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Experimental Determination of the Critical Correlation Function for a Binary Liquid Mixture: Evidence for Universality*

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Using light-scattering techniques we have measured the correlation function of a binary liquid near the critical point with a precision of 0.2%. The data cover a range $0.18 < k\xi < 26$ in terms of the scaling variable $k\xi$ and yield critical-exponent values in close agreement with recent theoretical calculations for the Ginzburg-Landau-Wilson model.

Near a second-order phase transition the intensity of scattered radiation for scattering vector $\vec{k}$ is proportional to the Fourier transform $\chi(k)$ of the order-parameter correlation function. Sufficiently close to the critical temperature T_c one expects the scaling form $\chi(k) = \Gamma t^{-\gamma} g(k\xi)$, where $t = (T - T_c)/T_c$, γ is the susceptibility exponent, ξ is the correlation length, and $g(x)$ is the correlation scaling function.[1] According to the universality hypothesis, systems having the same basic symmetries are expected to have identical critical exponents and scaling functions, and are said to belong to the same universality class.[2] This assumption implies that fluids near the gas-liquid critical point and binary liquids near the consolute point should belong to the same universality class as the three-dimensional Ising model. The critical exponents for this universality class have been calculated using different techniques.[3-6] The various calculations yield numerically similar results although some small unresolved discrepancies do exist.

During the past years the values reported for the critical exponents of gases as well as binary liquids appeared to deviate from those attributed to the Ising model. However, recent experiments indicate that the thermodynamic exponents β and γ for Xe, SF_6, and CO_2 [7] and β for isobutyric acid–water[8] and aniline-cyclohexane[9] do approach Ising-like values sufficiently close to the critical point.

In order to investigate further the hypothesis of universality, with particular emphasis on that for the critical correlation function, we have obtained a series of accurate light-scattering measurements for the binary liquid 3-methylpentane-nitroethane. Using photon counting techniques, the scattered intensity at a scattering angle of 90° was measured as a function of temperature. Data points taken in the range $1 \times 10^{-6} < t < 3 \times 10^{-3}$, corresponding to $0.18 < k\xi < 26$, were used in the analysis. Through a beam splitter, a reflector, and a diffuser, a small fraction of the incident light was delivered as reference light to the same photomultiplier measuring the scattering intensity. The photomultiplier was exposed, for a period of 100 sec, to either the scattered light or the reference light when one of the two shutters, installed in the two light paths, was opened. The data acquisition was automated with a real-time-operating minicomputer system. At each temperature, the mean of twenty consecutive normalized scattering intensities was taken as one datum point. A test run at a fixed temperature lasting 45 h yielded 41 data points with a standard deviation of 0.17% indicating the precision with which each datum point could be determined. The temperature of the sample cell was stabilized to within 0.2 mdeg using a two-stage thermostat. Further experimental details will be presented elsewhere.

When the precision of the data is a fraction of 1%, many small effects, previously ignored, must be taken into account. Therefore, the scattering intensity is to be compared with the expression

$$I = I_0(1 + at)t^{-\nu(2-\eta)} g(k\xi)\{1 - \tau(k\xi)[1 - R(k\xi)]\} + \Delta I. \tag{1}$$

I_0 is a proportionality factor, a is the temperature coefficient of the scattering intensity prefactor

$T\rho^{-1}(\partial\epsilon/\partial c)^2$ (ρ being the density, ϵ the dielectric constant, and c the concentration), $\tau(k\xi)$ is the turbidity correction, and $R(k\xi)$ is the double-scattering correction,[10] while ΔI represents the contributions from entropy fluctuations, Brillouin scattering,[11] and possibly cell wall scattering. These effects were either measured or estimated from known physical properties of the mixture. The magnitudes of the turbidity and double-scattering corrections are, respectively, about 4% and 1% at $k\xi = 10$ and 2% and 0.4% at $k\xi = 3$, and become progressively smaller as $k\xi$ decreases. The temperature dependence of the prefactor causes it to deviate from its value at T_c by 0.1% at $k\xi = 1$ and 2% at $k\xi = 0.18$. The extraneous contributions from entropy fluctuations and Brillouin scattering were found to be constant in the temperature range of the experiment and equal to about two thirds of the scattering intensity at $T - T_c = 10$ K.

Many light scattering experiments reported in the literature have been conducted at small values of $k\xi$ where the data are insensitive to the value of the exponent η. Furthermore, the interpretation of scattering experiments at intermediate and larger values of $k\xi$ was complicated in the past by the lack of a scaling function $g(x)$ of sufficient accuracy.[12] This deficiency may be remedied by using a "truncated Fisher-Langer" approximant which was recently proposed by one of the authors and which reproduces to high accuracy the theoretically known Ising-model correlation functions in two and $4-\epsilon$ dimensions.[13] To facilitate the analysis we use a form linearized in η valid for the small values of η considered here. The reciprocal of the scaling function is given by

$$g^{-1}(x) = 1 + x^2\left(1+\frac{x^2}{9}\right)^{-\eta/2}\left\{1+\eta\left[s_2(x) - \frac{w}{2}\ln\left(\frac{1+x^2/9}{1+x^2/36}\right)\right]\right\}, \qquad (2)$$

with

$$s_2(x) = \frac{x^2}{9}\int_1^\infty \frac{du}{u(u^2+x^2/9)}[1-F(u)].$$

Here $F(u)$ is the spectral function,[13] truncated so as to vanish for $u<2$, which is derived from the Fisher-Langer scaling function[14]

$$g_{\mathrm{FL}}(x) = (C_1/x^{2-\eta})(1 + C_2/x^{(1-\alpha)/\nu} + C_3/x^{1/\nu}), \text{ for } x \gg 1,$$

through the relation $\mathrm{Im}\, g_{\mathrm{FL}}^{-1}(i|x|) = [\sin(\pi\eta/2)/C_1]|x|^{2-\eta}F(|x|/3)$. The spectral function may be "fine tuned" by adding a constant contribution of strength $w \geq 0$ in the range $1<u<2$. A preliminary analysis indicated that ν and η are close to 0.625 and 0.015, respectively. Hence, we used an $s_2(x)$ evaluated with $\nu = \frac{5}{8}$ and $\alpha = 2-3\nu$, and the values $\eta = \frac{1}{54} \simeq 0.0185$, $C_2 = 1.773$, and $C_3 = -2.745$ derived from the ϵ expansion to $O(\epsilon^2)$.[15] The use of these values leads to a spectral function of the expected shape.[13] Since the best-fit values of the critical exponents were found to be insensitive to the choice of w within a physically reasonable range $[0 \leq w \leq F(2) \sim 0.17]$, the value $w=0$ was used in the final analysis. The closeness of the best-fit values of ν and η to those used in calculating $s_2(x)$ supports *a posteriori* the self-consistency of the analysis.

The experimental scattering data were fitted by (1) with scaling function (2) using the method of least squares with five adjustable parameters, namely I_0, ν, η, ΔI, and the correlation-length amplitude ξ_0. Eight experimental runs were available for this analysis, the results of which are presented in Table I. The values for γ and η_4, which are the basic exponents calculated by Baker *et al.*,[6] were deduced from ν and η using the scaling law $\gamma = \nu(2-\eta)$ and the definition $\eta_4 = \eta - 2 + 1/\nu$. The first six runs were obtained with the scattering beam located at the level where the meniscus would appear. In the last two runs this position was varied by 2 and 5 mm from the ear-

TABLE I. Experimental correlation function parameters.

Run	ν	η	ξ_0 (Å)	γ	η_4	r.m.s. error of fit (%)
1	0.6218	0.0288	2.324	1.2257	−0.3630	0.24
2	0.6295	0.0200	2.196	1.2463	−0.3913	0.23
3	0.6252	0.0169	2.275	1.2398	−0.3835	0.22
4	0.6227	0.0175	2.315	1.2344	−0.3765	0.12
5	0.6250	0.0058	2.319	1.2460	−0.3937	0.20
6	0.6278	0.0122	2.274	1.2479	−0.3949	0.15
7	0.6250	0.0108	2.313	1.2432	−0.3891	0.16
8	0.6237	0.0158	2.317	1.2376	−0.3809	0.18

lier position. The results appear to be indpendent of the level of the beam, indicating that gravity effects were negligible within that region of the sample. A run obtained by deviating the beam 11 mm away from the central position did yield slightly different results. This is exactly what one would expect since gravity-induced gradients start from the top and the bottom of the cell leaving the central section unaffected[16] during the course of our experiment.

The average values and the standard deviations of the critical exponents from the eight runs are

$$\nu = 0.625 \pm 0.003, \quad \gamma = 1.240 \pm 0.007, \\ \eta = 0.016 \pm 0.007, \quad \eta_4 = -0.384 \pm 0.010. \tag{3}$$

When an extended scaling correction factor $1 + Ct^{\Delta_1}$ with $\Delta_1 = 0.5$[6,17] was included in the analysis, we obtained $C = 0.3 \pm 0.3$ with insignificant shifts in the exponent values, indicating that the data are within the range of asymptotic simple scaling.

Our results are rather similar to the exponent values calculated by Baker *et al.* from the Ginzburg-Landau-Wilson model using the Callan-Symanzik equation[6]:

$$\nu = 0.627 \pm 0.01, \quad \gamma = 1.2410 \pm 0.002, \\ \eta = 0.021 \pm 0.02, \quad \eta_4 = -0.3842 \pm 0.003. \tag{4}$$

We should mention that the least-squares procedure yielded unrealistically small standard deviations of 0.001 for both ν and η for the individual runs. The errors quoted in (3) represent the standard deviations when the exponent values are averaged over the eight different determinations presented in Table I. More conservative estimates are obtained by allowing for two standard deviations, leading to $\nu = 0.625 \pm 0.005$ and $\eta = 0.016 \pm 0.014$. Thus, our work shows η to be significantly smaller than the values 0.11 ± 0.03 and 0.10 ± 0.05 previously reported for fluids on the basis of neutron scattering[18] and x-ray scattering[19] experiments which also probe the correlations at $k\xi > 1$. We also compared our data with the predictions of series expansions[3,4] by fixing ν and η at 0.638 and 0.041 and using an appropriate scaling function.[13] For comparison, two deviation plots of a representative experimental run (run No. 3) are presented in Fig. 1. Whereas the upper plot with $\nu = 0.625$ and $\eta = 0.017$ does not show any significant deviations, small but noticeable systematic deviations do appear in the lower plot with $\nu = 0.638$ and $\eta = 0.041$. Our data thus seem to support the Ising-model exponents as computed by Baker *et al.*[6]

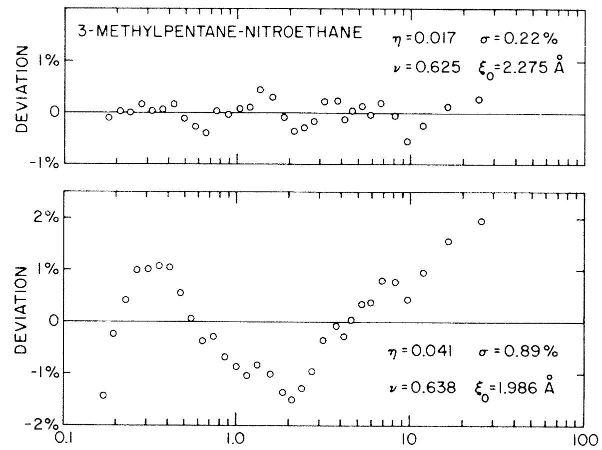

FIG. 1. Percentage deviations between the measured scattering intensity and the theoretical prediction using two different choices for ν and η.

The close agreement between our results and those of Baker *et al.* indicates that binary liquids and the Ising model belong to the same universality class. In conclusion we note that our experiments not only yield critical exponent values but also the critical correlation function for this universality class.

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Stability of Persistent Currents in Unsaturated Superfluid ^{4}He Films*

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We report the first measurements of the stability of persistent currents in unsaturated ^{4}He films in an open geometry. Persistent currents are generated without rotation and exhibit several features which place strong restrictions on models of thermally activated dissipation in thin films.

We report the first observation of persistent currents in unsaturated superfluid ^{4}He films on a large-diameter glass substrate in an open geometry. Although such currents have been observed in filter material[1] and Vycor,[2] previous attempts to observe these currents in similar open geometries have not been successful.[3,4] This led to the suggestion that high-quantum-number circulation states may be very unstable.[4] Recent experiments[5,6] in saturated films have shown this suggestion to be incorrect for those films. We present here the first measurements of the stability of persistent currents in unsaturated superfluid films as a function of film thickness at constant temperature and observe a strong thickness dependence for films thinner than about 12 atomic layers. Currently it is believed that thermally activated dissipation is responsible for the decay of persistent currents near critical velocity.[7] The observed behavior of our decays places strong restrictions on models used in this theory in the case of films. The decay of the flow velocity becomes immeasurably small for films thicker than 12 atomic layers. For these thicker films we find the circulation stable with respect to translation and to changes in the film thickness through condensation and evaporation.

The persistent currents are produced on the outside of a cylindrical Pyrex glass ring with posts attached to it schematically shown in Fig. 1(a). To produce the thin films investigated here the entire apparatus is enclosed in a copper cell where the pressure is maintained at a value below the saturated vapor pressure at the chosen operating temperature of 1.45 K. The film thick-

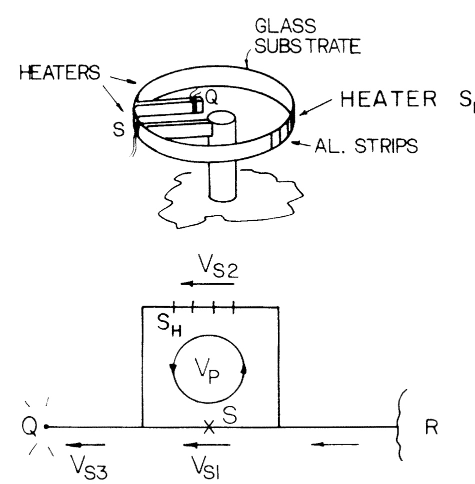

FIG. 1. (a) Pyrex glass ring substrate of average perimeter $l_1+l_2=21.15\pm0.2$ cm. The long- to short-flow-path ratio l_2/l_1 is 15.63 with height of 0.73 cm and thickness of 0.24 cm. The aluminum strips are on the outer side and directly opposite the two posts. (b) Schematic of the flow paths.