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F. Hardouin, M. Achard, G. Sigaud. CRITICAL BEHAVIOUR OF DYNAMIC TWIST VISCOSITY γ 1 NEAR POLYCRITICAL POINTS. Journal de Physique Colloques, 1979, 40 (C3), pp.C3-371-C3-373. 10.1051/jphyscol:1979372. jpa-00218768

HAL Id: jpa-00218768 https://hal.science/jpa-00218768

Submitted on 4 Feb 2008

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CRITICAL BEHAVIOUR OF DYNAMIC TWIST VISCOSITY γ_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 températureconcentration, 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 aminofluore-non (« 7 one »)





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

with octyloxycyanobiphenyl (8 OCB)

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

- 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 ($x_{8 \text{ OCB}} = 0.135$)

— a « N-A-C » triple point [7, 8] ($x_{8 \text{ OCB}} \simeq 0.087$: 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 wellknown rotating magnetic field method with a thermal stability of 10 mK [3b].

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 v 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 $\frac{T - T_{NA}}{T_{NI} - T_{NA}}$.



• For a weakly first order NS_A transition $(x_{8 \text{ OCB}} = 0.14)$, the v 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 $0.087 < x_{8 \text{ OCB}} < 0.135$) 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 ($x_{8 \text{ OCB}} = 0.09$) 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. — 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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