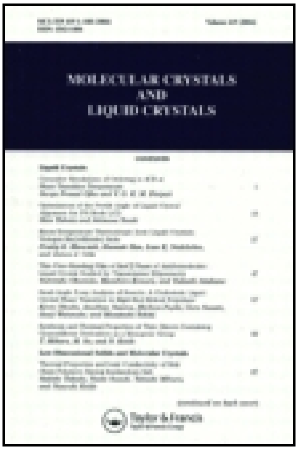

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Nature of the Smectic-A-Chiral-Smectic-C Phase Transition

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Nature of the Smectic-A-Chiral-Smectic-C Phase Transition†

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Details of a generalized mean-field model will be presented. A systematic fitting scheme will be discussed to acquire all eleven expansion coefficients in this mean-field model. Very good fitting results are obtained on our high-resolution heat-capacity, tilt-angle, and spontaneous polarization data in the vicinity of the smectic-A-chiral-smectic-C phase transition of *p*-(*n*-decyloxybenzylidene)-p-amino-(2-methyl-butyl)cinnamate (DOBAMBC). The fitting also gives qualitative account for one set of existing helicoidal pitch data.

Keywords: smectic phase, mean-field phase transition, heat capacity, tilt angle, polarization, pitch

I. INTRODUCTION

Based on their high-resolution heat-capacity results in the neighborhood of one smectic-A (SmA)-smectic-C (SmC) transition of one liquid-crystal compound, Huang and Viner[1] have proposed an extended mean-field model to describe the behavior of this phase transition. Soon after, many high-resolution heat-capacity and tilt-angle studies have demonstrated that not only the SmA-SmC transition but also the SmA-chiral-smectic-C (SmC*) transition are well described by the extended mean-field model.[2] The molecular tilt angle is the order parameter associated with the SmA-SmC transition. Besides the tilt angle, because of the existence of chirality in the SmC* phase,[3] both spontaneous polarization and helicoidal pitch are two additional important macroscopic quantities characterizing the SmA-SmC* tran-

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sition. The success of the extended mean-field model in describing the temperature variations of heat capacity as well as tilt angle near the SmA-SmC* transition supports the idea[3] that the tilt angle (θ) is the primary order parameter and the spontaneous polarization is the secondary one for this transition. This idea will gain quantitative support from our fitting results reported in this paper. To give full account of the observed temperature variation of spontaneous polarization (P) and helicoidal pitch ($L = 2\pi/q$) a reasonable number of coupling terms involving θ, P, and q have to be added to the free energy expression. The variable q is the wave vector for the helicoidal modulation.

Considering the symmetry of the group representation for the SmC* phase, Indenbom *et al.*[4] have proposed a phenomenological mean-field model including leading coupling terms containing P and/or q. This free energy expansion serves as a starting point to describe the SmA-SmC* transition but fails to explain most of the pertinent experimental results which are found just below the SmA-SmC* transition. In the light of our almost simultaneous measurements on the polarization and tilt angle in the neighborhood of the SmA-SmC* transition of *p*-(*n*-decyloxybenzylidene)-p-amino-(2-methylbutyl)cinnamate (DOBAMBC), we have demonstrated that a generalized mean-field model,[5,6] similar to the one recently proposed by Zeks,[7] can give us a fairly good fitting for the temperature variations of the heat capacity, tilt angle, polarization, and the ratio P/θ and a qualitative explanation for the anomalous behavior of the helicoidal pitch. In the next section we will present our experimental results on heat capacity, tilt angle and polarization in the vicinity of the SmA-SmC* transition of DOBAMBC. A brief discussion of the extended mean-field model proposed by Huang and Viner and the phenomenological mean-field model by Indenbom *et al.* will be given in Sec. III and IV, respectively. Finally, the generalized mean-field model, which gives a much better description for the SmA-SmC* transition, and our fitting results will be presented in Sec. V. In Sec. VI we draw our conclusions.

II. EXPERIMENTAL RESULTS

Details of our ac calorimeter for measuring heat capacity have been reported elsewhere.[8] After subtracting the addendum contribution of the sample cell, thermocouple etc., the sample heat capacity per unit area (thickness of the sample 72 μm) in both the SmA and SmC* phase of DOBAMBC is shown as solid dots in Figure 1. The data were taken in a fairly wide temperature range (about 25 K).[9]

Employing an electro-optical effect, we have measured tilt angle with resolution 1.7×10^{-3} rad. The temperature resolution is 3 mK. To eliminate the electro-clinic effect on the measured tilt angle, at a

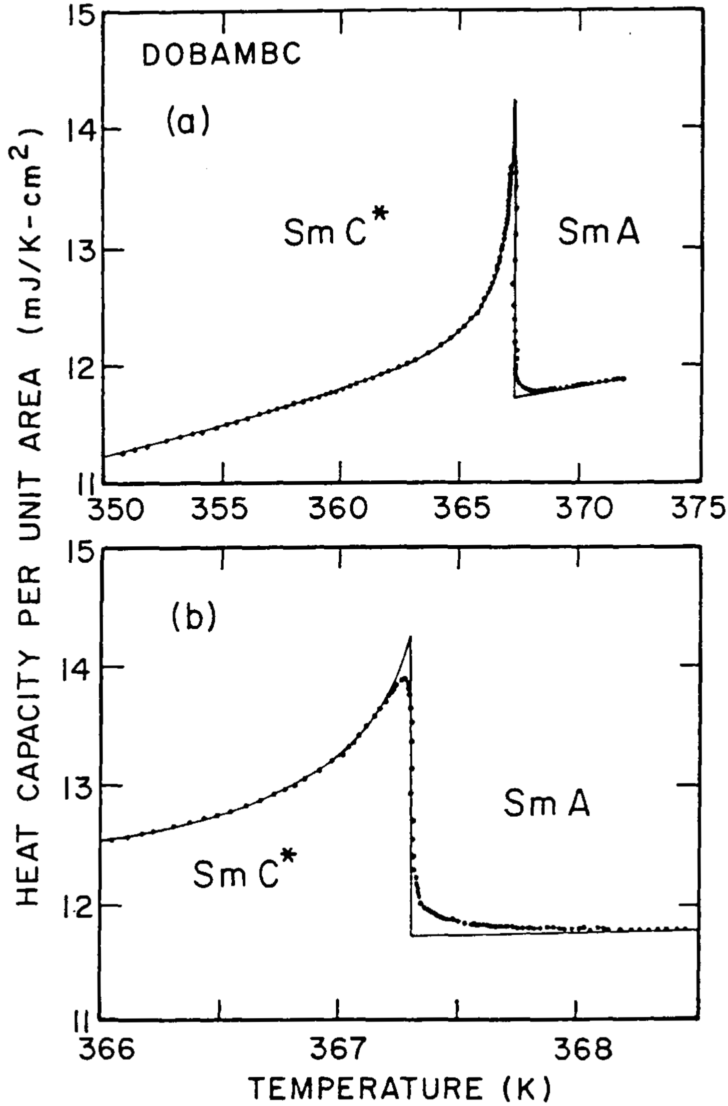

FIGURE 1 Heat capacity anomaly near the SmA-SmC* transition of DOBAMBC. Dots, experimental data; line, fitted line.

given temperature the tilt angles were measured with at least four different applied electric fields. An extrapolation to the zero-field value is our measured tilt angle. The data are displayed in Figure 2 as solid points.[5]

Finally, the spontaneous polarization is determined from measurements of the displacement current through the field-reversal method. To facilitate data reduction, the sample cells which were coated with transparent and conducting indium-tin-oxide (ITO) films were driven by an ac triangular wave.[10] Figure 3 shows our polarization data as solid dots.[5]

In order to eliminate the effect of the T_c shift (our samples for tilt-angle and polarization measurements had a T_c shift rate of about 20 mK/Hr) and obtain reliable data on the ratio P/θ, for a given temperature, we measured P and θ one after the other. This unique approach enables us to reveal a surprising anomaly in the temperature variation of the ratio P/θ. Our data are shown in Figure 4 as solid points.[5] The fast drop of the ratio P/θ in the region $T_c\text{-}T \lesssim 1.5$ K reminds us of the similar behavior of the helicoidal pitch. Since our first finding in the anomalous behavior in the ratio P/θ, similar anom-

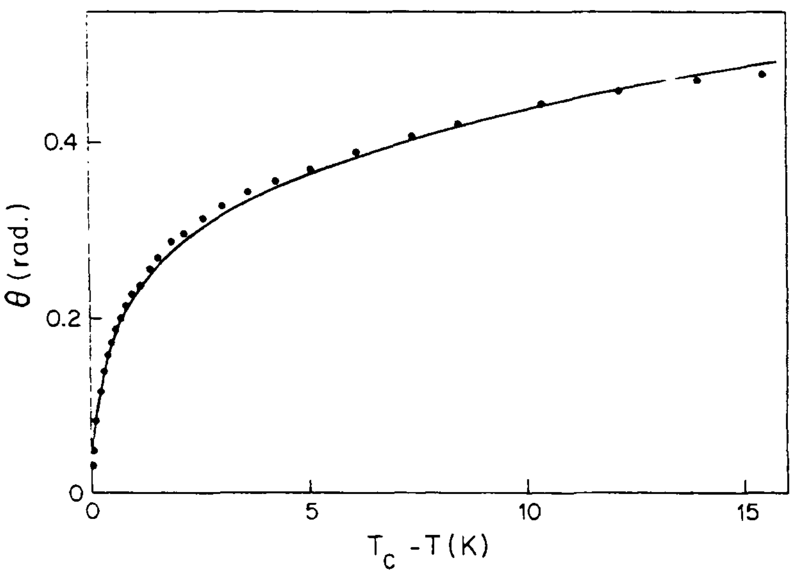

FIGURE 2 The temperature dependence of the tilt-angle for DOBAMBC. Dots, experimental data; line, fitted line.

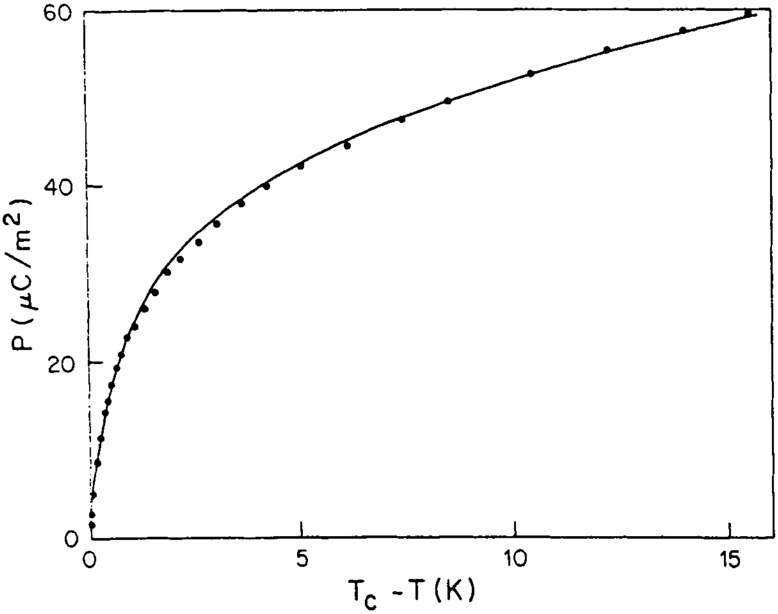

FIGURE 3 The temperature dependence of the spontaneous polarization for DOBAMBC. Dots, experimental data; line, fitted result.

aly has been observed in the majority of the reported P/θ.[11,12,13] However, at least one case with slightly different temperature variation in P/θ has been found also.[11]

III. EXTENDED MEAN-FIELD THEORY

So far all the high resolution studies near the SmA-SmC (or SmC*) transition have demonstrated that this phase transition can be well-described by the following extended mean-field model,[1]

$$G = G_0 + \frac{1}{2} a\, \bar{t}\theta^2 + \frac{1}{4} b\theta^4 + \frac{1}{6} c\theta^6. \qquad (1)$$

Here G_0 is the nonsingular part of free energy. The coefficients a, b, and c are positive for a continuous transition. For the convenience of the latter discussion, we choose $\bar{t} = T\text{-}T_c$. Here T_c is the transition temperature. The variable θ is the molecular tilt angle from the smectic layer normal and the amplitude of the SmC order parameter.

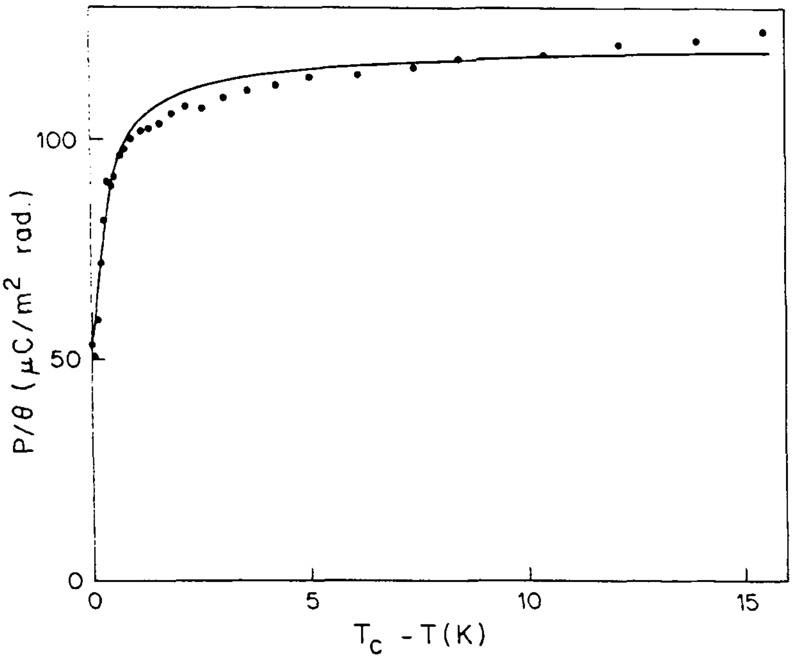

FIGURE 4 The temperature dependence of the ratio P/θ for DOBAMBC. Dots, experimental data; line, fitted result.

From Eq. (1), it is easy to obtain the following expressions for tilt angle θ and heat capacity.

$$\theta = \begin{cases} 0 & T > T_c \\ ((b/2c)((1-3\tilde{t}/T_c t_0)^{1/2}-1))^{1/2} & T < T_c \end{cases} \tag{2}$$

and

$$C_p = \begin{cases} C_0 & T > T_c \\ C_0 + AT(T_m - T)^{-1/2} & T < T_c. \end{cases} \tag{3}$$

Here $t_0 = 3b^2/(4acT_c)$. C_0 is the nonsingular part of the heat capacity. $A = (a^3/c)^{1/2}/4$ and $T_m = T_c(1 + t_0/3)$. The mean-field heat-capacity jump at T_c is $\Delta C_J = T_c a^2/(2b)$. Furthermore, one can show that θ and the anomalous part of heat capacity ($\Delta C_p = C_p - C_0$) are related by a simple relation, i.e.,

$$\theta = \left[\frac{2}{a}\int_T^{T_c} \frac{\Delta C_p}{T}\, dT\right]^{1/2} \tag{4}$$

Thus from Eq. (4) as well as the expression for t_0 and ΔC_J, the three mean-field expansion coefficients (a, b, and c) can be obtained from high resolution data of C_P and θ.

The least-squares fitting of our heat-capacity data to Eq. (3) is shown in Figure 1. Figure 1a displays the results for the entire set of data over approximately 25 K range.[9] The small rounding of our experimental data near the T_c is shown in Figure 1b. The origin of this small rounding may be caused by the existence of a small amount of impurities or optical isomers which act like impurities.[9] Two important parameters are determined in this fitting. The dimensionless parameters $t_0 = 1.42 \times 10^{-3}$ and mean-field heat-capacity jump at T_c, $\Delta C_J = 3.55 \times 10^5$ J/m^3-K. Furthermore, after subtracting C_0 from our measured heat-capacity data, one can calculate

$$\theta_c = \left[\int_T^{T_c} \frac{\Delta C_p}{T} \, dT \right]^{1/2} .$$

Then the ratios θ/θ_c are displayed in Figure 5. The fact that this ratio remains constant over more than two decades in T_c-T provides strong support for the mean-field description of the SmA-SmC* transition. Also, the constant leads to $a = 2.26 \times 10^4$ J/m^{3}K. This, combined with ΔC_J and t_0, gives $b = 2.62 \times 10^5$ J/m^3 and $c = 4.41 \times 10^6$ J/m^3.

IV. PHENOMENOLOGICAL MEAN-FIELD THEORY

Considering the symmetry change through the SmA-SmC* phase transition, Indenbom *et al.*[4] have proposed a mean-field model, which includes all the leading coupling terms among the P, θ, and q to describe the nature of this transition. This phenomenological mean-field model has the following seven expansion terms.

$$G = G_0 + \frac{1}{2} a\tilde{t}\theta^2 + \frac{1}{4} b\theta^4 + \frac{1}{2} Kq^2\theta^2 - \Lambda\theta^2 q + \frac{1}{2} P^2/\chi - fP\theta q - zP\theta. \quad (5)$$

Here K is the elastic modulus, Λ the coefficient of the Lifshitz-invariant term responsible for the helicoidal modulation, and f and z the coefficients of the flexoelectric and piezoelectric coupling between the tilt angle θ and the spontaneous polarization P. Again the coef-

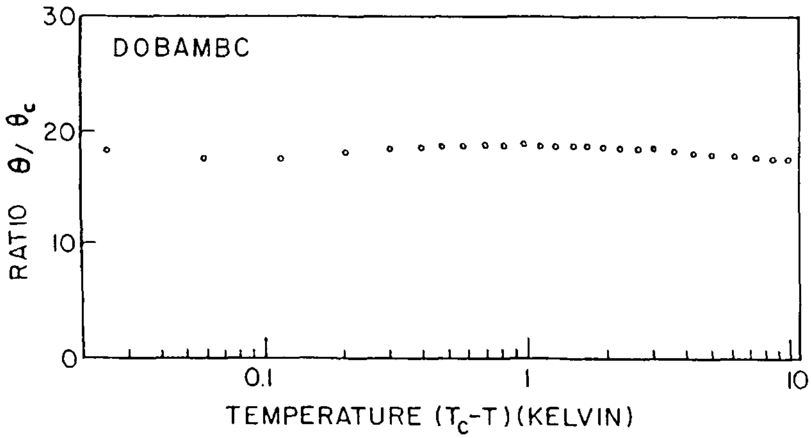

FIGURE 5 The ratio θ/θ_c versus $(T_c\text{-}T)$.

ficients a and b are positive constants and $\bar{t} = T\text{-}T_0$. T_0 is the "unrenormalized" transition temperature.

Minimizing this phenomenological free energy with respect to three variables, i.e., θ, P and q, one has the following three relations:

$$q = (\Lambda + fz\chi)/(K\text{-}f^2\chi) \tag{6}$$

$$P = \chi(fq+z)\theta \tag{7}$$

and

$$(a\bar{t} + (f^2\chi\text{-}K)q^2 - z^2\chi)\theta + b\theta^3 = 0. \tag{8}$$

The coefficient of θ term in Eq. (8) can be rewritten as a $(T\text{-}T_c)$. Then

$$T_c = T_0 + \frac{1}{a}((K\text{-}f^2\chi)q^2+\chi z^2) \tag{9}$$

Assuming that all the expansion coefficients are temperature independent, Eq. (6) indicates that the wave vector of the helicoidal modulation is constant of temperature. Then Eq. (7) suggests that P/θ is independent of temperature. We will discuss the transition temperature difference between the racemic and chiral compounds, which is related to Eq. (9), in Sec. V. Thus this phenomenological mean-field theory is insufficient to explain the temperature dependence of the helicoidal pitch[14,15] as well as the ratio P/θ.[5] In the view of the observed helicoidal pitch anomaly, at least three attempts[7,16,17] have been made to provide some theoretical understanding of the anomaly. To our knowledge, among the various theoretical attempts the generalized mean-field theory, which is proposed by Dumrongrattana and Huang[5] and is an extension of the mean-field model suggested by Zeks,[7] provides the best description of anomalies in heat capacity, tilt angle, spontaneous polarization, and helicoidal pitch.

Recently we have found that all the SmA-SmC (or SmC*) transitions are fairly closed to the mean-field tricritical point[18] such that θ^6 term becomes very important in describing the nature of the SmA-SmC (or SmC*) transition. Calculating the contribution of each term in Eq. (5) toward the total singular part of free energy at T_c-T = 5 K, we found[6] that all the additional terms involving P and/or q are at least several hundred times smaller than the three leading terms, namely, θ^2, θ^4, and θ^6. Consequently, some higher order coupling terms may be important as suggested by Zeks.[7]

V. THE GENERALIZED MEAN-FIELD THEORY

In order to explain their high resolution experimental data on heat capacity, tilt angle, and polarization near the SmA-SmC* transition of DOBAMBC, Dumrongrattana and Huang[5] have suggested the following free energy expansion to provide a very good account for their data as well as a reasonable description of the existing helicoidal pitch data.

$$G = G_0 + \frac{1}{2}a\tilde{t}\theta^2 + \frac{1}{4}b\theta^4 + \frac{1}{6}c\theta^6 + \frac{1}{2}Kq^2\theta^2 - \Lambda\theta^2 q + \frac{1}{2}P^2/\chi - fP\theta q - zP\theta - \frac{1}{2}eP^2\theta^2 + \frac{1}{4}gP^4 - dq\theta^4. \tag{10}$$

The importance of the θ^6 term has been addressed and demonstrated before. The last three terms were added by Zeks[7] in his attempt to give qualitative feature of the helicoidal pitch anomaly near the SmA-SmC* transition.

Minimizing Eq. (10) with respect to θ, P, and q and introducing new sets of parameters,[6] we obtain

$$\bar{P}^3 + \alpha\bar{P} - \beta = 0, \tag{11}$$

$$h_1(T\text{-}T_c')y + h_2y^3 + h_3y^5 - y\tilde{P}^2/3 - [h_4 + h_5y^2]\tilde{P} = 0, \quad (12)$$

and

$$q = h_6 + h_7y^2 + h_8(\tilde{P}/y). \quad (13)$$

Here two new variables $\tilde{P} = P/P_0$ and $y = \theta/\theta_1$ where $\theta_1^2 = (1/\chi - f^2/K)/e$ and $P_0 = 2\theta_1(e/3g)^{1/2}$. The new sets of parameters are $h_1 = (g/4e^2)a/\theta_1^2$; $h_2 = (g/4e^2)b(1-4\Lambda d/Kb)$; $h_3 = (g/4e^2)c\theta_1^2(1-3d^2/Kc)$; $h_4=(g/3)^{1/2}\ (z+\Lambda f/K)/(2\theta_1^2e^{3/2})$; $h_5 = (3g/e^3)^{1/2}fd/(2K)$; $h_6 = \Lambda/K$; $h_7 = d\theta_1^2/K$ and $h_8 = 2f(e/3g)^{1/2}/K$. Finally, $T_c' = T_0 + \Lambda^2/Ka$, $\alpha = 3(1-y^2)/4$ and $\beta = 3(3h_4y+h_5y^3)/4$.

The fact that the three leading terms dominate the contribution to the singular part of the free energy results in the smallness of the correction terms in h_2 and h_3, namely, $4\Lambda d/Kb$ and $3d^2/Kc$ are very small in comparison with one as well as the fitting results being very insensitive to the variation of the common factor, i.e., $g/(4e^2)$ (see Table I). Thus the original eleven expansion coefficients (a, b, c, d, e, f, g, K, χ, Λ, z) are replaced by this new set of parameters (a, b, c, $g/(4e^2)$, θ_1, P_0, h_4, h_5, h_6, h_7, h_8). Fitting to heat-capacity and tilt-angle data gives us a, b, and c. The Eqs. (11) and (12) allow us to fit P or P/θ with five adjustable parameters ($g/(4e^2)$, θ_1, P_0, h_4 and h_5). Finally the fitting to helicoidal pitch data leads to h_6, h_7, and h_8. The fitting results are shown as solid lines in Figures 1–4, and 6. Overall the fittings to heat-capacity, tilt-angle, polarization, and the ratio P/θ data are very good. In the temperature range between one and six degrees below T_c, the fitted curves are slightly below the tilt-angle data and above the polarization data. Combination of these two opposite deviations from the fitted curve produces a large deviation in the experimental data of the ratio P/θ from the fitted curve. Presently we don't have any good explanation except such a systematic deviation does not exist in our fitting to the tilt-angle and polarization data of DOBA-l-MPC (p-decyloxybenzylidene-p'-amino-l-methylpropylcinnamate) (Ref. 12). As far as the helicoidal pitch is

TABLE I

The standard deviation (σ) of our fitting to the spontaneous polarization data with various value of the common factor ($g/4e^2$) for calculation h_1, h_2, and h_3 from a', b, c, and θ_1

$g/4e^2$ in m^3/J	7×10^{-4}	2.9×10^{-5}	7×10^{-6}
σ	2.32	2.34	2.41

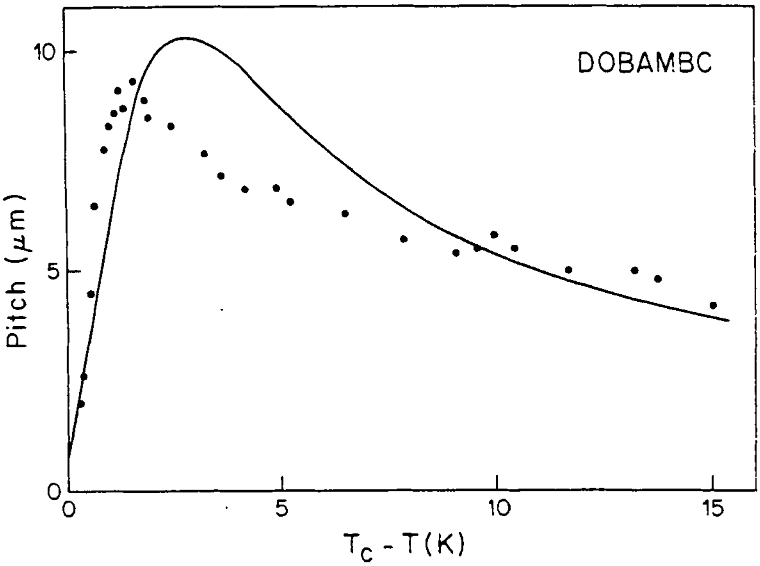

FIGURE 6 The temperature dependence of the helical pitch for DOBAMBC. Dots, experimental data; line, fitted result. Experimental results obtained by Ostrovskii *et al.* (Ref. 14) are used here.

concerned, this model only provides qualitative description of the existing data. Here we will offer two plausible explanations. First, among the reported data on helicoidal pitch of DOBAMBC there exists considerable discrepancy. The quality of the sample and the sample alignment and/or the thickness of the sample cell[19] may be the major sources of the discrepancy. Second, the generalized mean-field theory may be insufficient to describe the helicoidal pitch anomaly. Simultaneous measurements on tilt angle, polarization, and helicoidal pitch are in progress to provide a crucial test of the unique relation (i.e. Eq. 13) predicted by the theory.

Now all the expansion coefficients involving q and/or P terms can be related to the new fitting parameters and elastic constant K as follows: $\Lambda = h_6K$; $d = h_7K/\theta_1^2$; $f = h_8K\theta_1/P_0$; $e = h_7h_8K/(h_5P_0^2)$; $g = (2\theta_1)^2h_7h_8K/(3h_5P_0^4)$; $z = 2(3e^3/g)^{1/2}\theta_1^2h_4 - \Lambda f/K$, and $1/\chi = h_8K\theta_1^2(h_7/h_5 + h_8)/P_0^2$. Note that all these coefficients are proportional to the elastic constant. In principle, we can determine (g/e^2) from our fitting and obtain all the expansion coefficients. Practically, it is impossible as we discussed before. Thus in order to determine all the parameters we have to choose the most reliable experimental result to determine K. Among the experimentally determined results on f,

z (Ref. 20), K (Ref. 21), and χ (Ref. 22), we decided that χ = 2.60×10^{-11} F/m is the best one. Then the rest of the constants can be calculated and are listed in Table II with all the available experimental results. The common factor $g/(4e^2)$ has been determined through self-consistent method and is found to be 2.9×10^{-5} m^3/J. Moreover, the correction terms in h_2 and h_3 are 3.4×10^{-3} and 1.7×10^{-4}, respectively, which are much smaller than one. Finally, to check the relative importance of all the expansion terms, their contributions toward the singular part of free energy are calculated at T_c-T = 5 K and 0.48 K, respectively and are listed in Table III. Again, the dominance of the three leading expansion terms involving θ only is very clear.

With all eleven expansion coefficients being available, we can calculate temperature dependence of P and θ, in particular the ratio P/θ. Figure 7 displays the dimensionless ratio $\bar{P}/y(=(P/P_0)/(\theta/\theta_1))$ in four decades in T_c-T. It is very clear while T_c-T is small, both P and θ are small. Consequently, all four correction terms (θ^6, $P^2\theta^2$, P^4, $q\theta^4$) to the phenomenological free energy expansion are sufficiently small and can be ignored and the ratio $\bar{P}/y$ reach a constant value just as being predicted by the phenomenological mean-field model. However, the correction term θ^6 and $P^2\theta^2$ become important in comparing with θ^4 and $P\theta$ terms, respectively, in about the same temperature range (see Table III), i.e., T-$T_c \gtrsim 1$ K. Whether this is unique to DOMAMBC or fairly common among all the SmA-SmC* transitions remains to be answered by various experimental investigations.

TABLE II

The constant coefficients in the generalized mean-field model for describing the SmA-SmC* transition of DOBAMBC

Constant	Our fitted result	Other experimental result and reference		unit
χ	2.6[a]	2.6	(20)	10^{-11} F/m
K	2.5	3	(19)	10^{-12} N
Λ	2.3			10^{-5} J/m^2
d	2.5			10^{-5} J/m^2
f	−0.22	−0.4	(18)	V
e	5.7			10^{11} J-m/C^2
g	3.8			10^{19} J-m^5/C^4
z	2.8	3.4	(18)	10^6 V/m

[a]χ was chosen to be the same as the experimental result in order to determine the rest of the constants.

TABLE III

The magnitude of each individual terms in the generalized free energy at T_c-T = 5 K (case A) and 0.48 K (case B) in the unit J/m³

	Case A	Case B
$\frac{1}{2}a\bar{t}\theta^2$	8.0×10^3	1.7×10^2
$\frac{1}{4}b\theta^4$	1.2×10^3	60
$\frac{1}{6}c\theta^6$	1.9×10^3	20
$\frac{1}{2}K\theta^2q^2$	0.15	7.4×10^{-2}
$\Lambda\theta^2q$	3.1	0.47
$P^2/2\chi$	36	4.7
$fP\theta q$	3.4	0.84
$zP\theta$	45	7.6
$\frac{1}{2}eP^2\theta^2$	71	2.1
$\frac{1}{4}gP^4$	32	0.56
$dq\theta^4$	0.45	3.2×10^{-2}

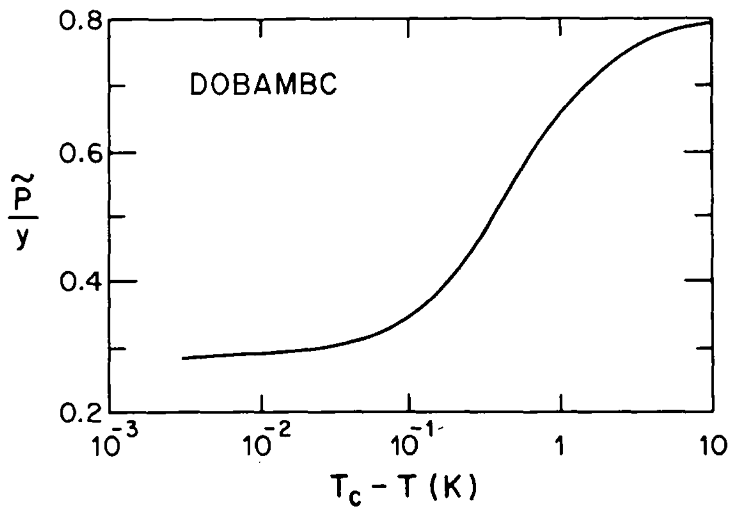

FIGURE 7 The ratio $\tilde{P}/y$ vs. T_c-T, calculated from the generalized mean-field model with the expansion coefficients determined by our fittings.

From the results in Table III, one sees that even at T_c-T = 5 K, the ratios $(1/6\, c\theta^6)/(1/4\, b\theta^4)$, and $(1/2\, eP^2\theta^2)/(zP\theta)$ are less than two. Consequently, judging from the relatively poor fitting for the helicoidal pitch data, one may argue that increasing the coefficient z by approximately two, then the biquadratic term may not be necessary in the generalized mean-field model. The failure of phenomenological mean-field model in describing both anomalies of helicoidal pitch and the ratio P/θ indicates the importance of the biquadratic terms!

Because the expression for T_c-T_0 is determined by the coefficients of θ^2 terms and all four additional terms in the generalized mean-field model are higher order terms in θ, this model should have the same expression for T_c-T_0 as the phenomenological mean-field model. Based on the available expansion coefficients, we can calculate the difference T_c-T_0 (see Eq. 9). The result is T_c-T_0 = 5 mK. On the other hand, the measured transition temperature difference between the chiral compound (SmA-SmC* transition) and the racemic compound (SmA-SmC transition) is about 0.7 K.[23] The large difference between our calculated value and the experimental value may be due to the impurities effect in the racemic compound, which slightly suppresses the transition temperature.

Finally one can show that Eq. (4) holds in general for the following singular part of free energy expansion

$$G_S = \frac{1}{2} a' t\theta^2 + \Delta G(\theta, \{x\}).$$

Here $\Delta G(\theta,\{x\})$ is all higher order expansion terms with temperature independent expansion coefficients and x is any set of relevant physical parameters characterizing the phase transition, besides the primary order parameter θ.

In this conference, Goodby *et al.*[24] have reported a cross-over of the sign of the spontaneous polarization in the SmC* phase of some biphenyl esters. Because flexoelectric coefficient f is normally negative, the parameter β in Eq. 11 may change sign at a temperature T_1 provided that normalized tilt angle (y) becomes sufficiently large. Under this circumstance the polarization will have an abrupt jump from $+P_1$ to $-P_1$ as the temperature being cooled through T_1 and the tilt angle does not change sign. In the case of DOBAMBC, the parameter β and polarization would change sign if the tilt angle reached 0.59 rad. However, this will not happen before the sample undergoes the SmC*-SmI* transition. Now let us get back to the case of biphenyl esters. The fact that the polarization does not change abruptly and has a positive temperature derivative over more than 10K range suggests that our generalized mean-field model is not applicable to the unique feature associated with biphenyl esters.

VI. CONCLUSIONS

Here we present our experimental results on the heat capacity, tilt angle and polarization near the SmA-SmC* transition of DOBAMBC. The anomalous behavior of the ratio P/θ which we have found in DOBAMBC seems to be fairly common among the compounds with the SmA-SmC* transition. A generalized mean-field model has been proposed to give fairly good account for all the anomalies near the SmA-SmC* transition. Recently, we have demonstrated that Eqs. 3, 11, and 12 again give very good fitting results to our heat-capacity, tilt-angle, and polarization data near the SmA-SmC* transition of DOBA-1-MPC (*p*-decyloxybenzylidene-*p*'-amino-1-methylpropylcinnamate).[12] Further experimental effort on the simultaneous measurement of P, θ, and q on the compound DOBAMBC as well as the other chiral-smectic-C materials is in progress.

Acknowledgment

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