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Theoretical approach to study the electroclinic effect very near to the Smectic C* –Smectic A* transition point of FLC molecules

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Abstract

We propose a theoretical approach considering Landau type free energy expansion in order to understand the electroclinic effect appearing very close to the transition region between Smectic-C* to Smectic-A* phases. A new secondary order parameter is considered, and expressing it as the fluctuations of the applied field very close to the transition point, the capacitive nature of the system is addressed successfully at this very region. More over we have been able to show that the modulated Smectic-C* phase may be considered to be responsible for the origin of the electroclinic behavior.

Keywords: Ferroelectric liquid crystals; Statistical physics; Dielectric relaxation

1. Introduction

Chiral liquid crystals show very interesting physical phenomena near the transition region [1]. Electroclinic effect is one of those phenomena, which can be attributed in general to the chirality of the liquid crystal molecules. There are several papers published [2-6] over the years to investigate this phenomenon in detail. We wish to highlight a few of them to emphasize the facts regarding the electroclinic effect. According to Meyer et al. [7], the application of external field may produce some net molecular tilt in the Smectic-A* phase with layer contractions, believed to be responsible for the appearance of the electroclinic behavior of the Smectic phases. These predictions are well equipped with strong experimental results [8,9]. Another approach to the electroclinic behavior was suggested by de Vries et al. [10] and according to them Smectic-A* phase initially contain some molecular tilt without any azimuthal order but due to the application of external field some azimuthal order may appear in the molecular environment without

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any layer contractions as suggested by the previous model. Meyer et al. [11] again suggested an idea to incorporate the field induced molecular tilt in Smectic-A* phase to describe the appearance of electroclinic effect very close to the transition point between Smectic-C* and Smectic-A* phases. But they proposed that the appearance of electroclinic effect in the Smectic-A* phase may not be considered as Smectic-A* phase, but it could better be a modulated Smectic-C* phase having no long-range azimuthal order in the absence of the external field and microscopically originating from a kind of defects present in the system. Thus it may closely resemble with the Smectic-A* phase. These predictions were also strongly supported by experimental observations [12]. In the present paper we extend these theoretical predictions more strongly following the de Vries approach and the concept of the modulated Smectic-C* phase i.e. disregarding the layer contractions and highlighting the fact that very close to the transition point the external field indeed induces some director tilt producing a net polarization in the medium from the theoretical point of view. By introducing a new secondary order parameter, displacement vector (D) instead of the polarization (P) of the medium, we have tried to explore new physical observations very close to the transition point as a result of appearance of the electroclinic effect. In this paper we observed the existence of the de Vries Smectic A* phase and the modulated Smectic-C* phase based on the same origin of electroclinic effect.

2. Theoretical approach and discussion

After the application of the significantly large electric field, the spatial configuration of the director in the ferroelectric phase does not change appreciably near the Smectic-C* to Smectic-A* transition point. Thus we can neglect the elastic contribution and the flexoelectric contribution to the Gibbs free energy in the vicinity of the transition point. Here we are considering the classical Landau model [13] by expressing the Gibbs free energy in terms of the primary order parameter θ (tilt angle) and the secondary order parameter P (spontaneous polarization) [14] and consider symmetry arguments to understand the electroclinic behavior of FLC molecules as proposed by Meyer et al. [15]. Again the de Vries type Smectic-A* phase, which is assumed to have some reorientations of the molecules by rotations around the Smectic cone with the electric field [16, 17] followed by some experimental evidence [18]. Since the modulated Smectic-C* phase may have some correlation with the ordinary Smectic-A* phase, we are trying to show the resemblance between the modulated Smectic-C* phase and the de Vries type Smectic-A* phase as discussed below. In the generalized Landau model, the Gibbs free energy in the presence of an oscillatory field (measuring field) of an unwound system (wave vector, q=0) for the SmC*-SmA* transition can be written as:

$$F = F_o + \frac{1}{2}a\theta^2 + \frac{1}{4}b\theta^4 + \frac{1}{6}c\theta^6 + \frac{\mu^2}{2\beta}P^2 + \frac{\lambda}{4!\beta}P^4 + \frac{\mu^2\varepsilon}{2\beta}E^2 + \frac{\lambda\varepsilon^4}{4!\beta}E^4$$
$$+ \frac{\mu^2}{\beta\varepsilon}PE + \frac{\lambda\varepsilon^2}{2\beta}P^2E^2 + \frac{\lambda\varepsilon}{6\beta}P^3E + \frac{\lambda\varepsilon^3}{6\beta}PE^3 \quad (1)$$

Here the parameters a, b, c, μ, β, and λ have the usual significance, and $\mu^2 = \alpha(T-T_0)$, where α is a constant and T_0 being the transition temperature. The free energy quite evidently contains both the primary order parameter (θ) and the secondary order parameter (P) with some coupling between P and the external field E. By visualizing the basic definition of the electric displacement vector $D=\varepsilon_0E+P$, the free energy expression now looks like the following i.e.,

$$F = F_0 + \frac{1}{2}a\theta^2 + \frac{1}{4}b\theta^4 + \frac{1}{6}c\theta^6 + \frac{\mu^2}{2\beta}D^2 + \frac{\lambda}{4!\beta}D^4 \quad (2)$$

The free energy expression is now expressed in terms of θ and the electric displacement vector D in equation (2). Since D contains both the secondary order parameter P and the external field E, we can now define a new secondary order parameter D instead of P in view of the Landau free energy expression (2). In this paper we have not only emphasized this fact but also shown the informative nature of D instead of P and thus justifying the introduction of D in our theoretical approach.

In consideration of all couplings the electroclinic free energy expression can be written as:

$$F = F_0 + \frac{1}{2}a\theta^2 + \frac{1}{4}b\theta^4 + \frac{1}{6}c\theta^6 + \frac{1}{2\chi_0}D^2 + \frac{1}{4}\eta D^4 - \gamma D\theta - \frac{1}{2}\Omega D^2\theta^2 \quad (3)$$

Most importantly, we have considered here the coupling between displacement current (D) and tilt angle (θ) both in the bilinear and biquadratic forms. χ_0 and η absorb the terms μ^2 and λ for simplifying the mathematical arguments. Therefore, to obtain a stable condition of the system the necessary minimizations were done with respect to θ and the new secondary order parameter D (instead of P). From the minimization condition with respect to D we obtain the following equation as given below:

$$\frac{\partial F}{\partial D} = 0 = \frac{D}{\chi_0} - \Omega\theta^2 D + \eta D^3 - \gamma\theta$$

$$\text{or } -\frac{1}{2}\Omega D^2\theta^2 = \frac{1}{2}\gamma D\theta - \frac{1}{2}\eta D^4 - \frac{D^2}{2\chi_0} \quad (4)$$

Putting the expression of equation (4) into equation (3) we have,

$$F = F_0 + \frac{1}{2}a\theta^2 + \frac{1}{4}b\theta^4 + \frac{1}{6}c\theta^6 - \frac{1}{4}\eta D^4 - \frac{1}{2}\gamma D\theta \tag{5}$$

Minimizing equation (5) with respect to θ we obtain

$$\frac{2a}{\gamma}\theta + \frac{2b}{\gamma}\theta^3 + \frac{2c}{\gamma}\theta^5 = D \tag{6}$$

The free energy now becomes

$$F = F_0 - \frac{1}{2}a\theta^2 - (\frac{3}{4}b + \frac{16\eta a^4}{4\gamma^4})\theta^4 - (\frac{5}{6}c + \frac{16\eta a^3 b}{\gamma^4})\theta^6 \tag{7}$$

If we consider the second order phase transition from SmC*-SmA*, then F (θ) can be expressed after transition as

$$F = F_0 + \frac{1}{2}a^*\theta^2 + \frac{1}{4}b^*\theta^4 + \frac{1}{6}c^*\theta^6 \tag{8}$$

One important aspect for the Landau free energy expansion is that the θ^6 term contributes to the free energy of the system when the parameter "b" or "b*" is less than zero. This can be obtained mathematically by setting the second order derivative of the free energy F with respect to θ to be greater than zero.

Now in equation (8) we have $a^*=a=\alpha(T-T_0)$, i.e. no shift of transition point noticed. It was observed earlier by several research groups [19-21] that the shift of transition depends on the thickness of samples or on the surface anchoring energy. But after defining the new secondary order parameter, D we did not observe theoretically any such shift. It does provide the fact that we can define a transition point, D=0 and θ=0 instead of P=0 and θ=0. That definition of transition point may be considered as unique point depending on sample properties itself but not depending on the thickness alike the other definition.

b* and c* are also the modified parameters for the free energy expression in equation (8), given by $b^* = (3b + \frac{16\eta a^4}{\gamma^4}),\ c^* = (5\ c + \frac{96\eta a^3 b}{\gamma^4})$ Since very close to the transition point θ→0, from equation (6), we have

$$\theta = eD \tag{9}$$

Where $e = \frac{\gamma}{2a} = \frac{\gamma}{2\alpha(T - T_0)}$ (10)

Here e is the electroclinic co-efficient, is the electroclinic coupling constant and $\alpha(T-T_0)$ is the first coefficient of the Landau free energy expansion with α as the tilt elastic modulus parameter (non-chiral) describes the restoring torque to return back the director to the layer normal. γ is the chiral parameter describes the coupling between D and θ in the SmC* phase. The linearity between D and θ is broken when we move far from the transition temperature T_0 corresponding to the second order transition between SmC*-SmA*. The linearity of D depends on the variation of temperature (which is quite small in the vicinity of the transition point) and on the coupling constant γ as similar to the variation obtained for P [6, 22].

Considering equation (9), the free energy F can be expressed as given below:

$$F = F_0 + \frac{1}{2}\mu' D^2 + \frac{1}{4}\lambda' D^4 + \frac{1}{6}\nu' D^6 \tag{11}$$

Here μ', λ' and ν' are the modified parameters governing the temperature dependence and the transition mechanism close to the transition point.

Therefore, we can obtain those parameters depending on temperature as given below:

$$\mu' = \frac{\gamma^2}{4a}, \lambda' = (\frac{3b\gamma^4}{64a^4} + \frac{\eta}{4}) \text{ and } \nu' = (\frac{5c\gamma^6}{384a^6} + \frac{\eta b\gamma^2}{4a^3}) \tag{12}$$

Now from equation (11) we have

$$\frac{dF}{dD} = E = \mu' D + \lambda' D^3 + \nu' D^5 \tag{13}$$

The E represents the applied field including both oscillatory and bias field.

Considering a small fluctuation in the applied field close to the transition point, we can express the electric displacement D using Taylor's series expansion in the following manner:

$$D(E + \delta E) = D(E) + \frac{\partial D}{\partial E}\delta E + \frac{1}{2!}\frac{\partial^2 D}{\partial E^2}(\delta E)^2 + ... \tag{14}$$

In view of equation (13) the Landau-Khalatnikov classical equation of motion for the electric displacement is given by the following equation:

$$\Gamma \frac{\partial D}{\partial t} + \mu' D + \lambda' D^3 + \nu' D^5 = E \tag{15}$$

Here Γ represents the kinetic co-efficient of the molecules of the system signifying the orientation mobility of its.

Comparing equations (14) and (15), we have

$$\Gamma \frac{\partial D(E)}{\partial t} + \mu' D(E) + \lambda' D^3(E) + \nu' D^5(E) = E \tag{16}$$

And $\Gamma \frac{\partial \varepsilon}{\partial t} + \mu' \varepsilon + 3\lambda' D^2(E)\varepsilon + 5\nu' D^4(E)\varepsilon = 1$ (17)

Considering $D=D_b+D_0\exp(i\omega t)$ and $E=E_b+E_0\exp(i\omega t)$, where D_0 (D_b) and E_o (E_b) are the magnitude of the oscillatory field (bias field) corresponding to D and E, respectively.

By substituting those in equation (16) we obtain the following relations:

$$\mu' D_b + \lambda' D_b^3 + \nu' D_b^5 = E_b \tag{18}$$

And $\Gamma D_0 i\omega + \mu' D_0 + 3\lambda' D_b^2 D_0 + 5\nu' D_b^4 D_0 = E_0$ (19)

From (18) we obtain the bias dependent dielectric variation as given below:

$$\varepsilon(b) = \frac{D_b}{E_b} = \frac{1}{\mu' + \lambda' D_b^2 + \nu' D_b^4}$$

By approximation of μ' to be very large

$$\varepsilon(b) = \frac{1}{\mu'} - \frac{1}{\mu'^2}[\lambda' D_b^2(E) + \nu' D_b^4(E)] \tag{20}$$

Now from (19) we have,

$$\varepsilon(\omega) = \frac{1}{\mu' + 3\lambda' D_b^2 + 5\nu' D_b^4 + i\omega\Gamma} \tag{21}$$

Equation (21) can be further simplified as,

$$\varepsilon(\omega)=\frac{\{\frac{1}{\mu'}-\frac{1}{\mu'^2}[\lambda' D_b^2(E)+\nu' D_b^4(E)]\}-i\omega\{\Gamma[\frac{1}{\mu'}-\frac{1}{\mu'^2}[\lambda' D_b^2(E)+\nu' D_b^4(E)]^2\}}{1+\omega^2\{\Gamma[\frac{1}{\mu'}-\frac{1}{\mu'^2}[\lambda' D_b^2(E)+\nu' D_b^4(E)]\}^2}$$

So $\varepsilon(\omega)\approx\dfrac{\{\frac{1}{\mu'}-\frac{1}{\mu'^2}[\lambda' D_b^2(E)+\nu' D_b^4(E)]\}-\frac{1}{\mu'}i\omega\tau}{1+\omega^2\tau^2}$ assuming μ′ to be large as stated earlier. The relaxation time is given by $\tau=\dfrac{\Gamma}{\mu'}$.

Now in view of equation (20) we have

$$\varepsilon'(\omega)=\frac{\varepsilon(b)}{1+\omega^2\tau^2}+\varepsilon(\infty)\ and\ \varepsilon''(\omega)=\frac{1/\mu'}{1+\omega^2\tau^2}\omega\tau \tag{22}$$

The new secondary order parameter D is absolutely crucial in defining the behavior of the above curve. The electroclinic behavior is one, which involves the external field inducing some net tilt (θ) in the medium and the absence of such an external bias would lead to no tilt (θ) present in the system. It is this behavior close to the transition point that assumes the system to be identical with the ordinary Smectic-A* phase though it physically appears in the modulated Smectic-C* phase as argued by Meyer et al. [11]. So there should not be any zero field polarization present in the system in the modulated Smectic-C* phase (as θ is zero) but from Fig.1 we are getting a non-zero value of ε′ for D_b=0 i.e. for zero bias field we have a very small contribution of $\frac{\gamma^2}{4a}$ to ε′ as highlighted in the Fig. 1. This contribution to ε′

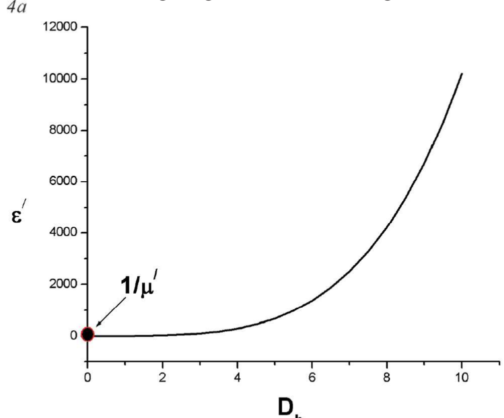

Figure 1: Variation of the real part of the dielectric permittivity (ε′) with the external bias (Db). The units are taken arbitrarily and show the induced polarization in medium by the applied bias.

though small but still significant to notice and as the field D_b increases from zero value ε′ also increases. Since the real part of the dielectric constant defines the polarization mechanism of the system, the zero field value of ε′ as shown in the above curve predicts a non-zero value of polarization present even in the modulated Smectic C* phase very close to the transition point. But recent studies [18] have shown that the de Vries type Smectic-A* phase does contain some initial orientational order with zero external field. The experimentally found value of the polarization present in the system is approximately 119 nC/cm^2. The correlation length corresponding to the above polarization is of the order of nanometers, approximately 22-45nm. In our case we have theoretically shown that there do exist some zero-field orientational order even in the modulated Smectic-C* phase responsible for the origin of the electroclinic behavior. This phenomenon is very interesting and reflects a very important concept about the electroclinic effect and its origin. The situation quite categorically predicts the fact that the modulated Smectic-C* phase is not just any other ordinary Smectic-A* phase but it is indeed identical with the de Vries concept of Smectic-A* phase very close to the transition point as we can't distinguish between these two phases at this very juncture visualizing their same zero-field tilt present in the system. So very close to the transition point our theory predicts a de Vries type behavior of the modulated Smectic-C* phase though the modulation (kind of defects, arrays, lines etc) is still present in the medium. Hence both the modulated phase and the de Vries Smectic-A* phase are in the identical resemblance and thus occupy the same footing as far as the theoretical considerations regarding the electroclinic phenomenon is concerned. Now these predictions are substantial when μ′ is significantly large (eq.19). If we observe the expression for μ′ i.e., $\mu' = \frac{\gamma^2}{4a}$ we see that a large value of μ′ is only observed when "a" is very small in fact $a \to 0$ i.e., $T \to T_0$ and γ is very large. $a \to 0$ means we are indeed minutely away from the transition point and a very large value of γ predicts a very strong coupling between D and θ i.e. a strong electroclinic nature of the medium. So our theory validates its predictions almost at the juncture of the transition point (may be some nano-kelvin difference in temperature) and if we can manage to attain such an experimental environment we would not be able to distinguish between modulated Smectic-C* phase and the de Vries Smectic-A* phase. The introduction of D plays an indispensable role in conceptualizing these theoretical arguments and hence is worth introducing.

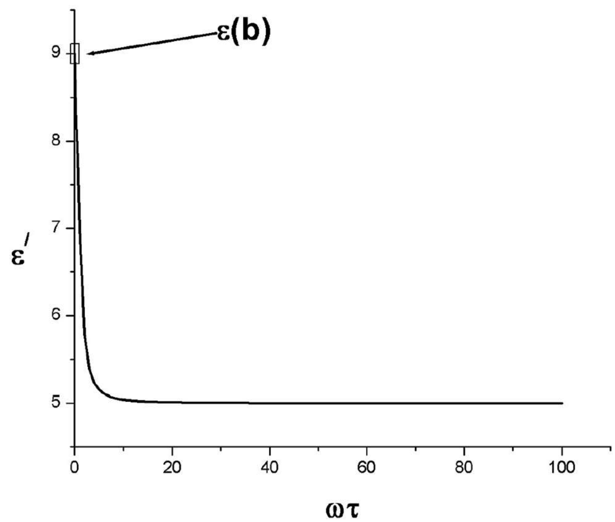

Figure 2: Variation of the real part of the dielectric permittivity (ε′) with frequency (ω). The units are taken arbitrarily.

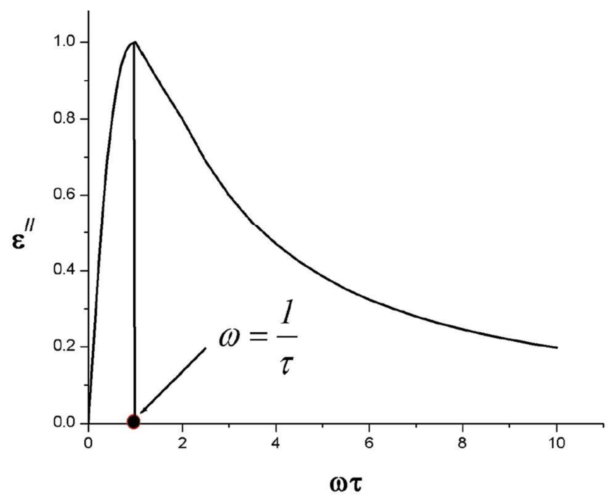

Figure 3: Variation of the imaginary part of the dielectric permittivity (ε″) with frequency (ω). The units are taken arbitrarily.

Figs. 2 and 3 demonstrate the frequency dependency of the real (ε′) and the imaginary (ε″) parts of the dielectric permittivity (ε). The behavior is the typical soft mode type relaxation for a Smectic C* liquid crystals [23]. ε′ decreases with the increase of frequency (Fig. 2) because of negligible effect of the dipolar polarizability at sufficiently high frequency due to the inertial response of the dipolar molecules towards a rapidly oscillating field. So at high frequency the dipolar polarizability contributes a little and electronic polarizability with substantially significant, corresponding to a rapidly oscillating field contributes to ε'. From Fig.3, at $\omega=1/\tau$ ($\tau = \Gamma/\mu'$.), the loss factor of the system is maximum. If μ' becomes very large, the randomization of the molecules are greatly increased and if μ' becomes very small, the molecules of the system almost freeze themselves at their respective lattice sites, thus contributing very little to the dielectric spectrum of the system.

By putting equation (14) to equation (13) and equating the coefficient of $(\delta E)^2$ we have

$$[3\lambda' D(E)+10\nu' D^3(E)]\varepsilon^2 = -[\frac{\mu'}{2}+\frac{3\lambda'}{2}D^2(E)+\frac{5\nu'}{2}D^4(E)]\frac{\partial\varepsilon}{\partial E} \tag{23}$$

The third bracketed term on the right-hand side of equation (23) is basically $\frac{\partial E}{\partial D}$ as can be seen from the equation (13). So, equation (23) now modifies to the following equation i.e.

$$\begin{aligned}[3\lambda' D(E)+10\nu' D^3(E)]\varepsilon^2 &= -\frac{1}{2}(\frac{\partial\varepsilon}{\partial E})(\frac{\partial E}{\partial D}) \\ &= -\frac{1}{2}(\frac{\partial\varepsilon}{\partial D})\end{aligned} \tag{24}$$

Equation (24) now can be further modified to yield the following expression i.e.

$$\varepsilon^2(\frac{\partial D}{\partial\varepsilon}) = -\frac{1}{6\lambda' D(E)}[1+\frac{10\nu'}{3\lambda'}D^2(E)] \tag{25}$$

Now taking the expression of D as D= D_b+ D_0 exp (iωt), equation (25) now takes the following form i.e.

$$\varepsilon^2(\frac{\partial D}{\partial\varepsilon}) = -\frac{1}{6\lambda' D_b}(1-\frac{D_0}{D_b}exp\,i\omega t)[1+\frac{10\nu'}{3\lambda'}(D_b^2+D_0^2\,exp\,2i\omega t+2D_bD_0\,exp\,i\omega t)]$$

The above expression can now be further modified in the manner stated below i.e.

$$\varepsilon^2(\frac{\partial D}{\partial \varepsilon}) = \xi_1(T,D) + \xi_2(T,D)\exp i\omega t + \xi_3(T,D)\exp 2i\omega t \tag{26}$$

The left-hand side of equation (25) i.e. $\varepsilon^2(\frac{\partial D}{\partial \varepsilon})$ is basically the charge density times the capacitance in the medium. The negative sign signifies the energy flow for the system as it is subjected to the external field.

Here the coefficients of equation (26) are provided by the following expressions i.e.

$$\xi_1(T,D) = (-\frac{1}{6\lambda' D_b} - \frac{5\nu' D_b}{9\lambda^2}), \xi_2(T,D) = (\frac{D_0}{6\lambda' D_b^2} - \frac{10\nu' D_b D_0}{9\lambda^2 D_b}), \xi_3(T,D) = -\frac{5\nu' D_0^2}{9\lambda^2 D_b}$$

Equation (26) is obtained by taking the co-efficient of $(\delta E)^2$. It provides an idea about the capacitive nature of the molecular environment very close to the transition point. The capacitance of a system provides us the energy storage mechanism i.e. how the system stores the charges in the medium for substantive amount of time. But if we see the expression (26) closely and its co-efficients we can conclude that very close to the transition point the system losses energy as the charges now flow out of the system in due course. This is perfectly understandable as the right-hand side of the equation (26) is $\varepsilon^2(\frac{\partial D}{\partial \varepsilon})$ signifying the capacitive nature of the medium being varying inversely with the charge density of the medium. As we increase the applied field on the system very close to the transition point the energy flux i.e. the energy flowing out of the system becomes appreciable. This is reasonable because very close to the transition point the system becomes very much constrained and in order to achieve a stable energetic situation it undergoes a transition to another phase i.e. the Smectic-A* phase. So, when the applied field is substantially large the system or the molecules as a whole become quite strained in their orientational freedom. This situation further aggravates if we assume that the arrays, lines and the defects in the medium now are also in the conspicuous situation resulting in the enhancement of the constrained nature of the system. Thus, the system releases the energy in order to achieve a stable equilibrium i.e. the stored charges for a considerable amount of time being released. For both zero and infinitely large value of D_b the energy flux diverges. For infinitely large value of D_b very close to the transition point the system no longer can follow Landau expression in terms of as usual secondary order parameter P because of the fluctuations in both of the applied field and the order parameters. Again, for zero value of D_b though there are some nanoscale orientational orders present in the medium (as demonstrated regarding our discussion about Fig.1) but still it is found to be insufficient to bring the stability in the system. We have to put some field of intermediate range in the system to make it stable at the vicinity of the transition point and to achieve a strong electroclinic effect, otherwise the system would continuously lose energy to achieve stability by going over to the Smectic-A* phase. Since the dielectric constant and the capacitance of a system show the similar behavior as obtained from equation (26), hence the physical arguments regarding the real and the imaginary parts of the expression $\varepsilon^2(\frac{\partial D}{\partial \varepsilon})$ may be assumed similar physical scenarios as demonstrated in the equations (19) and (20). We call the expression $\varepsilon^2(\frac{\partial D}{\partial \varepsilon})$ the response parameter since it reveals quite a lot about the physical situations of the system when it is exposed to the external field and thus is worth stating.

Conclusion

We have successfully obtained the behavior of modulated phases with the consideration of newly defined order parameter. By defining a new order parameter D we eventually obtained the dielectric function as a modulated nature taking into consideration of infinitesimal fluctuation from equilibrium. It definitely ascertains the existence of modulated Smectic-C* phase due to the fluctuation of the system as a whole under the influence of an external applied field. Besides we have been able to get an in-depth knowledge of the capacitive nature of the system by considering the new order parameter. We finally revealed several aspects such as response parameter from the electroclinic behavior of a system based on our newly defined concept in terms of newly defined order parameter.

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