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Short Communication

Characterizing the Nature of the Smectic A–Smectic C and Smectic A–Smectic C* TransitionsL. Benguigui ⁽¹⁾ and P. Martinoty ^(2,*)⁽¹⁾ Solid State Institute and Department of Physics, Technion-Israel Institute of Technology, 32000 Haifa, Israel⁽²⁾ Laboratoire d'Ultrasons et de Dynamique des Fluides Complexes (**), Université Louis Pasteur, 4 rue Blaise Pascal, 67070 Strasbourg Cedex, France*(Received 01 October 1996, revised 22 November 1996, accepted 16 December 1996)*

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Abstract. — We provide a theoretical explanation of the critical (*i.e.* non-mean-field) behavior of specific heat at the SmA–SmC transition, as it has recently been observed on certain compounds. This explanation rests on the Andereck and Swift free energy introducing terms which couple the order-parameter with both layer-compression and density. We show that the behavior of specific-heat depends on the intensity of these couplings, and that the behaviors which can be observed are therefore of the mean-field, Gaussian or 3DXY type.

Résumé. — Nous donnons une explication du comportement critique (c-à-d. non-champ moyen) de la chaleur spécifique, qui a été observé récemment sur certains composés. Cette explication repose sur l'énergie libre de Andereck et Swift qui introduit des termes couplant le paramètre d'ordre avec la compression de couche et la densité. Nous montrons que le comportement de la chaleur spécifique dépend de l'intensité de ces couplages et que les comportements qui peuvent être observés sont par suite de type champ moyen, gaussien ou 3DXY.

Two recent papers [1,2] give important information, concerning the nature of the smectic C–smectic A transition: through measurements of the specific heat, these results on two different compounds indicate a critical (*i.e.* a non-mean-field) behavior. From the symmetry of the order parameter, it was predicted by de Gennes [3] that this transition must belong to the universality class of the superfluid helium transition, the 3DXY model. However, a large amount of experimental data [4] have been well-described by the Landau theory with a sixth-order term, implying that this transition is near a tricritical point. In references [1] and [2], it was claimed that it was very difficult to explain these different behaviors for the same transition: meanfield or critical transition. In this report we present a very simple analysis which permits to explain these different behaviors of the specific heat. This analysis is based on the calculations

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which were developed in reference [5] to show that the ultrasound measurements are more sensitive to fluctuation effects than the specific heat measurements.

One possibility for the appearance of a tricritical point is the coupling of the order parameter with the strains. It is well-known that the quadratic coupling of the strains with the order parameter can bring a second order transition to a first order one [6]. Depending on the strength of the coupling, various possibilities may occur: second order transition, tricritical point or first order transition.

In the case of the SmA-SmC transition, Andereck and Swift [7] introduced terms in the free energy coupling quadratically the smectic C order parameter to the density and to the strain $x_3 = \partial u / \partial z$ (u is the deformation and the direction z is perpendicular to the layers). Their free energy used to study the behavior of sound attenuation and dispersion, can be written in the following form (with the notation of Ref. [5]):

$$F = \int d^3r \left[\frac{1}{2}a|\eta|^2 + \frac{1}{4}b|\eta|^4 + \frac{1}{2}C_{11}(x_1 + x_2)^2 + \frac{1}{2}C_{33}x_3^2 + C_{13}(x_1 + x_2)x_3 + \frac{1}{2}\gamma_u x_3 |\eta|^2 - \frac{1}{2}\gamma_\rho (x_1 + x_2 + x_3)|\eta|^2 + \frac{1}{2}m_\perp (\nabla_\perp \eta)^2 + \frac{1}{2}m_z (\nabla_z \eta)^2 \right] \quad (1)$$

where η is the smectic C order parameter. x_1 and x_2 are defined by $x_1 = \partial u / \partial x$ and $x_2 = \partial u / \partial y$. $m_\perp$ (the symbol $\perp$ refers to the plane of the layers), m_z and b are positive constants and $a = a_0(T - T_c)$. C_{11} , C_{33} and C_{13} are the elastic constants of the SmA-phase. γ_ρ and γ_u are parameters which couple the smectic C order parameter to the density variation and to the layer-spacing gradient. However this free energy is not suitable for analysis of the thermal properties (like the specific heat) since in that case it is the Gibbs free energy $G = F - \Sigma X_i \cdot x_i$ (the X_i 's are the stresses) which is relevant. As shown in reference [5], G is given by

$$G = \int d^3r \left[\frac{1}{2}a|\eta|^2 + \frac{1}{4}b^*|\eta|^4 \right] \quad (2)$$

where

$$b^* = b[1 + \chi] \quad (3)$$

and

$$\chi = \frac{2C_{13}\gamma_\rho(\gamma_\rho - \gamma_u) - C_{11}(\gamma_\rho - \gamma_u)^2 - C_{33}\gamma_\rho^2}{2[C_{11}C_{33} - C_{13}^2]}. \quad (4)$$

The key result for the following is that χ , and consequently b^* , is function of the parameters γ_u and γ_ρ . These two parameters are related to the jump Δ of the elastic constants at T_c by the equations

$$\begin{aligned} \Delta C_{11} &= -\frac{\gamma_\rho^2}{2b} \\ \Delta C_{13} &= -\frac{\gamma_\rho(\gamma_\rho - \gamma_u)}{2b} \\ \Delta C_{33} &= -\frac{(\gamma_\rho - \gamma_u)^2}{2b} \end{aligned} \quad (5)$$

where

$$\Delta C_{ij} = C_{ij}(T_c^-) - C_{ij}(T_c^+).$$

These relations allow χ to be rewritten as a function either of the ΔC_{ij} or of the de Gennes elastic constants $A = C_{11}$, $B = C_{11} + C_{33} - 2C_{13}$ and $C = C_{11} - C_{13}$.

$$\chi = \frac{\Delta(C_{11}C_{33} - C_{13}^2)}{C_{11}C_{33} - C_{13}^2} \quad (6a)$$

$$\chi = \frac{\Delta (AB - C^2)}{AB - C^2} \quad (6b)$$

The χ value can be determined experimentally since the elastic constants can be deduced from the velocity measurements through the relationship [8]

$$\rho V^2(\theta) = A - 2B\cos^2\theta + C\cos^4\theta \quad (7)$$

where θ is the angle between the normal to the smectic layers and the sound propagation direction.

Inspection of formulas (4) and (6b) shows that χ is always negative and given with a good approximation (because $A \gg B, C$ [9]) by $\Delta B/B$. Since B is always positive, $|\chi|$ is smaller than one. As a consequence b^* cannot be negative but the possibility that b^* may be small remains. Since χ is a function of the coupling parameters γ_u and γ_ρ , b^* can vary from a value near zero (and the transition will be almost a tricritical point) to a finite value. It means that the quadratic coupling cannot transform the order of the transition, contrarily to other examples of phase transitions (in ferroelectricity or magnetism) [6]. This unexpected result is due to the layer structure of the smectic A phase, responsible for the very particular properties of this phase.

To determine the nature of a second order transition, it is usual to calculate the width of the critical region with the help of the Ginsburg criterion. This width is given by [5]:

$$\Delta T_c = \frac{k^2 T_c}{(16\pi)^2 (\Delta C_p)^2 \xi_{0\perp}^4 \xi_{0\parallel}^2} \quad (8)$$

where $\xi_{0\perp}$ and $\xi_{0\parallel}$ are the bare correlation lengths for order fluctuations perpendicular and parallel to the director respectively, and ΔC_p the jump of the specific heat. This jump, deduced from equation (2), is given by:

$$\Delta C_p = C_p(T_c^-) - C_p(T_c^+) = \frac{T_c a_0^2}{2b^*} \quad (9)$$

Since $\Delta C_p \sim 1/b^*$, the width of the critical region is proportional to $(b^*)^2$. Thus a small change in b^* , may bring an important variation of ΔT_c and this explains the diversity of behaviors reported for the SmA–SmC transition. If b^* is very small, ΔT_c will be also small such that the specific heat behavior will be in accordance with the mean field theory of the tricritical point. However, if b^* is large enough, ΔT_c will get a significant value and deviations from the mean field theory will be detected giving Gaussian (as in [2]) or 3DXY (as in [1]) behaviors.

The critical or non critical nature of the transition is detected in a decisive manner by measuring the elastic constants through ultrasonic measurements. The relevant free energy for the C_{ij} 's is (1) and the width of the critical region is now proportional to b^2 which is always larger than $(b^*)^2$ [5]. It is why the compounds TBBA and 8S5 have a mean-field-like behavior from the specific heat measurements [10, 11], but distinct critical behavior from the ultrasonic measurements [12, 13].

We mentioned above that the mechanism of Andereck and Swift cannot bring the transition to be first order. However, first order SmA–SmC transitions have been observed, as mentioned in reference [2], but this results from another type of coupling. One example of such coupling is given in [14].

In conclusion, we showed that the different behaviors (Landau, Gaussian, 3DXY) which were observed in the specific heat are easily explained by means of the free energy of Andereck and Swift. The specific heat behavior is simply related to the strength of the coefficients which

couple the SmC order parameter with the layer-compression and with the density. We emphasize that there is no reason why the SmA–SmC transition should be close to a tricritical point, generically. It is only because the presence of the particular coupling mentioned above that such a tricritical transition could appear. Therefore, the several examples of tricritical behavior which were reported do not represent an intrinsic property of the SmA–SmC transition.

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