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Tricritical Exponents for the Isotropic-Nematic Transition: An Experimental Verification

P. H. Keyes
University of Massachusetts at Boston, Boston, Massachusetts 021215, and Francis Bitter National Magnet Laboratory, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

and

J. R. Shane
University of Massachusetts at Boston, Boston, Massachusetts 021125
(Received 11 December 1978)

The gap exponent for the isotropic-nematic phase transition in MBBA (N-[p-methoxybenzylidine]-p-butylaniline) has been determined from measurements of the induced birefringence in very strong magnetic fields. The experimental value, $\Delta = 1.26 \pm 0.10$, is consistent with the tricritical exponent, $\Delta = \frac{5}{4}$, but is clearly inconsistent with the exponent $\Delta = 2$ predicted by the mean-field theory. It is also shown that some of the observed effects are more than two orders of magnitude larger than the values predicted by the mean-field theory.

The purpose of this Letter is to present experimental results that clearly demonstrate that the critical exponents for the isotropic-nematic phase transition are those expected for a tricritical point. Measurements presented here of the birefringence induced by very strong magnetic fields yield, for the first time, the gap exponent Δ for this transition. It is found that the observed value of Δ is consistent with the value expected for a tricritical point.

It has recently been suggested[1] that tricritical exponents would be expected for this transition, rather than the exponents predicted by the de Gennes mean-field theory.[2] Until now, a clear experimental distinction between the two models has not been possible because of several accidental coincidences between the mean-field and tricritical exponents. However, the predicted values of Δ for the two models are considerably different and the agreement between the observed value and the tricritical exponent is unambiguous.

The experiment is in essence a determination of the equation of state for the order parameter[2] Q as a function of temperature T and magnetic field H on the high-temperature or "isotropic" side of the transition. For the purpose of discussion it shall be assumed that this relation can be represented in the scaling form[3]

$$h/Q^{\delta} + B/Q^{\omega} = g(Q/\epsilon^{\beta}) . \tag{1}$$

In the mean-field theory an equation of this form results from the Landau–de Gennes free-energy expansion

$$F = F_0 + \tfrac{1}{2} A Q^2 - \tfrac{1}{3} B Q^3 + \tfrac{1}{4} C Q^4 - hQ , \tag{2}$$

where $h = \chi_a H^2$, with χ_a the anisotropy of the magnetic susceptibility, and $A = A_0 \epsilon$, with $\epsilon = T - T^*$. Minimizing (2) with respect to Q results in (1) with $g(x) = C + A_0 x^{-2}$, and critical exponents $\delta = 3$, $\omega = 1$, and $\beta = \frac{1}{2}$.

For a tricritical point[4] the values would be $\delta = 5$, $\omega = 2$, and $\beta = \frac{1}{4}$. An explicit expression for $g(x)$ has not yet been given for this case, although, as shall be presently be demonstrated, its behavior as x approaches zero may be deduced in part.

This paper is primarily concerned with the susceptibility $\partial Q/\partial h$ and its derivative $\partial^2 Q/\partial h^2$. By definition the critical exponents for these quantities are, respectively, γ and $\gamma+\Delta$. Values for these indices may be obtained from the equation of state. Consider first the susceptibility; differentiation of (1) with respect to h gives

$$\partial Q/\partial h = [\delta Q^{\delta-1}g(x) + Q^{\delta}\epsilon^{-\beta}g'(x) - B(\delta-\omega)Q^{\delta-\omega-1}]^{-1}, \tag{3}$$

with the prime denoting the derivative taken with respect to the argument. In order that (3) have a sensible limit as $h, Q \to 0$, it must be the case that $g(x)$ has the asymptotic dependence $g \sim x^{1-\delta}$. Then the temperature dependence of the zero-field susceptibility will be $\partial Q/\partial h \sim \epsilon^{-\beta(\delta-1)}$, so that $\gamma = \beta(\delta-1)$. It should be noted that although the mean-field and tricritical models have different values for β and δ, they both give the same value of $\gamma = 1$.

The expression for $\partial^2 Q/\partial h^2$ is more interesting in that it distinguishes between the tricritical and mean-field cases. Differentiation of (3) with respect to h gives

$$\partial^2 Q/\partial h^2 = [\delta(1-\delta)Q^{\delta-2}g(x) - 2\delta Q^{\delta-1}g'(x) - Q^{\delta}\epsilon^{-2\beta}g''(x) + B(\delta-\omega)(\delta-\omega-1)Q^{\delta-\omega-2}](\partial Q/\partial h)^3. \tag{4}$$

The $x^{1-\delta}$ dependence previously taken for g causes complete cancellation of the first three terms in (4). Evidently a term varying as $x^{2-\delta}$ must survive in order to obtain a finite limit for (4) as $Q \to 0$. Therefore, the temperature dependence of $\partial^2 Q/\partial h^2$ in zero field is $\epsilon^{-2\delta-\beta}$ and thus $\Delta = \gamma + \beta$. Another possibility, however, is that the first three terms in (4) do actually vanish but $\delta - \omega - 2 = 0$ causing the fourth term to remain. Indeed, this situation is precisely what prevails for the de Gennes mean-field theory, as may be verified by direct calculation. In this case the zero-field temperature dependence is $\partial^2 Q/\partial h^2 \sim \epsilon^{-3\gamma}$ and hence $\Delta = 2\gamma$. In summary, if the de Gennes theory is correct then $\Delta = 2$, but if the tricritical hypothesis is valid then $\Delta = \frac{5}{4}$.

The experiment was performed on the material N-(p-methoxybenzylidine)-p-butylaniline (MBBA), which had been studied previously[5] up to about 1 T. The sample was placed in a water-jacketed cuvette having an optical path length of 1 cm. The temperature was measured at the sample and was controlled to within about 0.02°C by means of a circulator. To shield the sample from the heat of the magnet, the cuvette was placed in a glass Dewar. The Dewar was, in turn, inserted into the core of a Bitter coil having optical ports at right angles to the field direction. A helium-neon laser beam polarized at 45° to the field passed through the sample, then through a quarter-wave plate also oriented at 45° to H, and finally through an analyzer which could be rotated to produce an extinction. In this manner the birefringence, which is proportional to the order parameter Q, was measured.

The results of the experiment are displayed in Fig. 1; to avoid crowding only about half of the total data are plotted. The analysis is facilitated by the plot of $\Delta n/H^2$ vs H^2. Since

$$Q/h = (\partial Q/\partial h)_{h=0} + \tfrac{1}{2}(\partial^2 Q/\partial h^2)_{h=0} + \dots,$$

the intercept and slope in Fig. 1 are directly proportional to the susceptibility and its derivative, respectively.

The inverse of the intercepts is shown in Fig. 2. This result, similar to that obtained by others,[5] demonstrates that $\gamma = 1$ over a wide temperature range. Extrapolation of the linear fit gives the value of T^*. The small deviations from linearity near T_c have also been seen by others and are not at all understood.

The expression for Q/h may be rewritten as

$$Q/h = (\partial Q/\partial h)_{h=0}[1 + f + \dots],$$

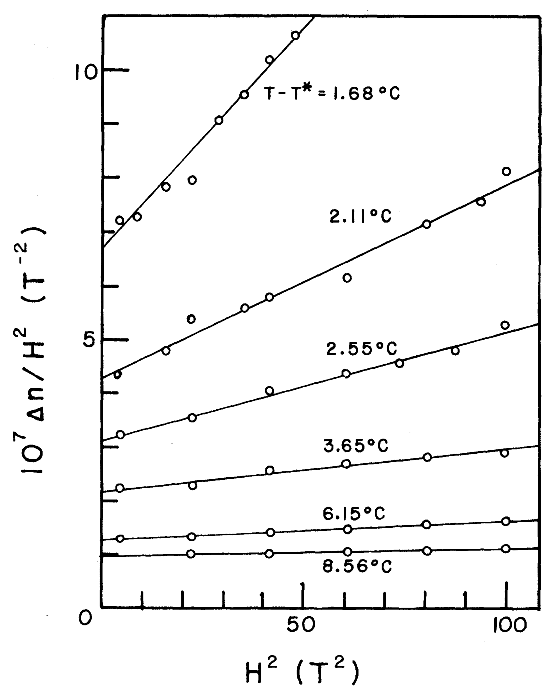

FIG. 1. The Cotton-Mouton coefficient, proportional to the order-parameter susceptibility at zero field, vs the square of the magnetic field for MBBA.

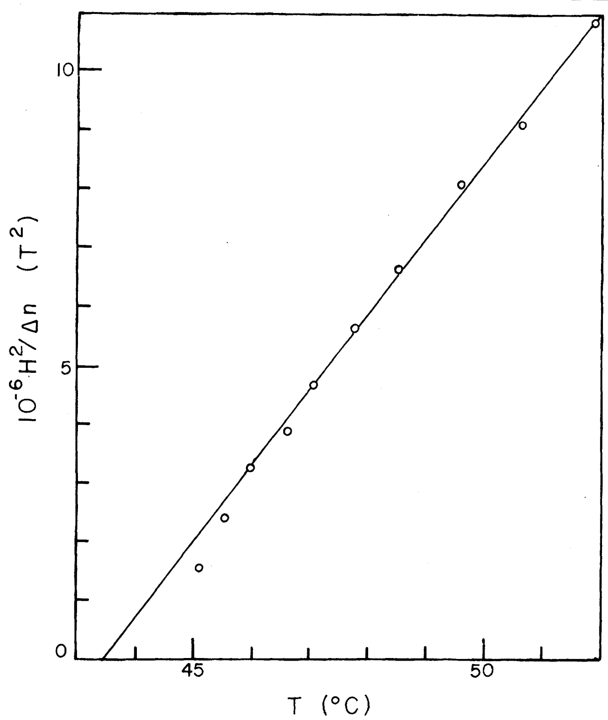

FIG. 2. Inverse of the Cotton-Mouton coefficient vs temperature.

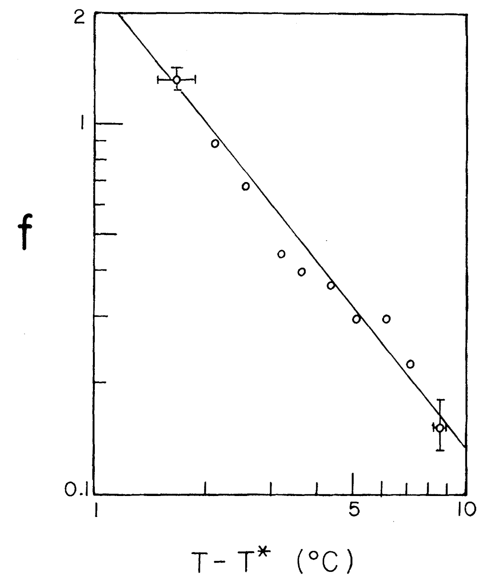

FIG. 3. The fractional increase f of the susceptibility at maximum field over its zero-field value vs $T - T^*$.

where

$$f = \tfrac{1}{2} h (\partial^2 Q/\partial h^2)_{h=0} / (\partial Q/\partial h)_{h=0}$$

is the dimensionless variable representing the fractional increase in Q/h over its zero-field value. It is easily seen that the critical exponent for the temperature dependence of f is Δ. In Fig. 3 is plotted a full logarithmic graph of f evaluated at the maximum applied field of 10 T vs $T - T^*$. A linear fit gives a slope of $\Delta = 1.26 \pm 0.10$. This result is compatible with the tricritical hypothesis and totally inconsistent with the mean-field theory.

Another inadequacy of the mean-field theory is revealied by the magnitude of the quantity f. It may be readily shown that $f = hB/A^2$ in the de Gennes theory. Using values for A, B, and χ_a which have been determined for MBBA from a mean-field analysis of experimental data,[6] it may be concluded that $f \simeq 0.01$ at $T = T_c$. Thus, as may be seen from Fig. 3, the mean-field theory is more than two orders of magnitude in error in the estimation of the effect reported here. Stated in another way, f can also be shown to be given as $f = 3H^2/4H_c^2$ in the mean-field theory, where H_c is the value of the magnetic field needed to reach a nematic critical point.[1] It has been estimated[7] that H_c is about 100 T, which would lead to a value of $f \simeq 0.008$ at $T = T_c$. The enormous contrast between the mean-field estimate and the experimental findings give encouragement to the belief that the nematic critical point may actually be accessible with experimentally available magnetic fields. A search for this critical point is now under way.

With the mean-field theory having been proven inapplicable, it now becomes necessary to invoke more sophisticated theoretical techniques in order to find the true locations of the critical and tricritical points. The critical-point exponents and the region of crossover from critical to tricritical behavior have also yet to be worked out. What has been accomplished so far is the calculation of the nonclassical exponents associated with the Landau point, which is an isolated critical point produced by the accidental vanishing of the cubic term in the Landau–de Gennes expansion.[3, 8–11] Whether or not this special type of critical point is relevant to the experimental situation is an open question.

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Superflow in Restricted Geometries

R. J. Donnelly, R. N. Hills, and P. H. Roberts[(a)]
Institute of Theoretical Science and Department of Physics, University of Oregon, Eugene, Oregon 97403
(Received 16 October 1978)

We propose a "competing barrier model" for nucleation of quantized vortices in small channels. The results are compared to experiments on decay of persistent currents, critical velocities, onset temperatures, and the effective superfluid density at onset.

For a number of years experimentalists have tried to use the Iordanskiĭ-Langer-Fisher[1,2] (ILF) theory of fluctuation dissipation for superflow in an unbounded region (intrinsic nucleation) to fit experiments in restricted geometries. Several years ago, two of us (DR)[3] pointed out the necessity of considering a permanent barrier ΔE for flow in a restricted geometry which is present, irrespective of any superflow. For a toroidal channel of circular cross section with zero circulation, DR point out that the probability of nucleating to the wall a vortex of one sign is equal to that for a vortex of opposite circulation. However, when there is a superflow present, the energy barriers for nucleation of the two types of vortex are not the same: For a large superflow the process associated with one type of vortex will completely dominate the other, while for slower flows the nucleation of both types of vortex have to be considered. This is the "competing-barrier model" of nucleation.

In this Letter we show that the competing-barrier model will qualitatively explain experiments on decay of persistent currents as reported, for example, by Hallock and co-workers[4] and Kojima *et al.*[5] We confine ourselves to the broad issues using a simplified one-dimensional nucleation model, neglecting the geometrical differences among various sorts of porous materials and thin films.

The nucleation probability, P, per unit time can be written

$$P = f\exp(-\Delta F/kT), \tag{1}$$

where f is the temperature-dependent attempt frequency discussed and tabulated by DR and ΔF is the barrier height. For the ILF model, ΔF is due to the superflow alone. Here, $\Delta F = \Delta E$ when the superflow $v_s = 0$ and the critical momentum is then p_c. With a flow, the $\vec{p}\cdot\vec{v}_s$ interaction shifts the barrier and (considering only one dimension) we have $\Delta F = \Delta E \pm p_c v_s$ so that (1) becomes

$$P = 2f\exp(-\Delta E/kT)\sinh(p_c v_s/kT). \tag{2}$$

Equation (2) shows that the dimensionless quantity V ($\equiv p_c v_s/kT$), the ratio of ordered flow energy to fluctuation energy, is important.

Suppose the superflow takes place in a toroidal geometry containing n candidates per unit length for nucleation; then (cf. Ref. 3)

$$dv_s/dt = -nP\kappa, \tag{3}$$

where $\kappa \equiv h/m$. Equation (3) can be written nondimensionally as

$$dV/d\tau = -\sinh V \tag{4}$$

by introducing the dimensionless time $\tau = \nu_0 t$ and the nucleation rate ν_0:

$$\nu_0 = \nu\exp(-\Delta E/kT),\ \ \nu = 2n\kappa f p_c/kT. \tag{5}$$

Equation (4) has the solution

$$V = \ln[(1 + e^{-\tau}\tanh V_0/2)/(1 - e^{-\tau}\tanh V_0/2)], \tag{6}$$