CRITICAL BEHAVIOUR OF DYNAMIC TWIST VISCOSITY $\gamma 1$ NEAR POLYCRITICAL POINTS

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Résumé. — Nous présentons ici les premières mesures réalisées près de points polycritiques révélés dans des mélanges binaires, de la viscosité dynamique de torsion γ_1.

Le long de la ligne de transition NS_A du deuxième ordre, tracée dans un diagramme température-concentration, nous observons un même comportement critique divergent. Par contre, près du point triple « N-A-C » du deuxième ordre le régime divergent disparaît.

Abstract. — We report here the first measurements performed close to polycritical points revealed in binary mixtures of the dynamic twist viscosity γ_1.

Along the second order NS_A line, drawn in a binary temperature-concentration diagram, critical behaviour is found to be the same. On the other hand, no divergence is observed near the « N-A-C » singular point.

1. **Introduction.** — The dynamical critical behaviour near a nematic-smectic A transition is still not quite understood. In particular, results concerning the dynamic twist viscosity are not all coherent.

Theoretically, two critical behaviours are expected :

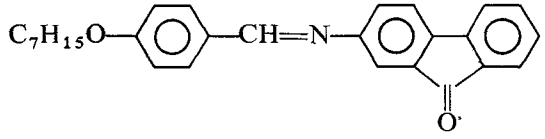
— either a Landau type one in a mean field theory [1],

— or an helium like one in a renormalized theory [2].

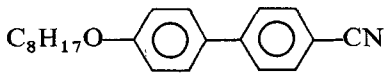
Numerous experimental results about the γ_1 divergence (as well as other viscoelastic coefficients divergences) have been already published, but the values of the corresponding critical exponents ν offer a great diversity [3]. Nevertheless, we must notice particularly all these measurements have been performed on pure compounds for which the N-S_A transition always remains weakly first order. But if we consider the truly second order transitions previously observed in binary mixtures of mesomorphic compounds [4] this disadvantage is of course removed ; we will present here the γ_1 measurements performed on such mixtures.

This approach is the more interesting as multricritical points have been also already revealed in these binary systems [5, 6].

2. **Experimental results.** — We have studied binary mixtures of p-n-heptyloxybenzylidene aminofluorenon (« 7 one »)



with octyloxycyanobiphenyl (8 OCB)



For small quantities of 8 OCB ($0 < x_{8\ OCB} < 0.20$) the temperature-concentration diagram (Fig. 1) presents three equilibrium *lines* :

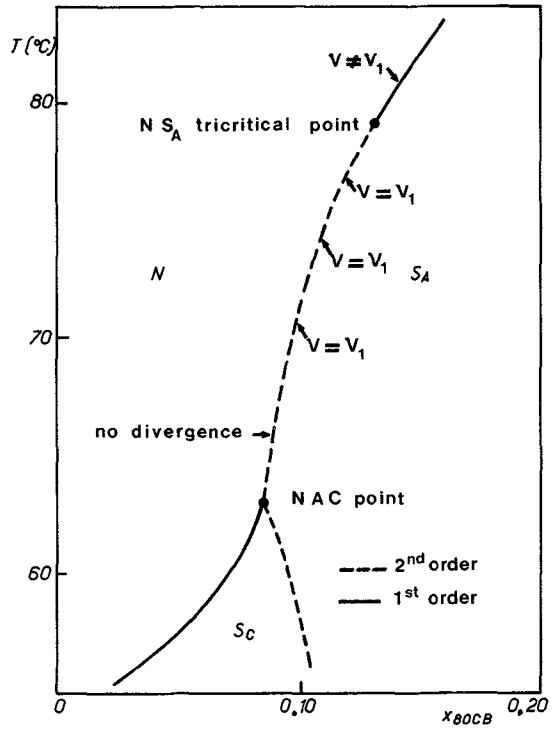


FIG. 1. — « 7 one-8 OCB » : temperature-concentration diagram near polycritical points.

— a nematic-smectic C transition *line*
— a nematic-smectic A transition line
— a smectic A-smectic C transition line

and exhibits two peculiar points :

— a tricritical point limit between first order and second order N-S_A transitions ($x_{8\,OCB} = 0.135$)

— a « N-A-C » triple point [7, 8] ($x_{8\,OCB} \simeq 0.087$: intersection of the second order N-S_A, the second order S_A-S_C lines and the first order N-S_C pseudo line) to be compared to a so-called Lifshitz point.

These results obtained by magnetic, enthalpic and refractive index anisotropy measurements have been already described in a previous study [9].

Now, we carry on this analysis by the determination of the critical behaviour of the dynamic twist viscosity coefficient γ_1 close to these points. We have performed these γ_1 measurements by the well-known rotating magnetic field method with a thermal stability of 10 mK [3*b*].

The experimental curves γ_1 *versus* the reduced temperature $\frac{T - T_{NA}}{T_{NI} - T_{NA}}$ are plotted on the figure 2 for five mixtures. In all cases, the twist viscosity coefficient increases strongly near the transition and reaches remarkably high values (5 poises).

We can deduce from these experimental results the ν critical exponent values by using a fitting method previously described [10] :

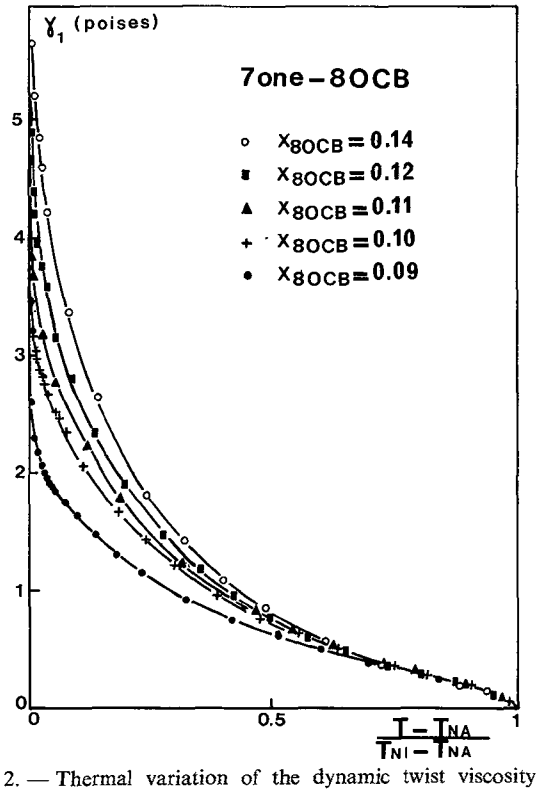


FIG. 2. — Thermal variation of the dynamic twist viscosity γ_1 *versus* the reduced temperature $\frac{T - T_{NA}}{T_{NI} - T_{NA}}$.

Erratum : In figure 2 read : ○ $x_{8\,OCB} = 0.09$, ▪ $x_{8\,OCB} = 0.10$, ▲ $x_{8\,OCB} = 0.11$, + $x_{8\,OCB} = 0.12$, ● $x_{8\,OCB} = 0.14$.

• For a weakly first order NS_A transition ($x_{8\,OCB} = 0.14$), the ν exponent differs from any theoretical prediction that is no wonder since we identify the experimental transition temperature with T_c^* and thereby the fitting may be contested.

• On the second order NS_A line (we recall that no discontinuity of the orientational order or of the transitional enthalpy has been detected within the accuracy of our experiment for mixtures in which $0.087 < x_{8\,OCB} < 0.135$) the analysis of the γ_1 divergence leads to a constant ν exponent. Nevertheless, we do not give the obtained value as the choice of the *normal* γ_1 contribution (no divergent part) used in the fitting may always be confuted. The invariability of the critical exponent remains in any case a very essential point.

• At last, a very surprising result is obtained for the mixture ($x_{8\,OCB} = 0.09$) the transition point of which is the farthest of the tricritical point on the second order NS_A line and close to the « N-A-C » triple point : as it is shown on the figure 3 no divergence is observed up to 10 mK from the NS_A transition.

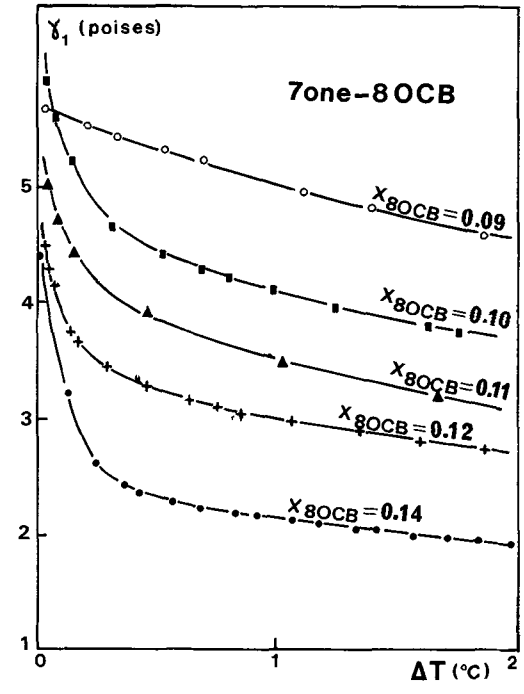


FIG. 3. — Thermal evolution of the dynamic twist viscosity γ_1 near the NS_A transitions.

Let us point out two remarks :

— after the experiment no important thermal modification of the mixtures has been detected : they are chemically stable ;

— the γ_1 non divergence could be explained by a first order NS_C transition (if some uncertainty in the mixture) but it is not so since no enthalpy discontinuity can be detected.

3. **Discussion.** — Thus, three types of behaviour seem to occur according to the different mixtures :

— one connected to the first order character where no correlation between experimental results and theory can be easily carried out (without adding an other parameter T_C^*),

— a second domain where the second order NS_A transition character leads to only one critical behaviour (at least in the part accessible to our experiment),

— and finally the particular influence of the « N-A-C » triple point seems to be felt and leads to a non divergent γ_1.

Unfortunately, the γ_1 behaviour near such a point has not been yet theoretically considered and only the elastic constants K_1 (splay), K_2 (twist) and K_3 (bent) have been taken in account [8] :

— near a second order N-S_A transition : K_2 and K_3 diverge as the coherence length ξ when K_1 does not diverge

— near a N-S_C transition : K_1, K_2 and K_3 diverge as ξ^2

— at the « N-A-C » point : K_3 diverges as ξ when K_1 and K_2 do not diverge.

So, if we can compare (as it seems consistent) the γ_1 behaviour to the K_2 one, the last result for which the dynamic twist viscosity does not diverge near a « N-A-C » point seems to be a very important result.

Nevertheless, it is clear that before arguing a cross-over directly connected to the « N-A-C » point, it is necessary to perform measurements on the NS_C line to further check the agreement with the theory.

Unfortunately, these experiments on the NS_C line are very difficult because of the metastability of these phases (when the composition of the mixture is slightly different of the pure « 7 one »).

We try now to find other systems to perform γ_1 measurements on both sides of the polycritical point.

The determination of properties already theoretically analysed (such as the twist elastic constant K_2 or directly the coherence length) could of course bring new informations on the critical behavior near the NS_A and NS_C transitions.

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