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Lifshitz point in the smectic A phases

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Résumé. — Le point triple, où trois phases smectiques ($Sm A_1$, $Sm A_2$, $Sm \tilde{A}$) coexistent, est un point de Lifshitz associé à une onde de polarisation parallèle aux couches smectiques. On montre que d'intéressantes propriétés diélectriques pourraient être observées.

Abstract. — The triple point where three smectic phases are present (the $Sm A_1$, the $Sm A_2$ and the $Sm \tilde{A}$ phases) is a Lifshitz point associated with a polarization wave parallel to the layers. We show that interesting dielectric properties may be observed in the vicinity of this Lifshitz point.

1. **Introduction.** — Until recently it was thought that the structures of the nematic and the smectic A phases of liquid crystals were well understood. However, the discovery of the reentrant nematic [1] and smectic [2] phases has shown the complexity of these phases. Furthermore, the number of smectic A phases with different structures is relatively large [3], when one considers that all these smectic A phases are described by layers of molecules with their director perpendicular to the layers.

In this paper, we are concerned with three smectic phases : the A_1 , A_2 and \tilde{A} phases [4]. The structure of the $Sm A_1$ phase is basically the simple picture of $Sm A$ with the spacing between layers approximately equal to the molecular length (in its most extended configuration). The $Sm A_2$ phase has a bi-layered structure [5]. There are two periodicities : the first with wavelength equal to the molecular length (as in the $Sm A_1$ phase) and the other with wavelength double that of the molecular length. A possible model is given in reference [5], and it implies that the first periodicity with $k = 2\pi/l$ (l : molecular length) is a density wave whereas the periodicity with $k = \pi/l$ is a polarization wave [13], since the molecules are polar. An experimental verification of this model is given by the anomaly in the dielectric constant observed at the nematic- $Sm A_2$ transition [6]. The $Sm A_2$ phase can be seen as an antiferroelectric phase. The third smectic phase [4] (\tilde{A}) is also a bi-layered structure as is the

$Sm A_2$ phase but now there is a modulation parallel to the layer with wavelength $2\pi/q$. In the $Sm \tilde{A}$ phase the polarization wave has components perpendicular and parallel to the layers.

A typical phase diagram in which these three phases appear is given in reference [4] and we reproduce it in figure 1. One notices the existence of a triple point with

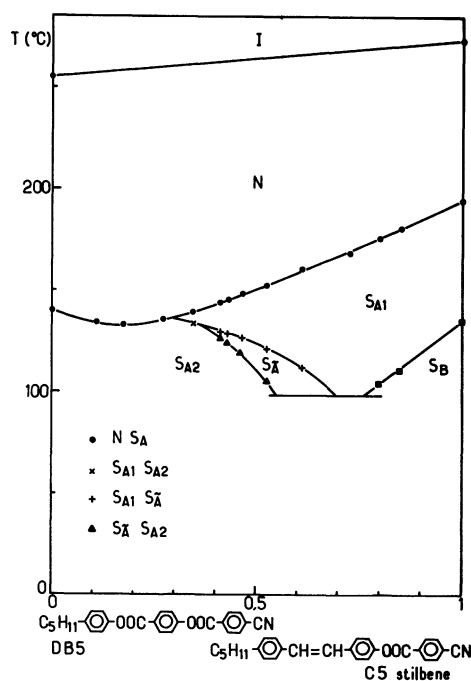


Fig. 1. — Experimental phase diagram showing the existence of the three smectic phases : A_1 , A_2 and \tilde{A} (from ref. [4]).

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coexistence of the three smectic phases. The purpose of this paper is to study the properties of the system around this particular point. In particular, we shall show that it could be a Lifshitz point [7]. We shall investigate first the thermodynamic properties and then the dielectric properties.

2. Phase transitions. — We shall use the formalism of Landau theory and the polarization P will be the order parameter. In the Sm A_1 phase, P is identically null and therefore we refer to this phase as the disordered phase. We write down the following free energy per unit volume :

$$G = \iint \left[\frac{a}{4} P^2(x, z) + \frac{b}{36} P^4(x, z) + \frac{K_1}{4} \left(\frac{\partial P}{\partial z} \right)^2 + \frac{\lambda_1}{4} \left(\frac{\partial^2 P}{\partial z^2} \right)^2 + \frac{K_2}{4} \left(\frac{\partial P}{\partial x} \right)^2 + \frac{\lambda_2}{4} \left(\frac{\partial^2 P}{\partial x^2} \right)^2 \right] dx dz. \quad (1)$$

The polarization P is dependent on z (perpendicular to the layer) and x (parallel to the layer). The free energy (1) is very similar to that proposed by Chen and Lubensky [9] for the study of NAC point. We could have added cross terms like $(\partial P/\partial x)^2 (\partial P/\partial z)^2$, but since the wave vector k which describes the z modulation is practically independent of T , we did not introduce them. As the procedure for minimizing (1) will show, we get a polarization modulated along the z direction if $K_1 < 0$ and $\lambda_1 > 0$. However, in the x direction we may either have (Sm \tilde{A}) or not have (Sm A_2) a modulation of P , and for that K_2 will be < 0 or > 0 and $\lambda_2 > 0$. Thus our choice of K_2 depends on the concentration c (in the case of a mixture) and/or the temperature T . K_1 , λ_1 and λ_2 may be dependent on T and c , but keep their signs as indicated.

We shall expand $P(x, z)$ in a Fourier series and retain only the first harmonic. It was shown that the higher harmonics are much smaller [15].

$$P(x, z) = P_0(e^{iqx} + e^{-iqx})(e^{ikz} + e^{-ikz}). \quad (2)$$

In the Sm A_2 phase $q \rightarrow 0$, and we get :

$$P(z) = 2 P_0(e^{ikz} + e^{-ikz}). \quad (3)$$

Putting (2) or (3) in (1), we obtain the free energy as a function of P :

$$G_{A_2} = 2 P_0^2 [a + K_1 k^2 + \lambda_1 k^4] + \frac{8}{3} b P_0^4 \quad (4)$$

(if $K_2 > 0$).

$$G_{\tilde{A}} = P_0^2 [a + K_1 k^2 + \lambda_1 k^4 + K_2 q^2 + \lambda_2 q^4] + b P_0^4 \quad (5)$$

(if $K_2 < 0$).

First we minimize with respect to k and q and get

$$k_0^2 = -\frac{K_1}{2\lambda_1} \quad (6a)$$

$$q_0^2 = -\frac{K_2}{2\lambda_2} \quad (6b)$$

and

$$G_{A_2} = 2 P_0^2 \left(a - \frac{K_1^2}{4\lambda_1} \right) + \frac{8b}{3} P_0^4 \quad (7a)$$

$$G_{\tilde{A}} = P_0^2 \left(a - \frac{K_1^2}{4\lambda_1} - \frac{K_2^2}{4\lambda_2} \right) + b P_0^4. \quad (7b)$$

Since a is linear function of T , we can write

$$a - \frac{K_1^2}{4\lambda_1} = \alpha_0(T - T_1) \quad (8)$$

$$a - \frac{K_1^2}{4\lambda_1} - \frac{K_2^2}{4\lambda_2} = \alpha_0(T - T_2), \quad (9)$$

and finally

$$G_{A_2} = 2 \alpha_0(T - T_1) P_0^2 + \frac{8}{3} b P_0^4 \quad (10a)$$

$$G_{\tilde{A}} = \alpha_0(T - T_2) P_0^2 + b P_0^4. \quad (10b)$$

Minimizing with respect to P_0 , the free energies as functions of T are :

$$G_{A_2} = -\frac{3}{8} \frac{\alpha_0^2(T - T_1)^2}{b} \quad (11a)$$

$$G_{\tilde{A}} = -\frac{1}{4} \frac{\alpha_0^2(T - T_2)^2}{b}. \quad (11b)$$

If $K_2 > 0$, we have only the Sm A_2 phase or the Sm A_1 phase, and a second-order transition (b is supposed to be positive) takes place at $T = T_1$. Now if $K_2 < 0$, we can have a Sm \tilde{A} phase. Since $(T_2 - T_1) = K_2^2/4 \alpha \lambda_2 > 0$ we have first the Sm A_1 -Sm \tilde{A} transition at T_2 , and at lower temperatures we have $G_{A_2} < G_{\tilde{A}}$. The Sm \tilde{A} -Sm A_2 transition appears at :

$$T_t = \frac{\sqrt{3} T_1 - \sqrt{2} T_2}{\sqrt{3} - \sqrt{2}}. \quad (12)$$

Since this transition is defined by the equality of the free energies G_{A_2} and $G_{\tilde{A}}$, it will be always a first-order transition. The phase diagram is shown in figure 2. One notes that the curves $T_2(K_2)$ and $T_t(K_2)$ are tangential to the line T_1 .

The wave vector q_0 is non zero at the Sm A_1 -Sm \tilde{A} and Sm \tilde{A} -Sm A_2 transitions. As the triple point, defined by $K_2 = 0$, $T = T_1$ is approached, q_0 goes to zero. One can show that $\lambda_2 q_0^4 = (\sqrt{3}/2 - 1) \alpha_0(T_1 - T)$

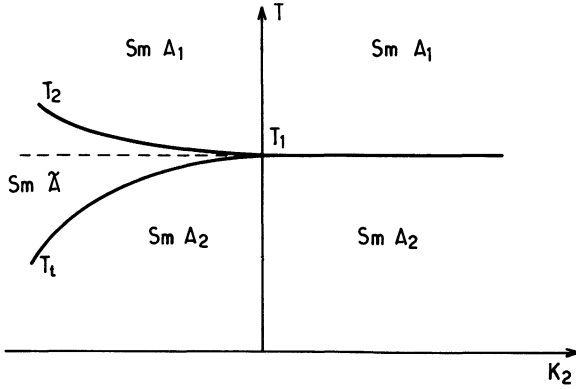


Fig. 2. — Theoretical phase diagram as functions of T and K_2 . Note the analogy with figure 1.

on the $Sm \tilde{A}$ - $Sm A_2$ line and $\lambda_2 q_0^4 = \alpha_0(T_2 - T_1)$ on the $Sm A_1$ - $Sm \tilde{A}$ line. Since experimentally T_1 is not always accessible, it is more interesting to write :

$$\alpha_0(T_2 - T_1) = \frac{\sqrt{3}}{\sqrt{3} - \sqrt{2}} \lambda_2 q_0^4. \quad (13)$$

This relation can be experimentally verified if T_2 and T_1 are taken for the same value of q_0 . One sees that for the q modulation, the triple point is a Lifshitz point. We expect very different fluctuations regimes in the k and q directions. This can be observed by measuring the intensity of the diffuse X-ray scattering [8]. In the q directions it will depend on q^4 and this effect will become stronger and stronger as the Lifshitz point is approached.

More specifically, we expect that around q_0 and k_0 , the X-ray intensity will be equal to $kT \left[\left(a - \frac{K_1^2}{4\lambda_1} \right) + \lambda_1(k^2 - k_0^2)^2 + K_2 q^2 + \lambda_2 q^4 \right]^{-1}$ if $K_2 > 0$ i.e. the intensity peaks at $k = \pm k_0$. But if $K_2 < 0$ the X-ray intensity is then

$$kT \left[\left(a - \frac{K_1^2}{4\lambda_1} - \frac{K_2^2}{4\lambda_2} \right) + \lambda_1(k^2 - k_0^2)^2 + \lambda_2(q^2 - q_0^2)^2 \right]^{-1} \quad (14)$$

and the maximum is along two rings defined by $(\pm k_0, q_0 \cos \varphi, q_0 \sin \varphi)$. This is analogous to the situation at the NAC [9] point. And this analogy indicates that our meanfield theory is probably not very good along the transition line $Sm A_1$ - $Sm \tilde{A}$. In the case of the NAC point the meanfield theory predicts a second-order transition for the N- $Sm C$ line, but a more complete theory [10] with fluctuations gives a first-order transition. It is very likely that the same thing applies for the $Sm A_1$ - $Sm \tilde{A}$ transition.

3. Dielectric permittivity. — First we shall calculate the influence of a constant electric field E directed along the z direction. This will give the parallel susceptibility. One might think that the only effect of the field would be to give a homogeneous component p_0 to the polarization. In fact, besides p_0 a second component modulated with wave vector equal to $2k$ will appear. To see this, we suppose that there is a small perturbation $2p(z)$ to P :

$$P(x, z) = P_0[e^{iqx} + e^{-iqx}] [e^{ikz} + e^{-ikz}] + 2p(z). \quad (15)$$

Expanding $p(z) = \sum_{\alpha} p_{\alpha} e^{iaz}$ (including $\alpha = 0$) we can calculate the new free energy. Supposing that $p(z)$ is much smaller than P , we get $G = G(P = 0) + \Delta G$, with :

$$\Delta G_{A_2} = a \sum |p_{\alpha}|^2 + \frac{8}{3} b P_0^2 \left(2 \sum |p_{\alpha}|^2 + \sum_{\alpha+\alpha'+2k=0} p_{\alpha} p_{\alpha'} + \sum_{\alpha+\alpha'-2k=0} p_{\alpha} p_{\alpha'} \right) + K_1 \sum \alpha^2 |p_{\alpha}|^2 + \lambda_1 \sum \alpha^4 |p_{\alpha}|^2 \quad (16)$$

and

$$\Delta G_{\tilde{A}} = a \sum |p_{\alpha}|^2 + \frac{4}{3} b P_0^2 \left(2 \sum |p_{\alpha}|^2 + \sum_{\alpha+\alpha'+2k=0} p_{\alpha} p_{\alpha'} + \sum_{\alpha+\alpha'-2k=0} p_{\alpha'} p_{\alpha} \right) + K_1 \sum \alpha^2 |p_{\alpha}|^2 + \lambda_1 \sum \alpha^4 |p_{\alpha}|^2. \quad (17)$$

In (16) and (17) we have retained only the terms containing p_0 . We see that, besides the term p_0^2 , there is a coupling of p_0 to $p_{\pm 2k}$. We shall now write down the free energy difference ΔG , in the presence of the field E :

$$\Delta G_{\tilde{A}} = p_0^2 \left(a + \frac{8}{3} b P_0^2 \right) + 2 |p_{2k}|^2 \left(a + \frac{8}{3} b P_0^2 + 2 k^2 K_1 + 8 k^4 \lambda_1 \right) + \frac{8}{3} b P_0^2 p_0 (p_{2k} + p_{-2k}) - 2 E p_0, \quad (18)$$

$$\Delta G_{A_2} = p_0^2 \left(a + \frac{16}{3} b P_0^2 \right) + 2 |p_{2k}|^2 \left(a + \frac{16}{3} b P_0^2 + 4 k^2 K_1 + 16 k^4 \lambda_1 \right) + \frac{16}{3} b P_0^2 p_0 (p_{2k} + p_{-2k}) - 2 E p_0. \quad (19)$$

We calculate p_0 from the conditions $\partial(\Delta G)/\partial p_0 = 0$ and $\partial(\Delta G)/\partial p_2 = 0$ (with $p_2 = p_{2k} = p_{-2k}$) and the inverse of parallel susceptibility $\chi_{\parallel}^{-1} = E/2 p_0$. After some tedious algebra, using the values of $k^2 = (K/4 \lambda_1)$ ($K = -K_1$) and the values of P_0 calculated through $\partial G/\partial P_0 = 0$, we get :

$$\chi_{\parallel}^{-1}(A_2) = \frac{\alpha_0}{2}(T_1 - T) + \frac{Kk^2}{4} - \frac{\alpha_0^2(T_1 - T)^2}{\alpha(T_1 - T) + \frac{9}{2}Kk^2}, \quad (20)$$

$$\chi_{\parallel}^{-1}(\tilde{A}) = \frac{\alpha_0}{6}(T_2 - T) + \frac{Kk^2}{4} - \frac{K_1 q^2}{4} - \frac{\frac{8}{9} \alpha_0^2(T_2 - T)^2}{\frac{2}{3} \alpha_0(T_2 - T) + 5Kk^2 - 9K_2 q^2}. \quad (21)$$

In the Sm A_1 phase, we have

$$\chi_{\parallel}^{-1}(A_1) = \frac{a}{2} = \frac{\alpha_0}{2}(T - T_1) + \frac{Kk^2}{4}. \quad (22)$$

It is interesting to consider the variation of χ_{\parallel}^{-1} in the vicinity of the different transitions. At T_1 , the Sm A_1 -Sm A_2 transition temperature, χ_{\parallel}^{-1} is continuous as may be seen from (20) and (22) : $\chi_{\parallel}^{-1}(T_1) = Kk^2/4$. There is only a change in the slope. However, if for small values of $(T_1 - T)$ we have $\alpha_0 | T_1 - T | \ll Kk^2/4$, this anomaly will be difficult to observe.

At T_2 , the Sm A_1 -Sm \tilde{A} transition temperature, χ_{\parallel}^{-1} is also continuous. Taking into account that $\alpha_0(T_2 - T_1) = K^2/4 \lambda_2$, we verify immediately from (21) and (22) that $\chi_{A_1}^{-1}(T_2) = \chi_{\tilde{A}}^{-1}(T_2)$.

However at T_3 , the temperature of the Sm \tilde{A} -Sm A_2 transition, we expect a discontinuity in χ_{\parallel}^{-1} . We can evaluate this discontinuity if we suppose that, in the temperature range of interest $\chi_{\parallel}^{-1}(A_2)$ and $\chi_{\parallel}^{-1}(\tilde{A})$ can be approximated by (see (20) and (21)) :

$$\chi_{\parallel}^{-1}(A_2) = \frac{\alpha_0}{2}(T_1 - T) + \frac{Kk^2}{4}$$

$$\chi_{\parallel}^{-1}(\tilde{A}) = \frac{\alpha_0}{6}(T_2 - T) + \frac{Kk^2}{4} - \frac{K_2 q^2}{4}.$$

We get at T_3 :

$$\chi_{\parallel}^{-1}(\tilde{A}) - \chi_{\parallel}^{-1}(A_2) \simeq -0.41 \lambda_2 q^4. \quad (23)$$

We note that this quantity is negative, the Sm \tilde{A} phase is more polarizable than the Sm A_2 phase. It is also interesting to note that the discontinuity depends on q and is larger far from the Lifshitz point.

Also interesting is the behaviour of the perpendicular susceptibility, which is measured with the electric field perpendicular to the director. We introduce the following new terms in the free energy :

$$\frac{A}{2} p_{\perp}^2 + \frac{B}{8} p_{\perp}^2 P^2(x, z) - E p_{\perp}. \quad (24)$$

After integration, we have :

$$\Delta G(A_2) = \frac{p_{\perp}^2}{2} (A + 2BP_0^2) - E p_{\perp}$$

$$\Delta G(\tilde{A}) = \frac{p_{\perp}^2}{2} (A + BP_0^2) - E p_{\perp}.$$

Thus, the perpendicular susceptibilities are :

$$\chi_{\perp}^{-1}(A_2) = A + 2BP_0^2 \quad \text{and} \quad \chi_{\perp}^{-1}(\tilde{A}) = A + BP_0^2.$$

As in the case of the parallel susceptibility, χ_{\perp} is continuous along the Sm A_1 -Sm A_2 and Sm A_1 -Sm \tilde{A} lines but it will exhibit a discontinuity at the Sm \tilde{A} -Sm A_2 transition.

4. Dielectric relaxations. — We shall consider only the parallel configuration, since we expect relaxation modes associated with the ordering. As seen above, the electric field E causes the appearance of the induced polarization p_0 and also of the $p_{\pm 2k}$ components. Thus with each of the components p_0 and $p_{\pm 2k}$ will be associated a relaxation mode.

We shall write the excess free energy ΔG (indicated above by the expressions (18) and (19)) in the following form taking into account that $p_{2k} = p_{-2k} = p_2$:

$$\Delta G = \alpha p_0^2 + 2 \beta p_2^2 + 2 \gamma p_0 p_2 \quad (25)$$

α , β and γ have to be identified with the coefficients appearing in (18) and (19).

We evaluate the relaxation times by means of the Landau-Khalatnikov equations [11] :

$$\frac{\partial p_0}{\partial t} = -\frac{\Gamma_1}{2} \frac{\partial \Delta G}{\partial p_0}, \quad (26a)$$

$$\frac{\partial p_2}{\partial t} = -\frac{\Gamma_2}{2} \frac{\partial \Delta G}{\partial p_2}. \quad (26b)$$

These are simplified equations since in general the Γ matrix is not diagonal. Here we use the same approximation as in reference [12], in the calculation of the smectic C* relaxation times. We get :

$$\frac{\partial p_0}{\partial t} = -\Gamma_1 \alpha p_0 - \Gamma_1 \gamma p_2, \quad (27a)$$

$$\frac{\partial p_2}{\partial t} = -\Gamma_2 \gamma p_0 - 2\Gamma_2 \beta p_2. \quad (27b)$$

The inverses of the relaxation times are the eigenvalues of the coefficient matrix in (27). We have :

$$s_{\pm} = \frac{\Gamma_1 \alpha + 2\Gamma_2 \beta}{2} \pm \frac{1}{2} \sqrt{(\Gamma_1 \alpha - 2\Gamma_2 \beta)^2 + 4\Gamma_1 \Gamma_2 \gamma^2}. \quad (28)$$

In the smectic A_1 phase, we have only one relaxation time since p_2 is not coupled to p_0 (the coupling of p_0 and p_2 takes place through P . See (12) and (14)) :

$$s = \Gamma_1 a = \Gamma_1 \left[\alpha_0(T - T_1) + \frac{K}{2} k^2 \right] \\ = \left[\alpha_0(T - T_2) + \frac{K}{2} k^2 - \frac{k_2 q^2}{2} \right] \Gamma_1. \quad (29)$$

At the $\text{Sm } A_1\text{-Sm } A_2$ transition we have $s_+ = \Gamma_1 K k^2/2$ and $s_- = 9 \Gamma_2 K k^2$. Thus, from (29) we easily see that the mode s_+ goes continuously into the mode s of the disordered phase. We have an analogous result at the $\text{Sm } A_1\text{-Sm } \tilde{A}$ transition : $s_+ = \Gamma_1 (K k^2 - K_2 q^2)/2$ and $s_- = \Gamma_2 (5 K k^2 - K_2 q^2)$. Here also we expect continuity between the modes s_+ and s . In both cases, there is a break in the slope. Whether s_- will be larger or smaller than s_+ depends on the ratio Γ_1/Γ_2 . At the $\text{Sm } \tilde{A}\text{-Sm } A_2$ transition, we predict a discontinuity between the two modes s_+ and s_- when crossing the transition.

We can have a more detailed picture if we assume that $\Gamma_1 = \Gamma_2 = \Gamma$ and that we are in the vicinity of the transitions, i.e. the difference $(T - T_1)$ and $(T - T_2)$ are small quantities. We get the following expressions for the inverses of the relaxation times : In the $\text{Sm } A_2$ phase

$$s_+ = \Gamma \left[\alpha_0(T_1 - T) + \frac{K k^2}{2} \right] \\ s_- = \Gamma [2 \alpha_0(T_1 - T) + 9 K k^2/2] \quad (30)$$

and in the $\text{Sm } \tilde{A}$ phase

$$s_+ = \left[\frac{\alpha_0}{3} (T_2 - T) + \frac{K k^2}{2} - \frac{K_2 q^2}{2} \right] \Gamma \\ s_- = \left[\frac{2 \alpha_0}{3} (T_2 - T) + 5 K k^2 - K_2 q^2 \right] \Gamma. \quad (31)$$

In particular, we note that at the $\text{Sm } \tilde{A}\text{-Sm } A_2$ transition, we have $s_+(\tilde{A}) < s_+(A_2)$ but $s_-(\tilde{A}) > s_-(A_2)$. These discontinuities are functions of q . It is possible to show that we have :

$$s_+(\tilde{A}) - s_+(A_2) \sim (1.63) K_2 q^2 \Gamma \quad (K_2 < 0)$$

and

$$s_-(\tilde{A}) - s_-(A_2) = \left(-\frac{4}{3} K_2 q + \frac{K k^2}{2} \right) \Gamma.$$

It is of practical importance to calculate the mode amplitudes of the dielectric constant. Writing

$$\chi = \frac{A_+}{1 + j\omega/s_+} + \frac{A_-}{1 + j\omega/s_-} \quad (32)$$

we can calculate the amplitudes, A_+ and A_- . We introduce the quantities $p_0 = \chi(\omega) E$ and $p_2 = K(\omega) E$ in (27), and then we deduce $\chi(\omega)$. We look for an expression for $\chi(\omega)$ which is identical to (32). The algebra is tedious but without difficulty. We find

$$A_+ = \frac{1}{s_+} \frac{\Gamma_1 \Gamma_2 \beta - \Gamma_1 s_+}{s_- - s_+} \quad (33a)$$

$$A_- = \frac{1}{s_-} \frac{\Gamma_1 \Gamma_2 \beta - \Gamma_1 s_-}{s_+ - s_-} \quad (33b)$$

with s_{\pm} given by (28).

It is interesting to know what are the relative values of A_+ and A_- , near the $\text{Sm } A_1\text{-Sm } \tilde{A}$ and the $\text{Sm } A_1\text{-Sm } A_2$ transition temperature. In this case γ which is proportional to $(T_1 - T)$ and $(T_2 - T)$ will be small. We find

$$A_+ = \frac{1}{\alpha} \quad (34a)$$

$$A_- \simeq \frac{\Gamma_1^2 \gamma^2}{2 \beta (\Gamma_1 \alpha - 2 \beta \Gamma_2)^2} \quad (34b)$$

This means that A_- will be very small in the vicinity of T_1 or T_2 , so it would be difficult to observe the mode s_- . Thus to detect it experimentally, it will be better to do measurements far from the transition temperatures.

5. Conclusion. — Our analysis of the triple point where the three smectic phases, $\text{Sm } A_1$, $\text{Sm } A_2$ and $\text{Sm } \tilde{A}$ coexist showed that it is a Lifshitz point very analogous to the NAC point where nematic, $\text{Sm } A$ and $\text{Sm } C$ phases coexist. Because the ordering is that of dipoles (and not of the density as in the NAC point) we expect interesting dielectric properties, in particular two modes in the dielectric relaxations. The preliminary results of Druon [14] show the existence of these two modes.

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