Pressure effect on the smectic-*A*–isotropic phase transition

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We examine the effect of pressure on the smectic-*A*–isotropic phase transition within the Landau phenomenological theory. The influence of pressure on the smectic-*A*–isotropic phase transition is discussed by varying the coupling between the orientational and the translational order parameter. The transition is found to be of first order even at elevated pressure. The pressure dependence nonlinear dielectric effect in the isotropic phase of the smectic-*A*–isotropic transition is calculated. The theoretical results are in good qualitative agreement with available experimental results.

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I. INTRODUCTION

While the research on the nematic-isotropic (*N*-*I*) phase transition has been going on over the past few decades, there is increasing interest in the smectic-*A*–isotropic (Sm-*A*–*I*) phase transition recently [1–10]. The first order behavior of the Sm-*A*–*I* transition was probably the first quantitative test in Kerr effect studies on dodecyl-alkylcyanobiphenyl (12CB) by Coles and Strazielle [11]. They showed the existence of the smectic-*A* (Sm-*A*) type cybotactic groups of molecules in the isotropic phase, which explain the high nonlinearity near the Sm-*A*–*I* transition. The strong first order character of the Sm-*A*–*I* transition was also confirmed on the investigations of the free surface in the isotropic phase of 12CB by Ocko *et al.* [12] and the measurement of strain birefringence induced by mechanical stress in side-chain elastomers by Olbrich *et al.* [5]. Recently the quantitative characteristic of the Sm-*A*–*I* transition delivered a series of static linear and nonlinear dielectric permittivity investigations in the homologous series of *n*-alkylcyanobiphenyls (*n*CB from $n=4-12$) and *n*-alkyl-2-4-(isothiocyphenyl-dioxabe, $n=4,6,8$) carried out by Drozd-Rzoska and co-workers [1–4]. The clear evidence for the pretransitional anomaly was obtained whose form was the same both for the *N*-*I* and Sm-*A*–*I* transition. Regarding the discontinuity of the Sm-*A*-*I* transition, it increases with increasing length of the molecule: in *n*CB from $\Delta T\approx 0.5$ K (4CB, *N*-*I* transition) to $\Delta T\approx 6.5$ K (12CB, Sm-*A*–*I* transition). The applied quasicritical, fluidlike equations give the exponents $\alpha\approx 0.5$ and $\gamma=1$ in each mentioned case even at the high pressure. It was further found that pressure reduces the discontinuity of the Sm-*A*–*I* transition, contrary to the *N*-*I* transition. It is likely that it is associated with the pressure induced nematic phase.

Theoretical studies [6–10,13,14] indicate that a direct Sm-*A*–*I* transition is possible. The first theoretical description of the Sm-*A*–*I* transition on a lattice model was developed by Ronis and Rosenblatt [13]. In a separate paper [14] Rosenblatt and Ronis pointed out that in the presence of an intense magnetic filed, materials can exhibit an intermediate nematic phase undergoing a direct Sm-*A*–*I* transition in the zero field limit. Recently, Mukherjee *et al.* [6] showed that the basic features of the Sm-*A*–*I* transition, particularly the relatively large value of the discontinuity, can be obtained within a simple Landau model. In this model they explained all the key features of the Sm-*A*–*I* transition. Brand *et al.* [7] studied the macroscopic dynamics behavior in the vicinity of the Sm-*A*–*I* transition. They calculated the macroscopic dynamic equations on the isotropic side as well as on the Sm-*A* side of the phase transition incorporating the effect of an external field.

In spite of these theoretical efforts, there remain still a number of key questions concerning the properties of the Sm-*A*–*I* transition. It is worthwhile here to point out that the Landau model presented in Ref. [6] is able to explain some of the main experimental observations at ambient pressure. There is no theoretical study on the pressure effect on the Sm-*A*–*I* phase transition. In contrast, experiments [1–3] show a one-to-one comparison between the *N*-*I* and the Sm-*A*–*I* transitions at high pressure. Although, some theoretical models [15,16] on the pressure effect in the *N*-*I* transition are available, there is no such study on the Sm-*A*–*I* transition.

The purpose of the present paper is to study the pressure effect on Sm-*A*–*I* transition within the Landau phenomenological theory. We calculate the pressure dependence nonlinear dielectric effect (NDE) in the isotropic phase of the Sm-*A*–*I* transition.

II. THEORY

Our aim is to calculate the pressure dependence of various thermodynamic quantities at the Sm-*A*–*I* transition. We start by using a standard Landau expansion for the free energy [6] in powers of the order parameters. Keeping homogeneous terms up to the quartic order and gradients only up to the relevant order, Gibbs free energy (g) near the Sm-*A*–*I* transition can be written as

$$g(P,T)=g_0(P,T)+\frac{1}{2}AQ_{ij}Q_{ij}-\frac{1}{3}BQ_{ij}Q_{jk}Q_{ki}+\frac{1}{4}C(Q_{ij}Q_{ij})^2+\frac{1}{2}\alpha|\psi|^2+\frac{1}{4}\beta|\psi|^4+\frac{1}{2}\delta|\psi|^2Q_{ij}Q_{ij}+\frac{1}{2}b_1|\nabla_i\psi|^2+\frac{1}{2}b_2|\Delta\psi|^2$$

$$+\frac{1}{2}eQ_{ij}(\nabla_i\psi)(\nabla_j\psi^*), \tag{1}$$

where $g_0(P,T)$ is the free energy of the isotropic phase. A, B, C, α, β, δ, b_1, b_2, and e are material parameters. All parameters are assumed to be a function of pressure. $Q_{ij}=\frac{1}{2}S(3n_in_j-\delta_{ij})$ is a symmetric traceless tensor as was originally proposed by de Gennes and Prost [17]. The quantity S defines the strength of the nematic ordering. The smectic order parameter $\psi(\vec{r})=\psi_0\exp(-i\phi)$ is a complex scalar quantity whose modulus ψ_0 is defined as the amplitude of a one-dimensional density wave characterized by the phase ϕ. δ is a biquadratic coupling constant. There is no coupling term $|\psi|^2Q_{ij}$ in the free energy (1) since such a term would preclude the existence of the isotropic phase. The gradient term associated with coefficient b_2 favors ψ the spatial modulation of wave-vector magnitude q_0. The gradient term associated with e governs the relative direction of the layering with respect to the director. The negative value of e favors the Sm-A phase over the nematic phase. The material parameters A and α can be assumed as $A=a[T-T^*_{N\text{-}I}(P)]$ and $\alpha=\alpha_0[T-T^*_{\text{Sm-}A-I}(P)]$. $T^*_{N\text{-}I}$ and $T^*_{\text{Sm-}A-I}$ are the supercooling temperatures. a and α_0 are constants.

From the experimental phase diagrams [1,18] one observes at elevated pressure, $T^*_{N\text{-}I}(P)$ and $T^*_{\text{Sm-}A-I}(P)$ can be portrayed as

$$T^*_{N\text{-}I}(P)=T^0_{N\text{-}I}+fP, \tag{2a}$$

$$T^*_{\text{Sm-}A-I}(P)=T^0_{\text{Sm-}A-I}+hP, \tag{2b}$$

where f and h are constants.

For the determination of the thermodynamic quantities near the Sm-A–I transition we consider a spatially uniform system in which the order parameter values are spatially invariant. This means that we will now discard the spatial derivative terms of Eq. (1). Substitution of the values of the order parameters Q_{ij} and ψ into Eq. (1) gives

$$g(P,T)=g_0(P,T)+\frac{3}{4}AS^2-\frac{1}{4}BS^3+\frac{9}{16}CS^4+\frac{1}{2}\alpha\psi_0^2+\frac{1}{4}\beta\psi_0^4+\frac{3}{4}\delta\psi_0^2S^2. \tag{3}$$

To ensure the stability of the isotropic phase at high pressure, the condition $\beta C-\delta^2>0$ should be fulfilled. It is clear that $S\neq0$ and $\psi_0\neq0$ are the equilibrium conditions realized in the Sm-A phase. The negative value of δ favors the Sm-A phase over the nematic phase. In order to ensure the condition of the Sm-A phase to be stable, it is required that

$$\frac{\partial^2g}{\partial S^2}\frac{\partial^2g}{\partial\psi_0^2}-\left(\frac{\partial^2g}{\partial S\partial\psi_0}\right)^2=9S^2\psi_0^2\left(\Delta-\frac{B\beta}{6S}\right)>0, \tag{4}$$

where $\Delta=\beta C-\delta^2$. Equation (4) shows that $\Delta>B\beta/6S$ for the stability of the Sm-A phase. Minimization of Eq. (3) with respect to ψ_0 gives

$$\psi_0^2=-\frac{1}{\beta}\left(\alpha+\frac{3}{2}\delta S^2\right). \tag{5}$$

By substituting the solution of Eq. (5) into Eq. (3), we get the Gibbs free energy expansion for the Sm-A–I transition as a function of S alone, which can be written as

$$g(P,T)=g_0(P,T)-\frac{\alpha^2}{4\beta}+\frac{3}{4}A^*S^2-\frac{1}{4}BS^3+\frac{9}{16}C^*S^4, \tag{6}$$

where the renormalized coefficients are

$$A^*=A-\frac{\delta\alpha}{\beta}, \tag{7a}$$

$$C^*=C-\frac{\delta^2}{\beta}. \tag{7b}$$

At the Sm-A–I transition there are two minima ($\partial g/\partial S=0$) at the same free energy g. One is a trivial one (the isotropic phase) and the other has $S>0$ (the Sm-A phase). The jump of the orientational order parameter at the Sm-A–I transition is given by

$$S_{\text{Sm-}A-I}=\frac{2B}{9C^*}. \tag{8}$$

The Sm-A–I transition temperature is given by

$$T_{\text{Sm-}A-I}=T^{01}_{\text{Sm-}A-I}+mP, \tag{9}$$

where

$$T^{01}_{\text{Sm-}A-I}=\frac{[\delta T^0_{\text{Sm-}A-I}-\delta_0(T^0_{N\text{-}I}+B^2/27aC^*)]}{(\delta-\delta_0)}, \tag{10a}$$

$$m=\frac{(\delta h-\delta_0 f)}{(\delta-\delta_0)}. \tag{10b}$$

Similarly, the Sm-A–I transition pressure reads

$$P_{\text{Sm-}A-I}=-T^{02}_{\text{Sm-}A-I}+m^{-1}T, \tag{11}$$

where

$$T^{02}_{\text{Sm-}A-I}=\frac{[\delta T^0_{\text{Sm-}A-I}-\delta_0(T^0_{N\text{-}I}+B^2/27aC^*)]}{(\delta h-\delta_0 f)}, \tag{12}$$

where $\delta_0=a\beta/\alpha_0$. Since $C^*<C$, Eq. (8) shows that the jump of the orientational order parameter $S_{\text{Sm-}A-I}$ at the Sm-A–I transition is larger than that at $T_{N\text{-}I}$ ($S_{N\text{-}I}=2B/9C$) even at elevated pressure. Equation (9) predicts that the Sm-A–I transition temperature increases with the pressure and is always higher than that the N-I transition temperature ($T_{N\text{-}I}=T^0_{N\text{-}I}+B^2/27aC+fP$). The orientational order parameter in the Sm-A phase can be expressed as

$$(S-S^{+})^2=(S^{+})^2-\frac{2a}{3C^*\delta_0}[(\delta_0-\delta)T-(\delta_0 f-\delta h)P-\delta_0 T^0_{N\text{-}I}+\delta T^0_{\text{Sm-}A\text{-}I}], \tag{13}$$

where $S^{+}=B/6C^*$ is the order parameter in the superheated Sm-A phase. Thus S decreases with the pressure in the Sm-A phases. When S is fixed, Eq. (13) may be rewritten as

$$T=T_1+mP, \tag{14}$$

where

$$T_1=T_1(S)=\frac{1}{(\delta_0-\delta)}\left[\delta_0 T^0_{N\text{-}I}-\delta T^0_{\text{Sm-}A\text{-}I}-\frac{3C^*\delta_0}{2a}\times S(S-2S^{+})\right]. \tag{15}$$

Differentiating Eq. (6) with respect to P one has the volume in the Sm-A phase,

$$V_{\text{Sm-}A}=V_I-\frac{3a}{4\delta_0}(\delta_0 f-\delta h)S^2(T,P), \tag{16}$$

where $V_I=(\partial/\partial P)(g_0-\alpha^2/4\beta)$ is the volume in the isotropic phase. Therefore the volume change at the Sm-A–I transition point reads as

$$\Delta V_{\text{Sm-}A\text{-}I}=\frac{3a}{4\delta_0}(\delta_0 f-\delta h)S^2_{\text{Sm-}A\text{-}I}. \tag{17}$$

The enthalpy change at $T_{\text{Sm-}A\text{-}I}$ is given by

$$\Delta H_{\text{Sm-}A\text{-}I}=\frac{aB^2}{27\delta_0 C^{*2}}(\delta_0-\delta)T_{\text{Sm-}A\text{-}I}. \tag{18}$$

Equation (18) shows that the jump of the latent heat of transition at the Sm-A–I transition is also higher than that the N-I transition ($\Delta H_{N\text{-}I}=(aB^2/27C^2)T_{N\text{-}I}$). We see from Eqs. (17) and (18) that discontinuity decreases with the rise of pressure, since the coefficients B and C change with the rise of pressure, which does not contradict experimental observations [1]. The above discussion shows that the Sm-A–I transition is more strongly of first order than the N-I transition even at the elevated pressure.

III. HEAT CAPACITY AND NONLINEAR DIELECTRIC EFFECT IN THE ISOTROPIC PHASE OF THE Sm-A–I TRANSITION

In this section we will calculate the pressure dependence of the heat capacity and the NDE in the isotropic phase of smectogenic liquid crystals. The Gibbs free energy density associated with the long wavelength part of the orientational order parameter fluctuation in the isotropic phase can be expanded as

$$g=g_0+\frac{1}{2}AQ_{ij}Q_{ij}-\frac{1}{3}BQ_{ij}Q_{jk}Q_{ki}+\frac{1}{4}C(Q_{ij}Q_{ij})^2+\frac{1}{2}\alpha|\psi|^2+\frac{1}{4}\beta|\psi|^4+\frac{1}{2}\delta|\psi|^2Q_{ij}Q_{ij}+\frac{1}{2}D(\nabla_i Q_{kl})^2-\frac{\varepsilon_0\Delta\varepsilon_0}{3}Q_{ij}E_iE_j, \tag{19}$$

where $A=a(T-T^0_{N\text{-}I}-fP)$, $\alpha=\alpha_0(T-T^0_{\text{Sm-}A\text{-}I}-hP)$, ϵ_0 is vacuum permittivity, $\Delta\epsilon_0$ is the anisotropy of the dielectric permittivity, and β, δ are functions of pressure. Substituting the value of Q_{ij} in Eq. (19) and transforming the free energy (19) as a function of S only we find

$$g=g_0-\frac{\alpha^2}{4\beta}+\frac{3}{4}A^*S^2-\frac{1}{4}BS^3+\frac{9}{16}C^*S^4+\frac{3}{4}D(\nabla S)^2+\frac{9}{4}DS^2(\nabla_i n_k)^2-\eta E^2S, \tag{20}$$

where A^* and C^* are same as in Eq. (7a) and Eq. (7b), respectively, and $\eta=\varepsilon_0\Delta\varepsilon_0/3$. The order parameter induced by a electric field in the isotropic phase is calculated to a first approximation ($B=0$, $C=0$, and $D=0$) from Eq. (20) and can be expressed as

$$S(E)=\frac{2\eta E^2}{3A^*}. \tag{21}$$

The NDE describes the shift of the anisotropy of the dielectric permittivity caused by a strong electric field. To a large extent the NDE is analogous to the Kerr effect for radio frequencies [19,20]. An anisotropy property is proportional to the induced order. Hence the dielectric permittivity in the isotropic phase can be expressed as [20,21]

$$\Delta\varepsilon(E)=\varepsilon(E)-\varepsilon=\Delta\varepsilon_f S(E), \tag{22}$$

where $\varepsilon(E)$ and ε are dielectric permittivities in a strong (E) and weak (measuring) electric field. $\Delta\varepsilon_f$ denotes the anisotropy of the dielectric permittivity for the given frequency f. Combining Eqs. (21) and (22) we find

$$\varepsilon_{NDE}=\frac{\Delta\varepsilon(E)}{E^2}=\frac{\gamma}{(T-T_0-mP)}, \tag{23}$$

where

$$\gamma=\frac{2\beta\epsilon_0\Delta\varepsilon_0\Delta\varepsilon_f}{9\alpha_0}\frac{1}{(\delta_0-\delta)}, \tag{24a}$$

$$T_0=(\delta_0 T^0_{N\text{-}I}-\delta T^0_{\text{Sm}A\text{-}I})/(\delta_0-\delta), \tag{24b}$$

where m is same as that given by Eq. (10b). From an alternative point of view, we can take $A=a(T)(P^*_{N\text{-}I}-P)$ and $\alpha=\alpha_0(T)(P^*_{\text{Sm-}A\text{-}I}-P)$ and regard β, δ, and P^* as functions of temperature. $P^*_{N\text{-}I}$ and $P^*_{\text{Sm-}A\text{-}I}$ are supercooling pressures. In this case ε_{NDE} can be expressed as

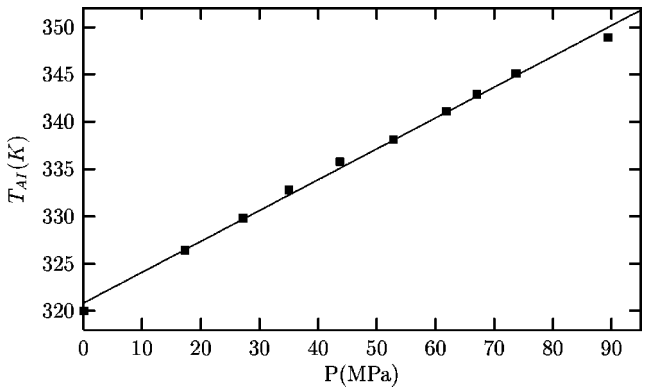

FIG. 1. The pressure dependence of the Sm-*A*-*I* transition temperature of 10CB. The measured data (square) are from Ref. [1], and the line is the best fit of Eq. (9).

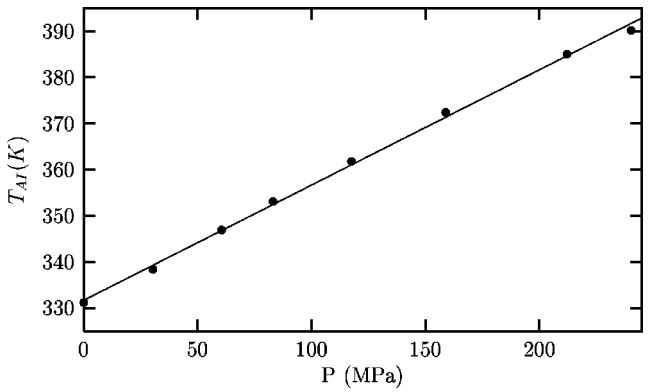

FIG. 2. The pressure dependence of the Sm-*A*-*I* transition temperature of 12CB. The measured data (circle) are from Ref. [1], and the line is the best fit of Eq. (9).

$$\varepsilon_{NDE}=\frac{\Delta\varepsilon(E)}{E^2}=\frac{\gamma}{(P^*-P)}, \tag{25}$$

where $P^*=(\delta_0 P^*_{N\text{-}I}-\delta P^*_{\text{Sm-}A-I})/(\delta_0-\delta)$ is the supercooling pressure.

For calculating the heat capacity in the isotropic phase of the Sm-A–I transition we consider the same free energy Eq. (19) but with zero electric field ($E=0$). Applying the same method as adopted by Imura and Okano [22] and Mukherjee and Deutsch [23], the excess heat capacity in the isotropic phase (above the Sm-A–I transition) due to the fluctuation is given by

$$\Delta C_P=ET^2(T-T_0-mP)^{-1/2}, \tag{26}$$

where $E=(3ak_B/64\pi D)[a\delta_0/D(\delta_0-\delta)]^{1/2}$. k_B is the Boltzmann constant.

IV. COMPARISON WITH EXPERIMENT

In this section we will compare some of the theoretical results presented in the preceding sections with the available experimental results. For the $T_{\text{Sm-}A-I}$ vs P phase diagram of 10CB and 12CB, there exists only one experiment by Drozd-Rzoska *et al.* [1]. The experimental phase diagrams for $T_{\text{Sm-}A-I}$ vs P in Figs. 5 and 6 of Ref. [1] is a straight line. From Eq. (14), when S is fixed, T vs P should be a straight line, which agrees with Figs. 5 and 6 of the above reference. Hence, in principle, Eqs. (9) and (14) are same. We have, therefore, fitted Eq. (9) with the experimental data for 10CB and 12CB of Drozd-Rzoska *et al.* [1]. The fit (line) and the measured data for 10CB and 12CB are shown in Figs. 1 and 2. The fits to the measured values are found to be good. Because of the large number of unknown parameters in Eq. (9) we take $T^{01}_{\text{Sm-}A-I}$ and m as a fit parameters. The values obtained for the fit parameters are $T^{01}_{\text{Sm-}A-I}=320.84$ K, $m=0.32$ K/MPa (for 10CB) and $T^{01}_{\text{Sm-}A-I}=331.68$ K, $m=0.25$ K/MPa (for 12CB). From the slope of the fitted curves we obtain $dT_{\text{Sm-}A-I}/dP=0.32$ K/MPa (10CB) and $dT_{\text{Sm-}A-I}/dP=0.25$ K/MPa (12CB), close, not surprisingly, to the experimental values 0.31 K/MPa and 0.27 K/MPa, respectively. Experiment shows that $dT_{N\text{-}I}/dP > dT_{\text{Sm-}A-I}/dP$. The experimental value of $dT_{N\text{-}I}/dP$ for 6CB gives [18] $dT_{N\text{-}I}/dP=0.36$ K/MPa. Thus we always find $dT_{N\text{-}I}/dP>dT_{\text{Sm-}A-I}/dP$. This can also easily be verified from Eq. (9), where we get $dT_{\text{Sm-}A-I}/dP=m$, whereas $dT_{N\text{-}I}/dP=f$. Since the value of m [Eq. (10b)] is smaller than the value of f, we find $dT_{N\text{-}I}/dP>dT_{\text{Sm-}A-I}/dP$. The expressions (13), (16), and (18) cannot be fitted with experimental data since the pressure dependences of such data are not available in the literature. However the value of the latent heat of transition at the Sm-A–I transition is always higher than that the N-I transition, which confirms the experiment and is also satisfied by the relation (18). The NDE expressions (23) and (25) can easily be verified with Fig. 4 of Drozd-Rzoska *et al.* [1]. In their measurement they obtained the exponent $\gamma'=1$. The NDE expressions (23) and (25) also give the exponent $\gamma'=1$. Furthermore, to check Eq. (23), ε_{NDE} vs P of Drozd-Rzoska *et al.* [1] for constant temperature is plotted in Fig. 3. The form of Eq. (23) shows that there are eight unknown phenomenological parameters. It is unphysical to take all these parameters as fit parameters while fitting Eq. (23). Unfortunately, to the best of the author's knowledge there are still no experimental estimations of the anisotropy of dielectric permittivity $\Delta\varepsilon_0$, even for

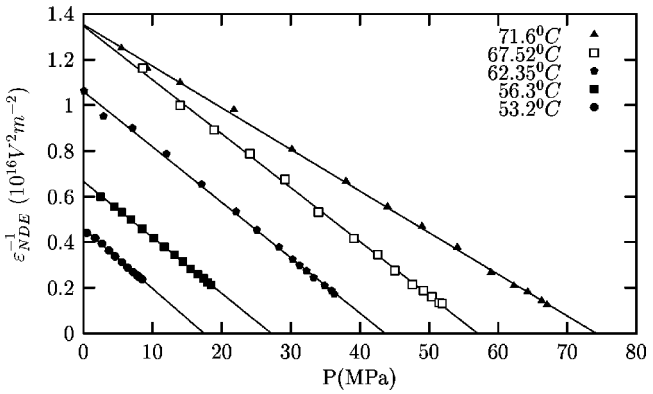

FIG. 3. The pressure dependence of the reciprocals of NDE in the isotropic phase of 10CB. The measured data are from Ref. [1], and the line is the best fit of Eq. (23).

TABLE I. Values of the experimental temperatures (T) and the corresponding various fitted parameters in the isotropic phase of the Sm-A-I transition in 10CB, as derived from a fit of Eq. (23) to the measured data in Ref. [1].

T (°C)	γ (10^{-16} $m^2\,V^{-2}\,K$)	T_0 (°C)	m (°C/MPa)
53.2	53.90	28.61	1.40
56.3	50.20	22.84	1.23
62.35	44.28	15.35	1.07
67.52	41.08	12.11	0.97
71.6	43.34	12.91	0.79

classic smectogens such as 10CB and 12CB. This can be related to essential problems by inducing preferably orientational ordering in the Sm-A phase. Hence, a reliable estimation of the coefficients a, α_0, β, δ, f, and h is not possible. We have, therefore, fitted Eq. (23) with the measured $\varepsilon_{NDE}^{-1}(P)$ data using γ, T_0, and m as a fit parameters. The line in Fig. 3 is the best fit of the reciprocal of the NDE (ε_{NDE}^{-1}) vs P of Eq. (23) for the different constant temperatures. As can be observed, the agreement of the measured data with Eq. (23) is extremely good considering the experimental error. The values of the fitted parameters are listed in Table I. For comparison, we have also fitted Eq. (25) with the same measured $\varepsilon_{NDE}^{-1}(P)$ data. Equation (25) yields a line that overlaps our solid line everywhere in Fig. 3 with different parameter values. While examining Eq. (26) we see that $d\varepsilon_P/dT \propto \Delta C_P \propto [T-T^*(P)]^{-\alpha'}$. The dielectric test measurement [1–3] on the isotropic phase of the Sm-A–I transition gave a clear evidence for the critical exponent $\alpha \approx 0.5$, which is same as the value in the isotropic phase of the N-I transition. Equation (26) gives $\alpha = 0.5$, which agrees fairly with experiment. We also observe that the amplitude of the orientational order parameter fluctuation increases abnormally near $T_{\text{Sm-}A-I}$ and brings about the anomalous increments in heat capacity. Thus we point out the same pretransitional phenomena observed in the isotropic phase of the Sm-A–I transition. This may be caused because the dielectric method or the heat capacity measurement probably detect only the orientational ordering.

V. CONCLUSIONS

We have presented here a Landau theory analysis to describe the pressure effect on the Sm-A–I transition. The effect of pressure on the Sm-A–I transition is to increase the transition temperature. But the pressure decreases the discontinuity of the transition. The theory predicts a first order character of the Sm-A–I transition even at high pressure. The same pretransitional phenomena is observed in the isotropic phase of the Sm-A–I transition similar to the N-I transition. The critical exponent $\alpha' = 0.5$ and $\gamma' = 1$ indicate the fluidlike analogy in the isotropic phase of the Sm-A–I transition. Although we have made some progress in comparing the theoretical results with available experimental results, there is still lack of basic data that would make possible a more complete comparison with the theory and the quantitative estimation of the phenomenological parameters.

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