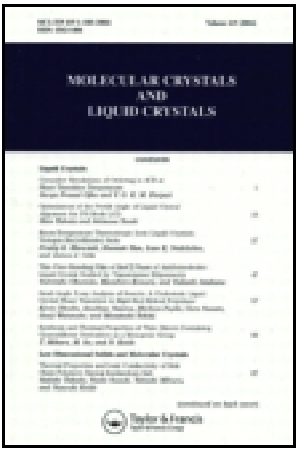

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The Nature of the Smectic A-Smectic C Transition

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THE NATURE OF THE SMECTIC A - SMECTIC C TRANSITION

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__Abstract__ We apply the Ginzburg criterion to the SmA-SmC transition and conclude that if sometimes the transition is mean-field like, this is very likely due to a coupling between the order parameter and the strains, and not to a large bare correlation length.

INTRODUCTION

From the nature of the order parameter characteristic of the smectic C ordering, de Gennes[1] concluded that the SmA-SmC transition belongs to the 3D XY universality class. However numerous experimental results seemed to be in contradiction with this prediction since they were analyzed with the help of the Landau theory which is equivalent to the mean field approximation. In particular, it was claimed that the heat capacity of several compounds[2], measured in the vicinity of the SmA-SmC transition temperature, is well described by the Landau free energy expansion including a sixth order term. Safinya et al.[3] tried to understand this situation using the Ginzburg criterion. This criterion permits to estimate a critical interval of temperature ΔT_C in which the Landau theory breaks. If this critical region ΔT_C is very small, it is not possible to observe the 3D XY properties of this transition. But already in the first work on the SmA-SmC transition, there were the discordant results of Delaye[4] and Galerne[5] who claimed that their results are not consistent with the mean field approximation.

Recently a renewal of interest arose from two different kinds of measurement. First the acoustic measurements of the Strasbourg group[6] showed the presence of anomalies in the elastic constants above the transition temperature T_C. These observations are not compatible with the mean field theory which predicts a total absence of anomaly above T_C.

Secondly, new specific heat measurements were found to be incompatible with the mean field theory and were analyzed with the help of different critical models: 3D XY model[7], gaussian model[8] and not conventional tricritical point[9]. May be it should be good to renew the analysis of some specific heat measurements in which the anomaly above T_C was considered as non intrinsic.

The purpose of this article is to make a careful application of the Ginzburg criterion in order to find the basic reason for this variety of situations. Here we limit ourselves only to the question to know why in some cases the specific heat behaves as in the mean field theory (figs. 1 and 2) and in another cases not (fig. 3).

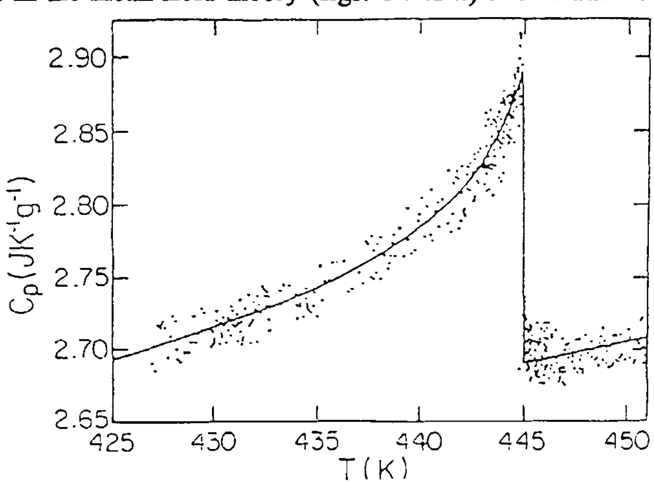

FIGURE 1: Specific-heat capacity of TBBA near the SmA-SmC transition. The line represents the Landau fit to these data. From Ref. 16.

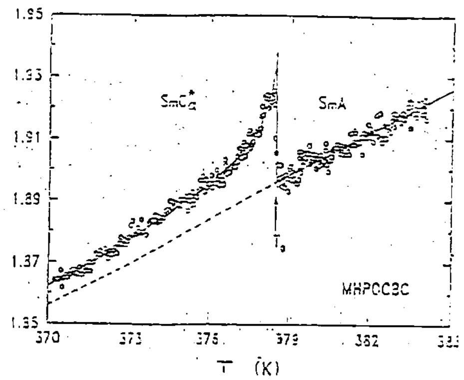

FIGURE2: Specific heat near the SmA-SmC* transition of MHPOCBC. From ref. 12.

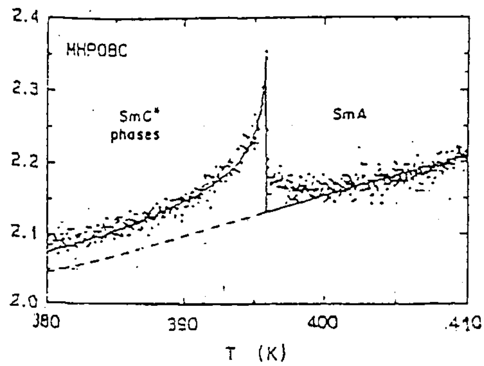

FIGURE3: Specific heat near the SmA-SmC* transition of MHPOBC From ref. 12.

THE GINSBURG CRITERION

We shall not give a derivation of the Ginzburg criterion (we refer the reader to the original article of Ginzburg[10]) and recently it was presented by us[11].

The Ginzburg criterion may be formulated in two different and equivalent forms. The first given by Ginzburg included only measurable quantities: the step in the specific heat at T_C as it is given by the mean field theory (Δ C) and the bare correlation length ξ_0. The correlation length itself is given by

$$\xi = \xi_0 \frac{\sqrt{\Delta T}}{T_C} \qquad (1)$$

$$(\Delta T = |T - T_C|)$$

In the second form the critical region ΔT_C is determined by means of the parameters of the Landau free energy expansion. Since ΔC and ξ_0 can be expressed in terms of the Landau coefficients, these two formulations are equivalent. However, as we shall see, the consideration of the two forms may be very useful.

In the first form ΔT_C is given by

$$\Delta T_C = \frac{k^2 T_C}{(16\pi)^2} \frac{1}{(\Delta C)^2 \xi_0^6} \tag{2}$$

The important point is that the bare correlation length appears in (2) at the sixth power in the denominator. Ginzburg uses this point to explain why some transitions (like ferroelectric or superconductor transitions) appear like mean field transitions when others (like ferromagnetic transitions) are observed as critical transitions. Transitions for which ξ_0 is large enough (i.e. long range interactions) appear like mean field transitions and those for which ξ_0 is small (i.e. short range interactions) are always critical.

The free energy expansion is written as

$$G = 1/2\ A\ |\theta|^2 + 1/4\ B^*\ |\theta|^4 + 1/6\ C\ |\theta|^6 + 1/(2\ m_\perp)\ |\nabla_\perp\theta|^2 + 1/(2\ m_\parallel)\ |\nabla_z\theta|^2 \tag{3}$$

In (3) θ is the order parameter and $A = A_0(T-T_C)$ and the critical region is

$$\Delta T_C = \frac{k^2 T_C^2 \left(m_\perp \sqrt{m_\parallel}\right)^2 B^{*2}}{16\pi^2 (4A_0)} \tag{4}$$

<u>THE BARE CORRELATION LENGTH</u>

Delaye[4] and Galerne[5] gave estimates of ξ_0. Delaye found $\xi_0 = 6.8$ Å and Galerne 2Å. Safinya et al.[3] proposed that for the compound $8\bar{S}5$, the bare correlation length is well larger then 13 Å. It was their explanation of the smallness of ΔT_C.

In ref. 11, we found that for TTBA, ΔT_C is 0.1^0C and using (2), we get that ξ_0 is about 4Å.

Ema et al.[12] measured the specific heat of several compounds and it is also possible to get values of ξ_0 from (2). For MHPOBC the critical region is about 1^0C and one finds ξ_0 = 3Å. However, for the compound MHPOCBC which exhibits a mean field behavior, one has ξ_0 = 7.5Å if one supposes a critical region of 0.1^0C.

Comparing the various estimates of ξ_0 (which can be seen as only order of magnitude), one finds that for all the compounds studied here, the bare correlation length is around the shortest distance between the long axis of two neighboring parallel molecules. Interesting also is the fact that the two compounds with the largest ξ_0 (6.8 Å and 7.5Å) present different behaviors: one is critical and the other is mean field. We conclude that there is no clear cut correlation between critical or non critical behavior and the value of the bare correlation length. We mention that in all the cases ξ_0 is the mean value of the two bare correlation lengths ($\xi_0 = (\xi_{0\perp}{}^2\, \xi_{0\parallel})^{1/3}$).

THE MODEL OF ANDERECK AND SWIFT

Andereck and Swift[13] proposed a model permitting the calculation of the elastic constants and their anomalies in the vicinity of the SmA-SmC transition temperature. It is based on coupling of the order parameter with the strains: the density ρ and the strain ($\partial u/\partial z$) when z is the direction perpendicular to the smectic layers. The starting point is the following free energy

$$F = 1/2\, A\, |\theta|^2 + 1/4\, B\, |\theta|^4 + 1/6\, C\, |\theta|^6 + 1/(2\, m_\perp)\, |\nabla_\perp \theta|^2 + 1/(2\, m_\parallel)\, |\nabla_z \theta|^2$$
$$+ 1/2\, \gamma_\rho\, \rho|\theta|^2 + 1/2\, \gamma_u \left(\frac{\partial u}{\partial z}\right) |\theta|^2 + 1/2\, A\rho^2 + 1/2\, B\left(\frac{\partial u}{\partial z}\right) + C\rho\left(\frac{\partial u}{\partial z}\right) \qquad (5)$$

This free energy is not identical with (3) since it is dependent of the strains (ρ and $\partial u/\partial z$) when the free energy given by (3) is dependent of the stress. The relation between the fourth order coefficients B and B^* is such that we have always $B^* < B$ (see ref. 11). However, contrarily to the other phase transitions with quadratic coupling between the order parameter and the strains (see ref. 14), B^* cannot be negative[15]. This means that the transition may be very near a tricritical point but cannot become of first order. The difference $(B - B^*)$ depends on the coupling coefficients γ_ρ and γ_u.

In the case of a strong enough coupling, B^* may be small enough to make the critical region also very small (see (4)). This argument provides also a convenient explanation of the frequent observation that the SmA-SmC transition is near a tricritical point.

Using the results of the specific heat measurements and those of the elastic constants, we have effectively verified that the model of Andereck and Swift gives the reason for the specific heat behavior of TTBA[11] in accordance with the mean field theory. In particular, we showed that B^* is small and equal to B/10. In spite of a relatively small value of the bare correlation length, ΔT_C is small because the presence of a quadratic coupling. Our suggestion is that this model is good also for other compounds near a tricritical point.

CONCLUSION

We applied the Ginzburg criterion to the specific heat at the SmA-SmC transition which belongs to the 3D XY universality class. If it appears that the specific heat follows a mean field behavior, this is likely due to the coupling of the order parameter with the strains (ρ and $\partial u/\partial z$). The proposal explanation of a large bare correlation length does not seem to be the good one. From our estimates for different compounds, we concluded that ξ_0 is about the shortest distance between two neighboring molecules and consequently is not large enough to reduce the critical region significantly. Finally, we stress once again that study of the specific heat alone is not enough to characterize the nature of the transition and that the elastic constant measurements remain an essential tool for the study of this transition[6].

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