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OPTICAL SOLITONS IN LIQUID CRYSTALS

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I. INTRODUCTION

Liquid crystal^{1,2} is a state of matter that is intermediate between the liquid state and the crystalline state. The molecules of liquid crystals can be rod-like, disc-like or bowl-like.³ In the crystal state, the molecules are arranged in a periodic structure, and there is long range order of the orientation of the molecules. In the liquid state, there are no translational and orientational orders, and the molecules can move rather freely. Liquid crystals are not as ordered as crystals, but, also not as disordered as liquids. For example, in some temperature range, the rod-like molecules of certain organic compounds tend to align preferably along one direction, commonly labelled by a unit vector n , the so-called director, while their positions are in disorder. These anisotropic liquids are called nematic liquid crystals. Another example is the smectic liquid crystals with one-dimensional order. The system can be viewed as a set of two-dimensional liquid layers stack on top of each other.

Liquid crystal is a highly nonlinear optical medium.⁴ This is because of the presence of π electrons which are extremely susceptible to electromagnetic fields. In addition to nonlinear optics, liquid crystal research is important in various areas in physics, chemistry and biology. Furthermore, there are numerous industrial applications for liquid crystals, e.g. in liquid crystal displays (LCD's) and sensors. At present, the market size

for these various applications is about half billion dollars and is growing rapidly.

In this paper, we will discuss theoretically the possible existence of optical solitons in the isotropic liquid and in the nematic phases. For the same compound, when heated, the nematic phase will go through a first order transition at temperature T_C to the isotropic liquid phase. As temperature increases from below T_C , the orientation order parameter, Q , decreases, drops to zero abruptly at T_C and remains zero for $T > T_C$.

II. ISOTROPIC LIQUID PHASE

In the isotropic liquid phase, the medium is isotropic. In the presence of an electric field \mathbf{E} (in the \hat{x} direction, say), dipole \mathbf{p} is induced in the molecules. Due to the torque, $\mathbf{p} \times \mathbf{E}$, the molecules reorient and redistribute themselves. Near T_C , fluctuation of the order parameter Q is very large, resulting in large optical nonlinearity.

The induced dipole is given by $\mathbf{p} = \overleftrightarrow{\chi} \mathbf{E}$, where $\overleftrightarrow{\chi}$ is the macroscopic susceptibility tensor. Specifically

$$\begin{aligned} \chi_{yy} = \chi_{zz} &= \bar{\chi} - \frac{1}{3} \chi_a Q, \\ \chi_{xx} &= \bar{\chi} + \frac{2}{3} \chi_a Q, \end{aligned} \quad (2.1)$$

where $\bar{\chi}$ is the average linear susceptibility $[(1/3)(\chi_{xx} + \chi_{yy} + \chi_{zz})]$ and χ_a is the anisotropic part.

According to the Landau-de Gennes theory, the free energy can be expanded as

$$\begin{aligned} F = F_0 + \frac{1}{2} a(T-T^*)Q^2 + \frac{1}{3} BQ^3 + \dots \\ - 2\left(\bar{\chi} + \frac{2}{3}\chi_a Q\right) |E(\omega)|^2, \end{aligned} \quad (2.2)$$

where T^* is the supercooling temperature, a and B are constants. Near T_C , $Q \ll 1$, hence Q^3 and higher order terms can be dropped from (2.2). We then have

$$\frac{\partial F}{\partial Q} = 0 = a(T-T^*)Q - \frac{4}{3} \chi_a |E(\omega)|^2. \quad (2.3)$$

This implies

$$Q = \frac{4}{3} \frac{\chi_a}{a} \frac{|E(\omega)|^2}{T-T^*} \sim n_2 |E|^2, \quad (2.4)$$

where n_2 is the nonlinear term in the refractive index defined by $n = n_0 + n_2 E^2$, n is the refractive index of an isotropic medium, and n_0 is the linear term.

For thermotropic liquid crystals, e.g., M3BA, one may have $T-T^* \sim 1K$, and so $n_2 \sim 10^{-9}$ esu. This is much larger than the case in ordinary liquids for which $n_2 \sim 10^{-12}$ to 10^{-11} esu. For lyotropic liquid crystal such as C₈PFO/H₂O, cesium perfluorooctanoate in water,⁵ one could have $T-T^* \sim 2 \times 10^{-2} K$. And a could be twenty times smaller than that in thermotropics (at least in the DACl/NH₄Cl/H₂O system⁶). We then expect $n_2 \sim 10^{-6}$ esu (or $\sim 10^{-13}$ mks). This is to contrast with a much smaller $n_2 \sim 10^{-22}$ mks in SiO₂.

By the same mechanism as invoked in ordinary isotropic medium,⁷ one expects optical solitons to occur in the isotropic liquid phase of liquid crystals. The only difference is that in the latter, n_2 is much larger and the effect should be much pronounced. In fact, as given in Ref. 7, the distance over which the effect of nonlinearity appears, i.e. the distance for solitons to form,

$$L_{NL} \sim \frac{v_g \epsilon_0 n_0^2}{2 \pi n_2} \left(\frac{\lambda S}{P_0} \right), \quad (2.5)$$

where λ is the wavelength, S is the cross sectional area of the beam, P_0 and v_g are the peak power and group velocity of the wave, respectively. For SiO₂, if $\lambda = 1.3 \mu m$, $P_0 = 10$ mW, $L_{NL} \sim 14$ km. Instead, if liquid crystal is used, $L_{NL} \sim 1 \mu m$. That is, the use of a very thin liquid crystal cell is sufficient to produce a soliton from an initial wave profile.

III. NEMATIC PHASE - THERMAL EFFECT

Consider a homeotropically aligned (director n perpendicular to the container walls) nematic liquid crystal cell of thickness d , in the presence of an electric field E which is perpendicular to n . Define $\epsilon_a = \epsilon_{\parallel} - \epsilon_{\perp}$, where ϵ_{\parallel} , ϵ_{\perp} are the parallel and perpendicular components of the dielectric tensor. If $\epsilon_a > 0$, and $E < E_{th}$, a certain threshold value, the molecules will remain

undisturbed due to the boundary effects. For $E > E_{th}$, the molecules in the bulk will start to rotate and tend to align in parallel to \mathbf{E} . This is analogous to a second order phase transition, and is called the Frederiks transition.^{1,2}

Below the Frederiks transition, $E < E_{th}$, the medium is a uniform uniaxial material. Nonlinear optical properties still exist because of the temperature dependence of ϵ_{\parallel} and ϵ_{\perp} (Fig. 1). In this case, $n_2 \cdot d\epsilon_{\perp}/dT > 0$.⁸ Because of this thermal effect, we still have $n = n_0 + n_2 E^2$ and the nonlinear Schrödinger (NLS) equation, resulting in optical solitons. Note that n_2 in this case is much smaller than that in the isotropic liquid case discussed in Sec. II.

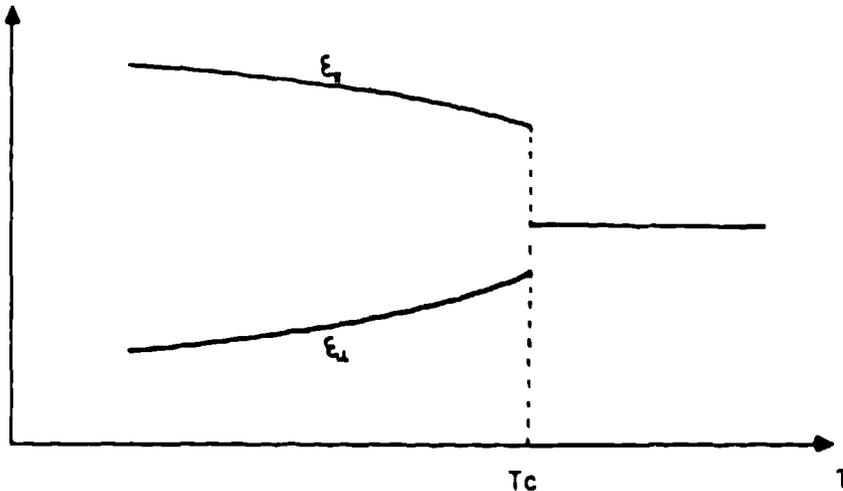


Fig. 1. Typical ϵ_{\parallel} and ϵ_{\perp} as functions of T .

IV. NEMATIC PHASE - REORIENTATION EFFECT

We now consider the case of $E > E_{th}$ (i.e., above the Frederiks transition) in the nematic phase where the dielectric constant, ϵ_{ij} , is given by

$$\epsilon_{ij} = \epsilon_{\perp} \delta_{ij} + \epsilon_a n_i n_j, \quad (4.1)$$

where $\mathbf{n} = (n_1, n_2, n_3) = (\sin\theta, 0, \cos\theta)$. Equivalently

$$(\epsilon_{ij}) = \begin{pmatrix} \epsilon_{\perp} + \epsilon_a \sin^2 \theta & 0 & \frac{1}{2} \epsilon_a \sin 2\theta \\ 0 & \epsilon_{\perp} & 0 \\ \frac{1}{2} \epsilon_a \sin 2\theta & 0 & \epsilon_{\perp} + \epsilon_a \cos^2 \theta \end{pmatrix}. \quad (4.2)$$

Above the Frederiks transition, reorientation of molecules occur. If the cell is thick enough, we can ignore the boundary effects and assume θ to be independent of z . Specifically,

$$\theta - \theta_m = 2 \sqrt{\frac{|E|}{E_{th}} - 1}, \quad (4.3)$$

where θ_m is the maximum director angle occurring at the center of the cell, and $E_{th} = (\pi/d) (4\pi K/\epsilon_a)^{1/2}$.² Here, K is an elastic constant. For $E \geq E_{th}$, $\theta \ll 1$ and

$$\sin^2 \theta - \theta^2 = \frac{4}{E_{th}} (E - E_{th}) = \frac{2}{E_{th}^2} (E^2 - E_{th}^2). \quad (4.4)$$

From Maxwell equations (in cgs units),

$$\nabla \times \nabla \times \mathbf{E} = - \frac{1}{c^2} \frac{\partial^2 \mathbf{D}}{\partial t^2}, \quad (4.5)$$

where $D_i = \epsilon_{ij} E_j$. Let \hat{z} be the direction perpendicular to the glass plates of the homeotropic nematic cell, and assume a light beam linearly polarized with $\mathbf{E} = (E(y, z, t), 0, 0)$, which satisfies $\nabla \cdot \mathbf{E} = 0$ identically. From the vector identity, $\nabla \times \nabla \times \mathbf{E} = -\nabla(\nabla \cdot \mathbf{E}) - \nabla^2 \mathbf{E}$, (4.5) becomes

$$\left[\frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} (\epsilon_{\perp} + \epsilon_a \sin^2 \theta) \right] E = 0. \quad (4.6)$$

Substituting (4.4) into (4.6), we have

$$\left(\frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2} \right) E - a_1 \frac{\partial^2 E}{\partial t^2} = a_2 \frac{\partial^2}{\partial t^2} |E|^2 E, \quad (4.7)$$

where

$$a_1 = (3\epsilon_{\perp} - 2\epsilon_{\parallel})/c^2, \quad \text{and} \quad a_2 = 2\epsilon_a/(E_{th}^2 c^2). \quad (4.8)$$

Following Ref. 9, we set

$$E = A(y, z, t) e^{i(\omega t - kz)} + \text{c.c.}, \quad (4.9)$$

where $A(y, z, t)$ is a slowly varying complex amplitude. By (4.7) and (4.9), one gets

$$(a_1\omega^2 - k^2)A - 2ik \frac{\partial A}{\partial z} + 2i\omega a_1 \frac{\partial A}{\partial t} + \frac{\partial^2 A}{\partial y^2} + a_2\omega^2 |A|^2 A = 0. \quad (4.10)$$

Consider the linearized case, i.e. $A = \text{constant}$ and the nonlinear term is small, we have the linear dispersion relation, $a_1\omega^2 - k^2 = 0$. In the weakly nonlinear region, we can assume ω and k still satisfy the linear dispersion relation. Then (4.10) becomes

$$\frac{\partial^2 A}{\partial y^2} + a_2\omega^2 |A|^2 A + 2i\omega a_1 \frac{\partial A}{\partial t} - 2ik \frac{\partial A}{\partial z} = 0. \quad (4.11)$$

In the steady state, (4.11) becomes

$$\frac{\partial^2 A}{\partial y^2} + a_2\omega^2 |A|^2 A - 2ik \frac{\partial A}{\partial z} = 0, \quad (4.12)$$

which describes self-focussing. On the other hand, if A is independent of z , or a new "time" variable is introduced so that after scaling, we have

$$\frac{\partial^2 A}{\partial y^2} + a_2\omega^2 |A|^2 A + 2i\omega a_1 \frac{\partial A}{\partial t} = 0. \quad (4.13)$$

Equation (4.13) describes self-phase modulation. In both case, we have NLS equation and hence optical solitons. For example, A and z in (4.12) can be rescaled to give

$$\frac{\partial^2 A}{\partial y^2} + 2|A|^2 A - i \frac{\partial A}{\partial z} = 0, \quad (4.14)$$

which has the soliton solution⁹ given by

$$A(y, z) = \frac{2\eta \exp[4i(\xi^2 - \eta^2)z + 2i\xi y + i\phi]}{\cosh[2\eta(y - y_0) - 8\eta\xi z]}, \quad (4.15)$$

where the free parameters are the amplitude 2η , the carrier phase ϕ , the envelop speed 4ξ and the envelop phase y_0 .

V. CONCLUSION

Liquid crystals are excellent materials for studying nonlinear optical effects and solitons.¹⁰ In this paper, we show that optical solitons can be generated easily in the isotropic

liquid phase due to the large magnitude of n_2 . In the nematic phase it is possible to generate optical solitons either through the thermal effect below the Frederiks transition, or through the reorientation effect above the Frederiks transition.

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