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Research Article

Temperature Dependence of the Tilt Angle for the Smectic A-Smectic C* Transition in a Mixture of C7-70PDOB Ferroelectric Liquid Crystals near the Tricritical Point

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The temperature dependence of the tilt angle θ is studied in the smectic C* phase near the smectic A-smectic C* tricritical point for a mixture of 70PD0B in the ferroelectric liquid crystal C7 ($X = 16.92$). The mean-field models with the biquadratic $P^2\theta^2$ (P is the spontaneous polarization) and $P^2\theta^2 + P\theta$ (bilinear) coupling terms in the free energy expansion are used to analyze the experimental data for the tilt angle in this binary mixture. From our analysis, the coefficients given in the free energy expansion of the mean-field models are determined. Our results show that the mean-field theory explains adequately the observed behaviour of the C7-70PD0B mixture near the AC* tricritical point.

1. Introduction

Smectic transitions in ferroelectric liquid crystals have been the subject of various experimental and theoretical studies. In particular, smectic A and C (or C*) transitions have been investigated and reported in the literature.

In the smectic A phase, the liquid molecules are oriented along the director (orientational order parameter) and they show a transitional order in layers, whereas in the smectic C (or C*) phase, additionally they make a tilt angle with the director which is perpendicular to the smectic layers. If the chiral molecules, which are optically active, exist in the liquid crystalline material, then the smectic C* phase occurs.

The smectic A-smectic C* (AC*) transition has been predicted by de Gennes [1] as the transition which belongs to the three-dimensional XY universality class ($n = 2$, $n = 3$). This has not been confirmed by some early experiments which are in agreement with a Landau mean-field theory of the AC* transition [2–7]. Some experimental [8–13] and theoretical [3, 14, 15] studies have shown that from the first-order to a second order AC* transition, there occurs a tricritical point (TCP). In our recent studies, tricritical behaviour of mixtures of C7+10.04 [16] and SCE9+SCE10 [17, 18] has been shown using the mean-field models. Mean-field models are applicable to many liquid crystalline systems and they predicted adequately the observed behaviour of those materials exhibiting phase transitions. The physical quantities measured to high accuracy can then be analyzed by a mean-field model and the type of transition (first order, second order, or tricritical) can be characterized. From this point of view, mean-field model is a more realistic theoretical model to be applicable to the experiments.

The mean-field models which describe the AC (or AC*) transitions have the free energy expanded in terms of the order parameters (tilt angle θ and the spontaneous polarization P) and their bilinear ($P\theta$) and/or biquadratic ($P^2\theta^2$) couplings [3, 14, 19, 20]. Dipolar interactions ($P\theta$ coupling) are of a chiral character which becomes importantly close to the AC* transition, whereas quadrupolar interactions ($P^2\theta^2$ coupling) are of a nonchiral one which covers a wide range of temperatures below the transition temperature in the smectic C or AC* phase. On the basis of our earlier mean-field models with the biquadratic coupling ($P^2\theta^2$), recently we have calculated tilt angle and the temperature shifts

as a function of concentration for a mixture of 10.04+C7 [16] and the temperature dependence of the spontaneous polarization and the tilt angle for a mixture of SCE9+SCE10 [17, 18] for the AC* transition. Very recently, we have studied the dielectric constant as a function of temperature for the AC* transition in 4-(3-methyl-2-chlorobutanoyloxy)-4′-heptyloxybiphenly (A7) [21] and we have calculated a generalized smectic-hexatic phase diagram in various mixtures of liquids crystals [22].

In this study, we focus on the temperature dependence of the tilt angle for a mixture of C7-70PDOB ferroelectric liquid crystal close to the tricritical point (TCP). The ferroelectric material 4-(3-methyl-2-chlorobutanoyloxy)-4′-heptyloxy biphenly (C7) exhibits a large spontaneous polarization [11]. It undergoes a first-order AC* transition. When adding a second compound of 4-heptyloxy-4′-decloxybenzoate (7OPDOB), this transition is driven towards a second order by passing through a tricritical point (TCP) [9]. The transition temperatures of C7 for the relevant phases are as follows: SmC* (53.9°C) and SmA (54.06°C) [8]. The AC* transition temperatures of a binary mixture of C7+7OPDOB depending on the X values (mol percent of 7OPDOB) [9] are given in Table 1. Ferroelectric liquid crystals with large spontaneous polarizations such as C7 can be used in switching display devices and liquid crystals devices. Their carbon-13 nuclear magnetic resonance can be obtained experimentally to determine the temperature dependence of order parameter. In our previous study [21], we used our mean-field model with the $P^2\theta^2$ coupling and calculated the dielectric constant (dielectric susceptibility) at various temperatures under constant electric field for A7. Our calculation for the tilt angle (order parameter) as a function of temperature was performed [21] using the molecular field theory [23] for A7. The experimental data for the dielectric constant of A7 [24] was then analyzed according to the expression derived from the mean-field model [21]. Similarly, we have used our mean-field model with the quadratic couplings $\Psi^2\theta^2$ (Ψ is the orientational order parameter and θ is the tilt angle) for the smectic-hexatic phase transitions in various mixtures of liquid crystals in our recent study [22]. By expanding the free energy in terms of the order parameters (Ψ and θ), we derived the phase line equations and calculated a generalized phase diagram (T-X) on the basis of the experimental data [25].

In the present study, not only the quadric coupling $P^2\theta^2$ but also $P^2\theta^2 + P\theta$ coupling in the free energy of our mean-field model is considered and the temperature dependence of the tilt angle θ is derived in both cases. Instead of using the temperature dependence of the order parameter from the order parameter, from the molecular field theory [23], as we have also needed in our recent study [21], the θ versus T relation derived here is directly used to analyze the experimental data [9]. This is more general in the sense that the experimental data can be analyzed freely with the fitted parameters determined, whereas the molecular field theory [23] predicts the temperature dependence of the order parameter according to the critical exponent 1/2. This, however, can restrict the analysis of the experimental data for various mixtures of liquid crystals. In order to get good fits, our θ versus T expression is favorable. On this basis, we have fitted to the experimental data [9] the expression for the tilt angle as a function of temperature, which we derive from the mean-field models with the $P^2\theta^2$ and $P\theta + P^2\theta^2$ couplings for the AC* transition close to the TCP. Coefficients given in the mean-field energy are calculated from our fits and they are interpreted within the AC* transition in this mixture of C7-70PDOB.

In Section 2, we give an outline of our mean-field models, where the temperature dependence of the tilt angle and the spontaneous polarization are derived. In Section 3, the tilt angle expression is fitted to the experimental data and the results are presented. Sections 4 and 5 give our discussion and conclusions, respectively.

2. Theory

The free energy of the smectic C* phase can be expanded in terms of the tilt angle θ and the spontaneous polarization P for the AC* transition in ferroelectric liquid crystals. By considering the quadrupolar interaction between the θ and P (biquadratic coupling $P^2\theta^2$), the free energy can be expressed as

$$g = \frac{1}{2}a(T - T_0)\theta^2 + \frac{1}{4}b\theta^4 + \frac{1}{6}c\theta^6 + \frac{1}{2\chi_0\varepsilon_0}P^2 - DP^2\theta^2 + \frac{1}{4}eP^4 - EP \tag{1}$$

in the presence of the electric field E, where a, b, c, D, and e are constants. T_0 is the transition temperature, χ_0 is the static dielectric susceptibility, and ε_0 is the permittivity in free space. In the free energy expansion, the tilt angle θ is taken as the primary order parameter which does not exist in the smectic A phase. The spontaneous polarization P is the secondary order parameter which is defined in both the smectic A and C* phases. We have introduced this model in our previous study [19].

In order to describe the AC* transition, the energy is minimized with respect to the order parameters P and θ. By minimizing the free energy g with respect to the spontaneous polarization P, we get at the zero electric field ($E = 0$),

$$\frac{1}{\chi_0\varepsilon_0} - 2D\theta^2 + eP^2 = 0. \tag{2}$$

Also, by minimizing the free energy g with respect to the tilt angle θ, we find that

$$a(T - T_0) + b\theta^2 + c\theta^4 - 2DP^2 = 0. \tag{3}$$

Equations (2) and (3) give the temperature dependence of the spontaneous polarization and tilt angle, respectively. By substituting (2) into (3), the temperature dependence of the primary order parameter θ (tilt angle) can be written as

$$(T - T_C) = \frac{-c}{a}\theta^4 + \frac{1}{a}\left(\frac{4D^2}{e} - b\right)\theta^2 - \frac{3}{16 \cdot c \cdot a}\left(b - \frac{4D^2}{e}\right)^2. \tag{4}$$

Table 1: Transition temperature (T_{AC^*}) for the different mixtures of C7+7OPDOB system [9].

Mole percent	T_{AC^*} (°C)
16.92	44.668
15.49	45.120
14.02	45.738
13.60	46.095
13.30	46.271

In (4) the temperature shifts,

$$\Delta T = T_C - T_0 = \frac{3}{16ac}\left(b - \frac{4D^2}{e}\right)^2 - \frac{2D}{ea\chi_0\varepsilon_0}, \quad (5)$$

were used for a first-order AC* transition [19]. In (5) T_C denotes the experimentally measured AC* transition.

The free energy of the smectic C* phase can also be expanded in terms of the tilt angle θ and the spontaneous polarization P by considering both the dipolar interactions (bilinear coupling $P\theta$) and the quadrupolar interactions (biquadratic coupling $P^2\theta^2$) for the AC* transition in ferroelectric liquid crystals. The free energy can then be expressed as

$$g = \frac{1}{2}a(T - T_0)\theta^2 + \frac{1}{2}b\theta^4 + c\theta^6 + \frac{1}{2\chi_0\varepsilon_0}P^2 - DP^2\theta^2 - CP\theta. \quad (6)$$

By minimizing the free energy with respect to the spontaneous polarization P and the tilt angle θ as before, one gets

$$\frac{P}{\chi_0\varepsilon_0} - C\theta - 2DP\theta^2 = 0, \quad (7)$$

$$a(T - T_0)\theta + b\theta^3 + c\theta^5 - CP - 2DP^2\theta = 0, \quad (8)$$

respectively. By substituting (7) into (8), the temperature dependence of the tilt angle θ can be written as

$$(T - T_0) = \frac{-b}{a}\theta^2 - \frac{c}{a}\theta^4 + \frac{1}{a}\left(\frac{\chi_0\varepsilon_0 C^2}{1 - 2D\chi_0\varepsilon_0\theta^2}\right) + \frac{1}{a}\left(\frac{2D\chi_0^2\varepsilon_0^2 C^2\theta^2}{\left(1 - 2D\chi_0\varepsilon_0\theta^2\right)^2}\right). \quad (9)$$

Using (1) and (6) with the $P^2\theta^2$ and $P^2\theta^2 + P\theta$ coupling in the free energies, respectively, the temperature dependence of the tilt angle θ can be predicted for the AC* transition in ferroelectric liquid crystals.

3. Calculations and Results

The temperature dependence of the tilt angle θ was calculated for the AC* transition of a binary mixture of 70PDOB in C7 near the tricritical point ($X = 16.92$). The value X is the mol percent (mol %) of 70PDOB in C7, as given in Table 1 [9]. For this calculation, (4) was fitted to the experimental data [9] and the coefficients were determined. Table 2 gives values of the coefficients with the uncertainties from (4) according to the biquadratic coupling ($P^2\theta^2$) for this mixture of 70PDOB+C7 when the tricritical concentration of 70PDOB is $X = 16.92$ for the AC* transition. We plot θ versus $T - T_C$ in Figure 1. We also calculated the temperature dependence of the tilt angle θ by fitting (9) to the experimental data [9] in the case of both biquadratic ($P^2\theta^2$) and bilinear ($P\theta$) couplings. Coefficients with the uncertainties and plots of θ versus $T - T_0$ are given in Table 2 and Figure 2, respectively.

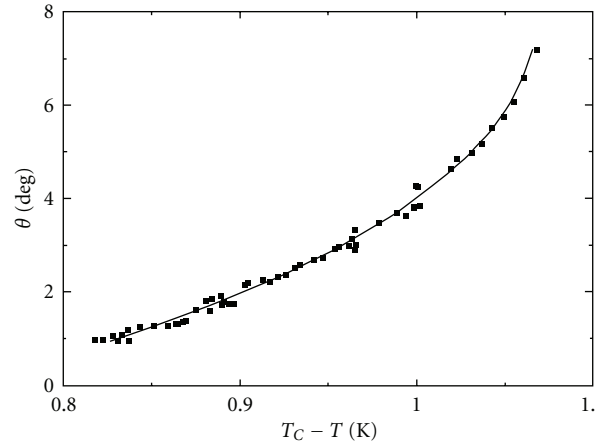

Figure 1: Tilt angle θ as a function of $T - T_C$ for the AC* transition in a mixture of 70PDOB+C7 near the tricritical point ($X = 16.92$) according to (4) with the $P^2\theta^2$ coupling.

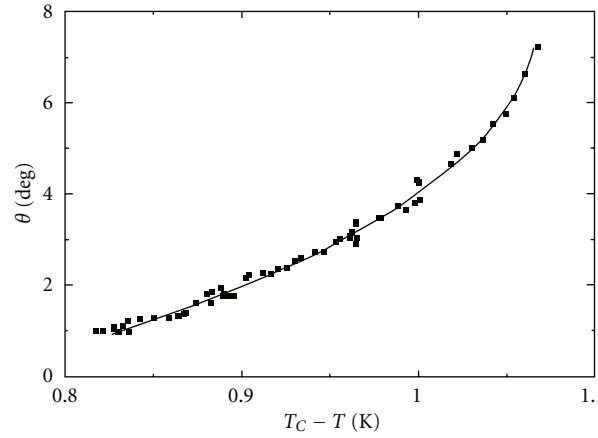

Figure 2: Tilt angle θ as a function of $T - T_C$ for the AC* transition in a mixture of 70PDOB+C7 near the tricritical point ($X = 16.92$) according to (9) with the $P\theta + P^2\theta^2$ coupling.

4. Discussion

The tilt angle θ was calculated as a function of temperature in the C* phase for the AC* transition near the tricritical concentration of 70PDOB ($X = 16.92$) in C7 using the mean-field models with the $P^2\theta^2$ (1) and $P\theta + P^2\theta^2$ (6)

Table 2: Values of the parameters given in the equations indicated for the AC* transition in a mixture of 70PDOB+C7 near the tricritical point ($X = 16.92$).

χ_0	T_0 (°C)	a (J/m^3 K deg^2)	b (J/m^3 deg^4)	c (J/m^3 deg^6)	C (J/m^3 C deg)	D (J·m/C^2 deg^2)	e (Jm^5/C^4)	Equation
4.59	44.668	1642.41 ± 1.30	15.41 ± 0.01	-5.32×10^{-5} $\pm 4.78 \times 10^{-8}$	—	2.546 ± 0.001	3.557 ± 0.002	(4)
4.59	44.668	1.77×10^{17} $\pm 1.7 \times 10^{14}$	8.31×10^{14} $\pm 4.6 \times 10^{12}$	3.18×10^{12} $\pm 1.13 \times 10^{11}$	6.83×10^{13} $\pm 2.6 \times 10^{7}$	4.05×10^{6} $\pm 1.4 \times 10^{5}$	—	(9)

couplings, as shown in Figures 1 and 2, respectively. Both equations, (4) with the $P^2\theta^2$ coupling and (9) with the $P\theta + P^2\theta^2$, when fitted to the experimental data [9], describe the observed behaviour of the tilt angle θ near the tricritical point (TCP) satisfactorily. We also calculated the standard deviations of the fitted parameters for both mean-field models, as given in Table 2. The uncertainties in the coefficients of the free energies ((1) and (6)) are comparatively small in most cases (Table 2). However, these uncertainties can be significant for the coefficients in the free energies for both models since the critical behaviour of the mesomorphic mixture depends on those coefficients, in particular the coefficients of the coupling terms ($DP^2\theta^2$ and $CP\theta$). For the mesomorphic mixture studied here, regarding the values of C and D (Table 2) for the second mean-field model (6), the bilinear $P\theta$ coupling is dominant in comparison with the biquadratic coupling $P^2\theta^2$. However, considering all the values of the fitted parameters in the free energy expansion (Table 1), the mean-field model with the $P^2\theta^2$ coupling (1) can still be preferred since (4) provides the values of the fitted parameters which are physically meaningful. This then indicates that the dominant mechanism of the smectic AC* transition near the tricritical point is due to quadratic interactions for a mixture of 70PDOP+C7. In (1) the biquadratic coupling terms ($P^2\theta^2$) characterizes the nonchiral properties and induces a transverse quadrupolar ordering. This is the dominant term in a wide temperature range far away from T_C. To stabilize the mesomorphic mixture for far away T_C, we included the P^4 term, which has also been considered in the generalized mean-field model for the smectic A-chiral-smectic C phase transition in the case of p-(n-decyloxybenzylidence)-p-amino-(2-methylbutly) cinnamate (DOBAMBC) in an earlier study [3]. Since the bilinear coupling term ($P\theta$) characterizes the chiral properties close to the T_C, the mesomorphic mixture is already stabilized with the biquadratic coupling term ($P^2\theta^2$) and, no additional term such as P^4 is required, in (6). So that the free energy of the smectic C* phase is given in terms of the spontaneous polarization up to P^2 with the bilinear coupling term $P\theta I$ as also given previously [3] and with this free energy the system is stable. Thus, the temperature dependence of θ is for both mean-field models studied here. In fact the case of DOBAMBC, the relative importance of biquadratic term $P^2\theta^2$ and P^4 term in comparison with terms involving $P\theta$ and P^2 has been demonstrated numerically using the generalized mean-field model for the AC* transition [3, Table III]. Thus, as explained previously [3], when relatively reliable experimental data is obtained in the temperature range studied, the biquadratic term $P^2\theta^2$ and P^4 are not negligible as we also demonstrate in our study here (Table 2). Thus, the terms θ^6, $P^2\theta^2$, θ^4, and P^4 (1), and $P^2\theta^2$, $P\theta$, and P^2 (6) are required to explain the major features of the AC* transition in C7-7OPDOB. We also note that from the data analysis point of view, if the P^4 term was added to the free energy of the C* phase (6), the fitting of the model to the experimental data would not have been a straightforward problem. This is due to the P^3 term appearing in (7) when the free energy was minimized with respect to the polarization P. As a cubic polarization equation, two solutions would correspond to a local minimum of the free energy, which may then incorporate the thermal fluctuations in the mean-field model, as also pointed out previously [14].

The temperature dependence of the tilt angle θ was calculated here near the tricritical point ($X = 16.92$) using the experimental data [9] where they obtained $\beta = 0.436$ according to a power-law formula

$$\phi = \phi_0 \left[\frac{T_{AC^*} - T}{T_{AC^*}} \right]^{\beta}. \tag{10}$$

In (10) ϕ is the tilt angle, ϕ_0 is the amplitude, T_{AC^*} is the AC* transition temperature, and β is the critical exponent for the order parameter [26]. The experimental tilt-angle data were analyzed within the temperature interval of 240 mK for $X = 16.92$ mixture of 70PDOB in C7 [9]. The experimental tilt-angle (ϕ) data were also the same as those obtained from the analyses within the temperature intervals of 103 mK ($\beta = 0.5$), 490 mK ($\beta = 0.38$), and 930 mK ($\beta = 0.343$) [9]. Similarly, the temperature dependence of the tilt angle can also be calculated for the AC* transition in a mixture of 70PDOB-C7 in the case of various concentrations of 70PDOB on the basis of our mean-field models given here. Since the tilt angle ϕ has been obtained from the layer spacing data as a function of $T_{AC^*} - T$ for C7 and its mixture with 70PDOB for concentrations of 0 (C7), 5.15, 9.68, 15.0, and 19.4 [8], (4) and (9) can be fitted to those data to describe the AC* transition in this mixture. Since $X = 0$ corresponds to a first-order AC* transition in C7, as the concentration of 70PDOB increases towards $X = 19.4$, the AC* transition then becomes a second order through $X = 16.92$ (tricritical transition) for this mixture of 70PDOB-C7. Thus, from a first order to a second order via the tricritical point (TCP) AC* transition, as the concentration (70PDOB) increases, can be described adequately by calculating the temperature dependence of the tilt angle using the experimental data [9] on the basis of our mean-field models studied. Mean-field to

tricritical crossover behaviour near the AC* tricritical point can then be better understood for a mixture of 70PDOB-C7. This work is under progress.

5. Conclusions

The tilt angle was predicted as a function of temperature for the AC* transition of a mixture of 70PDOB-C7 close to the tricritical point. The mean-field models which consider the quadratic and dipolar interactions between the tilt angle and the spontaneous polarization were used for this calculation of the tilt angle. Expressions for tilt angle which we derived from our mean-field models were fitted to the experimental data and the coefficients were determined.

Our results show that quadratic interactions seem to be dominant in the mechanism of the AC* transition for the 70PDOB-C7 mixture near the tricritical point in regard to the experimental data.

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