives,

$$A_1(T - T^*)S - B_1S^2 + C_1S^3 - \frac{\epsilon_a}{c}I = 0 \quad (6.2)$$

There exists a critical point at  $\partial^2 F/\partial S^2 = \partial^3 F/\partial S^3 = 0$ . The critical order parameter ( $S_c$ ), critical field intensity ( $I_c$ ) and critical temperature ( $T_c$ ) are given by

$$S_c = B_1/3C_1 \quad (6.3)$$

$$I_c = B_1S_c^2c/3\epsilon_a \quad (6.4)$$

$$T_c = T^* + B_1^2/A_1C_1 \quad (6.5)$$

This is like a liquid gas critical point. Beyond this point, the two phases can not be distinguished. Below this critical point there is a discontinuous *i.e.* first order transition from the nematic to the paranematic phase and *vice-versa*.

In addition, if there is a small absorption of the laser light, then there would be a rise in temperature of the medium. This rise in temperature (in the wide beam approximation) can be obtained by solving heat conductivity equation as described in the previous chapter (see Sec. 5.2.1) [22, 23]. In liquid crystals because of orientational order  $S$  we get,

$$\beta = \bar{\beta} + \frac{2}{3}\beta_a S \quad (6.6)$$

Here,

$$\bar{\beta} = \frac{\beta_{\parallel} + 2\beta_{\perp}}{3}, \beta_a = \beta_{\parallel} - \beta_{\perp} \quad (6.7)$$

$\beta_{\parallel}$  and  $\beta_{\perp}$  are the optical absorption coefficients, of a fully orientationally ordered ( $S = 1$ ) nematic, for light polarized parallel and perpendicular to the director respectively. The rise in temperature ( $\delta T$ ) can also be written as:

$$\delta T = \left( \bar{\alpha} + \frac{2}{3}\alpha_a S \right) I \quad (6.8)$$

Where,  $\bar{\alpha}$  and  $\alpha_a$  are related to the average optical absorption coefficient ( $\bar{\beta}$ ) and the optical absorption anisotropy ( $\beta_a$ ) as follows:

$$\bar{\alpha} = \left( \frac{d}{\pi} \right)^2 \frac{\bar{\beta}}{\kappa} \quad \alpha_a = \left( \frac{d}{\pi} \right)^2 \frac{\beta_a}{\kappa} \quad (6.9)$$

where,  $d$  is the sample thickness and  $\kappa$  is the thermal conductivity. The total temperature is now  $T_0 + \delta T$ , where  $T_0$  is the sample temperature when there is no laser illumination. The free energy density in such absorbing nematics is given by

$$F = \frac{A_1}{2}[T_0 - (T^* - \bar{\alpha}I)]S^2 - \frac{1}{3}(B_1 - \alpha_a I A_1)S^3 + \frac{C_1}{4}S^4 - \frac{\epsilon_a}{c}IS \quad (6.10)$$

This equation results from Eqn. (6.1) and Eqn. (6.8) and describes an isothermal state because the thermal conductivity equation has been solved for steady state. In this case the critical point,  $S'_c$ ,  $I'_c$  and  $T'_c$  are given by:

$$S'_c = (B_1 - \alpha_a I'_c A_1)/3C_1 \quad (6.11)$$

$$I'_c = (B_1 - \alpha_a I'_c A_1)S'^2_c / 3\epsilon_a \quad (6.12)$$

$$T'_c = T^* - \bar{\alpha}I'_c + (B_1 - \alpha_a I'_c A_1)^2 / A_1 C_1 \quad (6.13)$$

Minimization of total free energy gives us the value of order parameter ( $S$ ) as a function of laser intensity ( $I$ ) and initial temperature ( $T_0$ ). Both these parameters can be varied easily in an experiment.

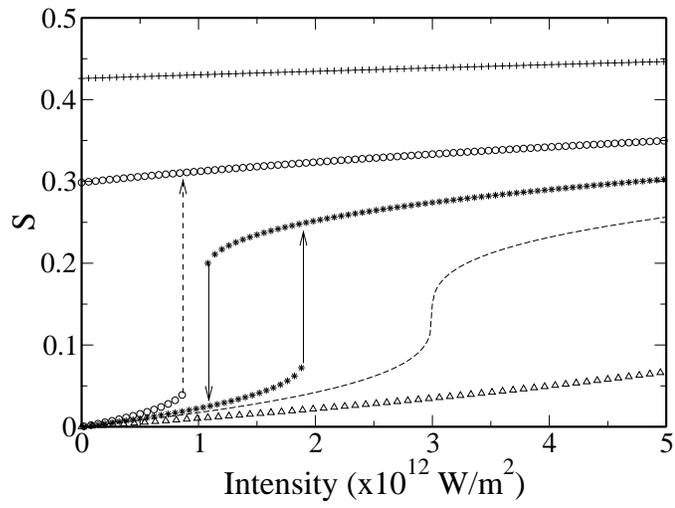
We present here the results of our calculations. We have taken the values of  $T^* = 50^\circ C$ ,  $\kappa = 10^{-3} W/cmK$ ,  $d = 10\mu m$  and  $A_1$ ,  $B_1$  and  $C_1$  from the literature [24], . Further, we should point out that in our calculations the absorption coefficient is taken to be small ( $10^{-5} cm^{-1}$ ). On the other hand, if we take a larger absorption coefficient then in order to get only a few degrees raise in temperature we have to use low intensities. But at low intensities the last term of free energy  $F$  will be negligible. We work in the limit of weak absorption and high laser intensities. Such a situation of very weak absorption can arise even in the so called transparent materials.

From Eqn. (6.10) it is clear that at large intensities the cubic term in  $S$  can change sign. This positive coefficient for cubic term, can lead to a negative order parameter. However, the linear term in  $S$  favors a positive order parameter. In such a case, one has to carefully work out the energetics. We find that this situation will not occur at presently attainable laser intensities. Hence, we have not considered such a case.

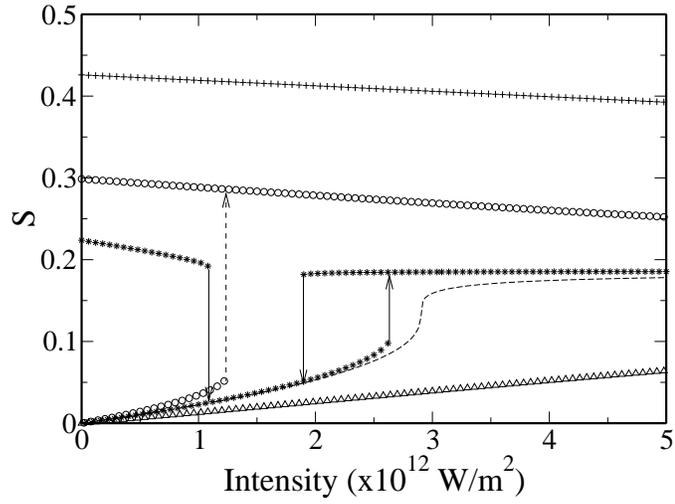
Before we discuss the results of this theory, we would like to make a remark. It is experimentally difficult to measure the total temperature ( $T$ ) of the laser illuminated sample since the temperature probe interferes with the laser beam. So, we consider a realistic situation where, we have a large enough sample in an ambient temperature controller. And only a small portion of it is illuminated by a laser beam. The temperature ( $T_0$ ) of the unilluminated part can be measured easily.

### 6.2.1 Order parameter dependence on intensity

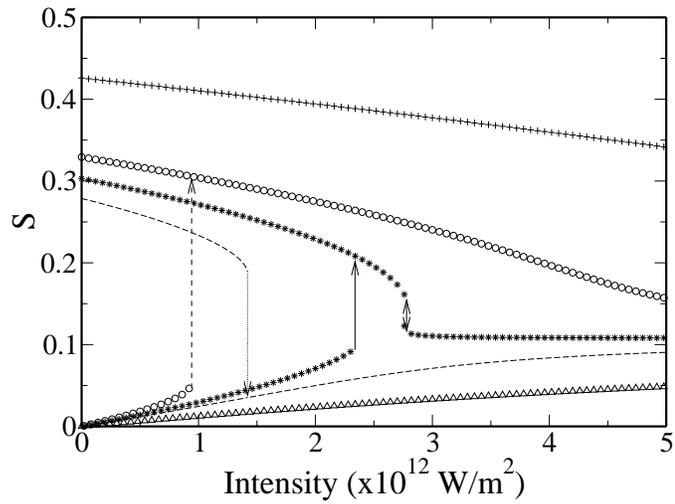
The variation of orientational order parameter ( $S$ ) with laser intensity ( $I$ ), at different initial sample temperatures ( $T_0$ ) is shown in Fig. 6.2. If the medium is not absorbing then on increase of the intensity the order parameter always increases both in nematic as well as in paranematic phase as shown in Fig. 6.2(a). But, when the medium is absorbing, then on increase of intensity the order parameter decreases in the nematic phase but increases in paranematic phase as shown in Figs. 6.2(b)-6.2(c). Generally, the laser field induces an orientational order in the isotropic phase which increases on increasing the intensity. However, for some initial temperatures ( $T_0$ ) we find that instead of a monotonic increase, a sudden jump in the order parameter at a particular intensity. Interestingly, the order of the medium is retained even on decreasing the intensity to a zero value. Thus, we get a one way transition from a paranematic to a nematic as indicated by dashed arrow in Fig. 6.2(a). If the system is absorbing, we find that the order parameter in the nematic phase decreases with increase in intensity as shown in Figs. 6.2(b)-6.2(c). This is due to the heating of the medium because of laser light absorption. In this case it is possible to get a one way transition from a nematic to a paranematic (Fig. 6.2(c)). There is also an interesting phase transition for  $\bar{\beta} = 1.3 \times 10^{-5} cm^{-1}$ ,  $\beta_a = 0.43 \times 10^{-5} cm^{-1}$  as indicated by (\*\*) in the Fig. 6.2(b). Here, if we start with a nematic phase, then, on increase of laser intensity there is a jump from the nematic to the paranematic phase which changes to the nematic on further increase of the intensity. This is like a reentrant nematic phase *via* a paranematic phase. But, the reentrance does not take place on decrease of the field. Similarly, there



(a)



(b)



(c)

Figure 6.2: The variation of the orientational order parameter ( $S$ ) with the laser intensity at various temperatures ( $T_0$ ) of the sample; ( $\Delta\Delta$ )  $T_0 > T^{**}$ ; ( $--$ )  $T_0 = T_c$ ; ( $**$ )  $T^* < T_0 < T_c$ ; ( $\circ\circ$ )  $T_{NI} > T_0 > T^*$ ; ( $++$ )  $T_0 < T^*$ ; (a)  $\bar{\beta} = \beta_a = 0$ ; (b)  $\bar{\beta} = 1.3 \times 10^{-5} \text{ cm}^{-1}$ ,  $\beta_a = 0.43 \times 10^{-5} \text{ cm}^{-1}$ ; (c)  $\bar{\beta} = 2.3 \times 10^{-5} \text{ cm}^{-1}$ ,  $\beta_a = 0.77 \times 10^{-5} \text{ cm}^{-1}$ .

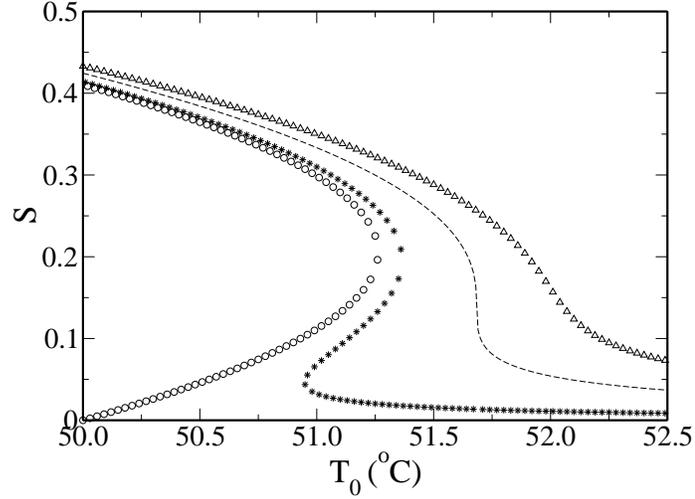
can be a paranematic→nematic→paranematic transition on increase of laser intensity for  $\bar{\beta} = 2.3 \times 10^{-5} \text{cm}^{-1}, \beta_a = 0.77 \times 10^{-5} \text{cm}^{-1}$  as indicated by (\*\*) in Fig. 6.2(c). Reentrance is not seen on decrease of intensity.

In order to realize experimentally these phase transitions, even at low laser intensities, one can use a low frequency electric field parallel to the director to augment the laser field contribution to the linear term in  $S$ . For example, in a low frequency field of the order of  $10^5 \text{V/cm}$ , the laser intensity required to observe similar phase transitions, decreases by an order of magnitude. Also, one has to start with a particular initial temperature of the sample and then increase the laser intensity. At each value of laser intensity one has to wait long enough to attain thermal equilibrium.

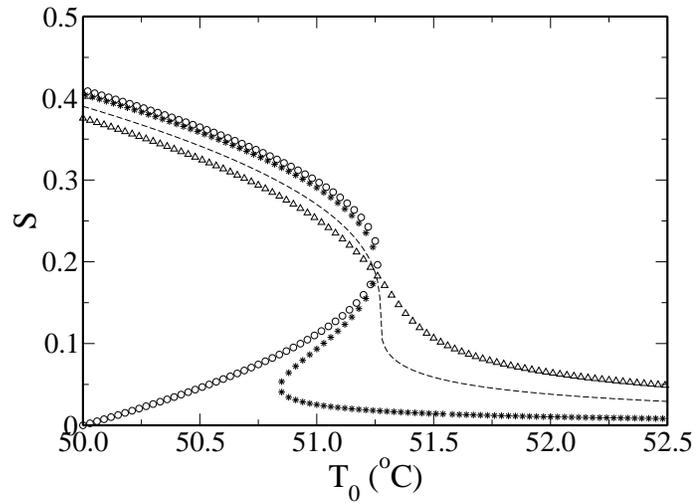
### 6.2.2 Order parameter dependence on temperature

Dependence of the orientational order ( $S$ ) on  $T_0$ , at different values of laser intensities are depicted in Fig. 6.3. As a reference we have shown in Fig. 6.3(a) the case when there is no laser absorption. Here, it is clear that below a critical intensity ( $I_c$ ) there is a first order transition from a nematic to a paranematic phase and *vice-versa*. Above  $I_c$ , there is a continuous transition from a nematic to a paranematic phase. In the case of absorbing nematics, as can be seen in Fig. 6.3(b)-6.3(c) all curves intersect at a single point. It should be noted here that if we consider the total temperature ( $T$ ) of the sample then these curves do not intersect. By subtracting out the extra temperature rise ( $\delta T$ ) due to laser heating we find an interesting result. The curves for various laser intensities intersect at a single point. As the absorption coefficient increases, the intersecting point moves down along the  $S$  versus  $T$  curve of typical first order N-I transition without any external field. We find that the critical intensity and critical temperature decreases slightly at relatively high absorption. In the region before the cross over point, the  $S^2$  term of free energy density dominates and in the region after the cross over point the linear term in  $S$  dominates. The cross over point which represents a switch over from one region to the other, is given by:

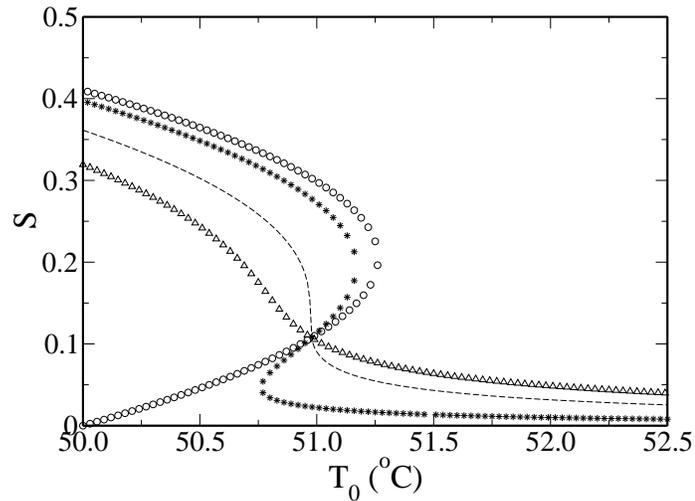
$$S_{cross} = \frac{\epsilon_a}{cA_1\bar{\alpha}} \quad (6.14)$$



(a)



(b)



(c)

Figure 6.3: The variation of the orientational order parameter ( $S$ ) with  $T_0$  the temperature of unilluminated sample at various intensities of laser beam: (a) no absorption, (b) and (c) for absorbing systems. ( $\Delta\Delta$ )  $I > I_c$ ; (- -)  $I = I_c$ ; (\*\*)  $I < I_c$ ; ( $\circ\circ$ )  $I = 0$ . The values of average absorption coefficient and absorption anisotropy are same as that given in Fig. 6.2.

### 6.3 Smectic A - nematic transition

Smectic liquid crystals have a layering order of molecules in addition to orientational order. In smectic A the molecules are preferentially aligned along the layer normal. Majority of smectic A liquid crystals go through a nematic phase before going to an isotropic phase. It is well known that if these two order parameters are weakly coupled, then A-N transition is a second order phase transition. However, the A-N transition can become first order if the coupling between orientational order and layering order is strong [12]. We find a new kind of coupling between nematic orientational order and smectic layering order arising due to absorption anisotropy. The smectic layer order parameter is denoted by  $\psi$  and the free energy density for A-N transition without the usual de Gennes ( $\psi - S$ ) coupling is given by:

$$F_A = F_0 + \frac{A_2}{2}(T - T_{AN})\psi^2 + \frac{B_2}{4}\psi^4 \quad (6.15)$$

Here  $A_2$  and  $B_2$  are positive constants. Consider the case when a linearly polarized laser beam is propagating through the medium such that its electric vector is polarized parallel to the director, as shown in the Fig. 6.4. In such a situation there is no torque on the director due to the electric field of the laser beam. The increase in temperature ( $\delta T$ ) of the medium due to light absorption is given by Eqn. (6.8). Then, Eqn. (6.15) becomes,

$$F_A = F_0 + \frac{A_2}{2} [T_0 - (T_{AN} - \bar{\alpha}I)] \psi^2 + \frac{B_2}{4} \psi^4 - \left(\frac{\epsilon_a}{c}I\right) S + \left(\frac{A_2}{3}\alpha_a I\right) \psi^2 S \quad (6.16)$$

Here,  $T_0$  is the sample temperature without laser heating. The last term indicates that there is a coupling between  $\psi$  and  $S$ . We now follow deGennes [12] and find that because of this coupling there will be an extra orientational order when smectic order ( $\psi$ ) is present. We can write orientational order parameter ( $S$ ) as

$$S = S_0 + \delta S$$

where  $S_0$  is the orientational order parameter in the nematic state and  $\delta S$  is the extra orientational order. The total free energy density must also include the nematic free energy density

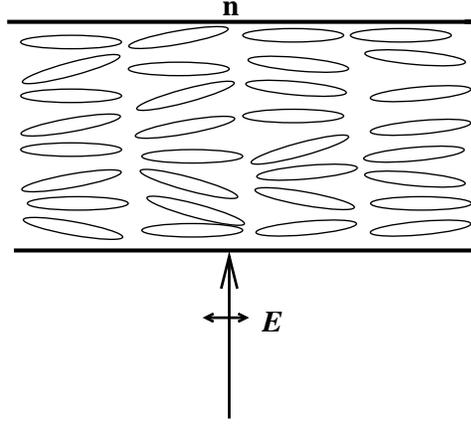


Figure 6.4: The geometry for (A-N) phase transition.

$F_N$ . Keeping in mind that  $F_N$  should be minimum for  $\delta S = 0$ , we write the nematic free energy density as:

$$F_N = F_N(S_0) + \frac{K}{2}(\delta S)^2 \quad (6.17)$$

where  $F_N(S_0)$  is the usual nematic free energy density and  $K$  is a positive constant. The minimization of total free energy, with respect to  $\delta S$  gives,

$$\delta S = -\left(\frac{A_2\alpha_a I}{3K}\right)\psi^2 + \frac{\epsilon_a I}{cK} \quad (6.18)$$

with this value of  $\delta S$  the total free energy density for A-N transition becomes,

$$F_{AN} = F_0 + F_N(S_0) - \frac{\epsilon_a}{c} I \left( S_0 + \frac{\epsilon_a I}{2cK} \right) + \frac{A_2}{2} [T_0 - (T'_{AN})] \psi^2 + \frac{B'_2}{4} \psi^4 \quad (6.19)$$

where,

$$T'_{AN} = T_{AN} - \left( \bar{\alpha} I + \frac{2}{3} \alpha_a I S_0 + \frac{2\epsilon_a \alpha_a I^2}{3cK} \right) \quad (6.20)$$

$$B'_2 = B_2 - 2(A_2\alpha_a I)^2/9K \quad (6.21)$$

The order of transition depends on the sign of  $B'_2$ . We now consider the different cases.

*Case I:* At low intensities the correction to  $B_2$  is small and  $B'_2$  will be a positive number. Then one can easily see that transition is of second order and transition temperature is  $T'_{AN}$ .

*Case II:* At high intensities  $B'_2$  can be negative. Then, to ensure stability  $\psi^6$  term should be

added. The transition will be first order.

*Case III:* At a particular intensity ( $I = \sqrt{(9KB_2/2A_2^2\alpha_a^2)}$ ),  $B'_2$  becomes zero. This is a tricritical point at which there is a change over from a second order to a first order transition. This is induced by the coupling between  $\psi$  and  $S$  due to absorption anisotropy. In the case of A-N transition which is first order because of de-Gennes coupling term ( $-C\psi^2\delta S$ ) [12], light induced coupling can exactly cancel out this term at a particular intensity [equal to  $(3C/A_2\alpha_a)$ ], as they are of opposite sign. In that case, A-N transition becomes second order at this particular laser intensity. Above and below this intensity it is a first order transition. Here, we should point out that though the sign of laser induced coupling is opposite to that of de-Gennes coupling it still leads to a negative correction in  $\psi^4$  term. This is because the correction is quadratic in the coupling coefficient.

It would be interesting to know the nature of A-N transition when the laser intensity alone is changed. To start with in the smectic phase, the coefficient of  $\psi^2$  is negative and that of  $\psi^4$  term is positive, leading to a nonzero smectic order parameter. From Eqn. (6.21) it is clear that above a certain laser intensity, the  $\psi^4$  term can become negative and therefore, for stability we should add a positive  $\psi^6$  term. With further increase of laser intensity,  $\psi^4$  term becomes more negative. This stabilizes the smectic order parameter. At the same time, the orientational order parameter ( $S$ ) decreases with increase of laser intensity and goes to zero at a certain laser intensity. Since,  $\psi$  can not exist without  $S$ , we get a smectic to isotropic transition at this laser intensity. Thus, on increase of laser intensity we get a smectic to isotropic transition. While on decreasing the laser intensity, it is possible for nematic order to set in and smectic order parameter to be zero. On further decrease of the intensity we get a smectic phase. Then we can get the familiar phase sequence isotropic→nematic→smectic on decrease of laser intensity. Thus, A-N transition can become one way when laser intensity alone is varied. The possibilities of such a one way transition are rather high when the smectic phase is very close to its tricritical point *i.e.* the coefficient  $\psi^4$  term is very small and positive to start with.

The above discussions do not include the effect of the thermal fluctuations of the director

on the A-N transition. Halperin *et al.* [16] showed that director fluctuations get coupled to smectic order  $\psi$  leading to a cubic term ( $\psi^3$ ) in free energy density. This coupling results in a first order A-N transition. If these fluctuations are suppressed in a smectic, then we can get a second order A-N transition [17–19]. The director fluctuations can be completely suppressed by subjecting the medium to an external electric or magnetic field. This has been shown in nematics [6]. A laser beam of appropriate intensity and polarization can also suppress director fluctuations [20]. Then the A-N transition becomes second order at and above such intensities. However, if the material is absorbing, the thermal heating of the medium will lead to an increase in the director fluctuations. In such cases, the associated field may not be strong enough to completely suppress the director fluctuations. Then, A-N transition will be first order.

## 6.4 Smectic C - smectic A transition

In the smectic C phase also the molecules are stacked in layers but tilted with respect to the layer normal. This tilt angle continuously goes to zero as one approaches the smectic C to smectic A phase transition. The tilt angle ( $\theta$ ) is the relevant order parameter for this transition. Free energy density for the C-A transition in the presence of a linearly polarized laser beam when its electric vector is along the layer normal (shown in Fig. 6.5), is:

$$F_{AC} = F_0 + \frac{A_3}{2}(T - T_{AC})\theta^2 + \frac{B_3}{4}\theta^4 + \frac{C_3}{6}\theta^6 - \frac{\epsilon_a}{c}I\cos^2\theta \quad (6.22)$$

Here,  $A_3$ ,  $B_3$  and  $C_3$  are positive constants. In this case, the rise in temperature of the medium due to light absorption is given by

$$\delta T = (\alpha_{\parallel} - \alpha_a \sin^2\theta) I \quad (6.23)$$

Expanding free energy density as a series in  $\theta$  up to  $\theta^6$ , we get,

$$F_{AC} = F_0 + A_3 (T_0 - T'_{AC}) \frac{\theta^2}{2} + B'_3 \frac{\theta^4}{4} + C'_3 \frac{\theta^6}{6} - \frac{\epsilon_a}{c} I \quad (6.24)$$

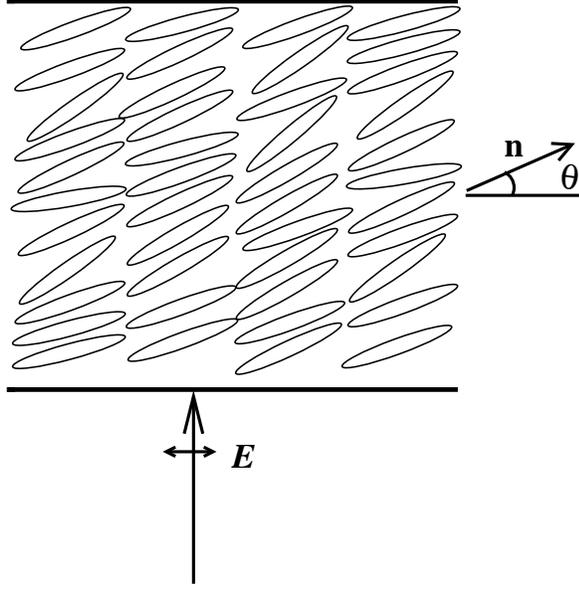


Figure 6.5: The geometry for (C-A) phase transition.

Here,

$$T'_{AC} = T_{AC} - \left( \alpha_{\parallel} + \frac{2\epsilon_a}{A_3 c} \right) I \quad (6.25)$$

$$B'_3 = B_3 - \left( 2A_3 \alpha_a + \frac{4\epsilon_a}{3c} \right) I \quad (6.26)$$

$$C'_3 = C_3 + \left( A_3 \alpha_a + \frac{4\epsilon_a}{15c} \right) I \quad (6.27)$$

We highlight some interesting consequences. There will be a laser field induced decrease in the C-A transition temperature. This is similar to the change in transition temperature due to an applied electric or magnetic field [2]. We stress here that the sign of  $B'_3$ , the coefficient of  $\theta^4$  term can change at high enough laser intensities. Then, next higher order term in  $\theta$  becomes important and the transition changes from a second order to a first order. The critical intensity at which  $B'_3$  vanishes is given by

$$I_{cr} = \frac{B_3}{(2A_3 \alpha_a + 4\epsilon_a/3c)} \quad (6.28)$$

This is a tricritical point at which C-A transition changes from second order to first order. If the laser beam propagates along the layer normal, with its electric vector in

the plane of molecular tilt, then in Eqn. (6.26) and Eqn. (6.27) intensity dependent term changes sign while in Eqn. (6.25) the coefficient of the intensity dependent term becomes  $[-(\alpha_{\perp} - 2\epsilon_a/A_3c)]$ . In this geometry the phase transition continues to be of second order at all intensities. But, the C-A transition temperature increases or decreases depending upon whether  $\alpha_{\perp}$  is lesser or greater than  $(2\epsilon_a/A_3c)$ .

## 6.5 Cholesteric - isotropic transition

As described earlier, cholesteric (Ch) liquid crystals can be considered as spontaneously twisted nematics with the twist axis normal to the nematic director. Here, we discuss qualitatively the cholesteric-isotropic (Ch-I) transition in the presence of a laser field. We consider the geometry where the light propagates along the helix axis of a cholesteric in the Mauguin's regime *i.e.* the cholesteric pitch is much larger than the wavelength of the laser light. Then, the base states are linear vibrations polarized along and perpendicular to the local director [12]. If the incident laser beam is polarized parallel to the director, then its electric vector follows the director inside the cholesteric medium. Now, if the medium gets heated due to laser light absorption, then the structure will melt at a particular laser intensity leading to an isotropic phase. In the isotropic phase, the electric field of incident laser beam will try to locally align the molecules. Such an alignment gets spontaneously twisted because of the local chirality. The pitch of this twisted structure is directly proportional to the Frank twist elastic constant  $K_{22}$  which in turn varies as  $S^2$ . Thus, the pitch will be very small since the field induced orientational order parameter ( $S$ ) is small. Then the pitch may not be very large enough compared to wavelength of light. In such case the cholesteric will not be in the Mauguin's regime. Then, spontaneously twisted structure cannot be sustained by the electric field of the laser beam, as it will not be parallel to the local director everywhere. Hence, we get only an isotropic phase. If we keep increasing the intensity of laser beam then at high enough intensities, the field induced orientational order in isotropic phase can become large enough. Then the resulting spontaneously twisted structure can have a pitch, large enough to be in the Mauguin's regime. Under these circumstances, the laser field can propagate with

its electric field along the local director. The electric field of the laser beam can sustain the twisted structure and we get a “paracholesteric” phase induced by the laser beam. Thus, with increase of intensity, we expect a cholesteric to isotropic transition, followed at much higher intensities by a paracholesteric phase. The transition from the isotropic phase to paracholesteric phase can only be gradual without a well defined transition point since there is no definitive pitch for the onset of Mauguin’s regime.

Finally, in this chapter, we have discussed many interesting phase transitions in liquid crystals in the presence of a linearly polarized light beam. There are also other possibilities, for example, in the N-I transition, as we describe, the cubic term in order parameter can change sign at some very high value of laser intensity. In that case, the order parameter will tend to have a negative value. An analysis on energetics and stable solution of free energy may lead to some really interesting situations. Also, in the case of smectic phase transitions, it will be useful to construct the phase diagram with three parameters, *viz.*, laser intensity, order parameter and temperature.

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