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Light Scattering by ^3He-^4He Mixtures near the Tricritical Point

P. Leiderer, D. R. Watts,* and W. W. Webb
School of Applied and Engineering Physics and Laboratory for Atomic and Solid State Physics, Cornell University, Ithaca, New York 14850
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Scattering intensity and related measurements have established the tricritical exponents for the divergences of the concentration susceptibility, γ, and of the correlation length for concentration fluctuations, ν, have indicated a probable value for the exponent η, and have established a region of tricritical scaling.

We have measured the intensity of light scattering by liquid isotropic mixtures of ^3He and ^4He around the tricritical point (T_t, x_t), where the λ line delineating the concentration-dependent temperature $T_\lambda(x)$ of the continuous superfluid transition terminates at the top of a miscibility gap and the mixture begins a first-order separation into two phases, one a normal ^3He-rich fluid and the other a ^4He-rich superfluid.[1-3] Light-scattering intensities give a direct measure of the concentration susceptibility $(\partial x/\partial\Delta)_{TP}$ near T_t in the small-scattering-vector limit $(q\to 0)$ in contrast with experiments on specific heat[2] and saturated vapor pressure,[4] and the q dependence gives the correlation length ξ for concentration fluctuations. Here x is the atomic fraction of ^3He and $\Delta = \mu_3 - \mu_4$ is the difference between the chemical potentials of ^3He and ^4He.

The scattering power is proportional to the extinction length

$$h = A(\lambda)\frac{k_B T}{v^2}\left[\frac{1}{v}\left(\frac{\partial v}{\partial x}\right)_{TP}^2\left(\frac{\partial x}{\partial \Delta}\right)_{TP} + \beta_{Tx}\right], \qquad (1)$$

where $A(\lambda) = (2\pi/\lambda)^4[4\pi\alpha N_0)^2/6\pi]$ and $\beta_{Tx} = -(1/v)(\partial v/\partial p)_{Tx}$. Here v is the molar volume,[5] αN_0 is the molar polarizability of helium, and λ is the light wavelength. The isothermal compressibility β_{Tx} varies only weakly near the tricritical point (T_t, x_t),[6] whereas the concentration susceptibility $(\partial x/\partial\Delta)_{TP}$ is expected to diverge. Since the atomic volumes of ^3He and ^4He differ, concentration fluctuations dominate the scattering and the critical opalescence is determined by the divergence of $(\partial x/\partial\Delta)_{TP}$.

In summary, our results are as follows: First, we found a steep increase in scattered intensity on cooling just below the λ line and an accompanying local scattering maximum at $x - x_t > 0.01$. Second, on approaching T_t we found a divergence of the scattering intensity indicating $\partial x/\partial\Delta \propto \epsilon^{-\gamma}$, where $\epsilon = |T_t - T|/T_t$ and $\gamma = 1.0$ with high accuracy. Third, we found evidence for a strong tricritical divergence in the superfluid phase of the correlation length ξ for concentration fluctuations that is consistent with model calculations of Furman and Blume.[7] The combined experiments provide a satisfactory test and confirmation of recent theoretical discussions on tricritical scaling.[8-12] Finally to cope with substantial concentration gradients near (T_t, x_t) due to the earth's gravitational field, we have measured them interferometrically and have determined the tricritical point and neighboring coexistence curve by using a reflectometric technique that is immune to gravitational perturbations. Our preliminary reports are corrected here.[13-15]

In our experiments, the temperature was (necessarily) controlled to better than $\pm 1\,\mu K$ for a few hours to attain demonstrable equilibrium, and was calibrated to ± 0.001 K absolute by vapor-pressure thermometry. Laser power was limited to avoid effects of heating at windows. Temperature differences $T_t - T$ were established to better than ± 0.1 mK, as described below. Actual compositions, x_1 to x_5, were known to ± 0.0016 absolute and ± 0.0007 relative to each other. (Watt's thesis provides some experimental details[15] and more will be published elsewhere.)

Preliminary light-scattering data[12] on four compositions around x_t were obtained with the scattering volume positioned at two fixed heights for measurements in coexisting normal and superfluid phases. They coincide in the phase-separated region only until gravitational effects begin to grow near T_t. A sharp step in scattered intensities occurs in the mixtures with $x < x_t = 0.6750$[3] as they reach the transition from superfluid to normal fluid at their λ temperatures. In the scattering by the mixture $x_1 = 0.6318$ we noted a small, reproducible, nonhysteretic, rounded maximum slightly below $T_\lambda(x_1)$. This maximum is about 5% above the intensity at $T \lesssim T_\lambda$ and about 10 mK wide, and was not observed closer to x_t in the mixture $x_2 = 0.6623$.[16] The origin of the maximum is uncertain; it implies a weak singularity, in either $(\partial x/\partial\Delta)_{TP}$ or β_{Tx}, that vanishes as $x \to x_t$.[16]

Strong gravitational effects on measurements near the tricritical point are expected as a result of vertical gradients in composition induced by the gravitational potential whenever the concentration susceptibility is large.[17,18] There is a predicted concentration change with height of $\delta x(z) \propto z^{1/\delta} \sim z^{1/2}$. We determined $\delta x(z, T)$ to about 0.001 over a range ~4 mm by using a modified Jamin interferometer to measure changes of the index of refraction n, and thus composition changes by using $dn/dx \cong -0.0070$ near (x_t, T_t). From the results we estimated corrections for gravitational effects in the phase diagram reported by Alvesalo *et al.*[2] and confirmed the diagram given by Ahlers and Greywall[3] in clear preference to all others proposed.[1,2]

Because these effects obfuscate determination of the tricritical point, we have also redetermined it. We measured the limiting angle of total reflectivity of the interface in the phase-separated mixture, obtaining values for the discontinuity of the refractive index across the interface and hence the concentration difference $\Delta x = x_n - x_s$ between normal and superfluid coexisting phases. Δx was found to decrease linearly on approaching T_t with exactly the absolute value expected from the corrected phase diagram. Thus we confirm $\Delta x \propto \epsilon^\beta$ where the tricritical exponent is precisely $\beta = 1.00$ in the range $8\times10^{-4} \leq \epsilon \leq 10^{-2}$. The value of the tricritical temperature was determined as $T_t = 0.866 \pm 0.001$ K, in good agreement with only the result of Ahlers and Greywall, 0.867 K.[3] This experiment provides calibration for temperature differences from T_t to better than 0.1 mK, and determines the phase diagram free of gravity perturbations, since only the interface itself is involved in the measurement.

To avoid the gravitational effects on light scattering for measurements very close to T_t in the mixture $x_5 = 0.6749$ ($x_t = 0.6750$)[3] the laser beam was focused and scanned vertically across the sample cell. In this was the scattered intensity was monitored $\lesssim 0.2$ mm from the interface, thus providing data essentially free of gravity effects when $T_t - T \gtrsim 0.002$ K. The wave-number dependence in x_5 was determined by using two wavelengths, $\lambda = 6328$ and 4880 Å, and two scattering angles, $\theta = 90$ and 15°, thus providing data at four wave numbers. Apparent concentration susceptibility data $(\partial x/\partial\Delta)_{TP}$, shown in Fig. 1, were

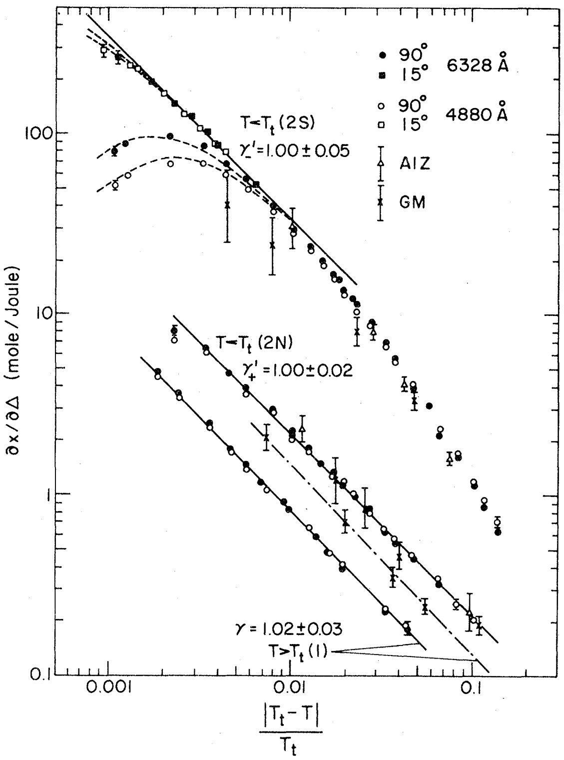

FIG. 1. Log plot of $(\partial x/\partial\Delta)_{TP}$ versus ϵ for the single phase (1), and for the coexisting normal (2N) and superfluid (2S) phases for the mixture $x_5 = 0.6749$. For comparison, data of Alvesalo *et al.* (AIZ) and Goellner and Meyer (GM) (Refs. 2 and 4) are shown. The dashed lines show the best fit to our data calculated according to Eq. (2). At $\epsilon \lesssim 10^{-3}$, the remaining small discrepancy is attributable to gravity effects.

obtained from the scattered intensities by using Eq. (1), i.e., by heuristically assuming isotropic scattering for the $q \to 0$ limit. The results were fitted by the expected power-law divergence of the form $(\partial x/\partial\Delta)_{TP} = G\epsilon^{-\gamma}$.

For the single-phase critical isochore and the normal coexisting phase, power laws were found to hold in the entire investigated temperature interval. In contrast, the tricritical region for the superfluid seems to be confined to $T_t - T \leq 10$ mK, about the same temperature interval in which the phase diagram is linear. The best-fit critical exponents γ and prefactors G (in mole/J) are $\gamma = 1.02 \pm 0.03$, $G = (7.7 \pm 1)\times10^{-3}$, $\gamma_+' = 1.00 \pm 0.02$, $G_+' = (2.2 \pm 0.2)\times10^{-2}$, $\gamma_-' = 1.00 \pm 0.05$, $G_-' = (3.2 \pm 0.6)\times10^{-1}$, where γ and G apply at $T > T_t$ along the critical isochore, and γ_+' and G_+', and γ_-' and G_-' apply at $T < T_t$ in the normal and superfluid coexisting phases, respectively. Values of $(\partial x/\partial\Delta)_{TP}$ obtained from the specific-heat data of Alvesalo *et al.*[2] and from the saturated-vapor-pressure data of Goellner and Meyer[4] are included in Fig. 1 for comparison.

The onset of q dependence of the 90° scattering intensities in the coexisting superfluid is clearly evident in Fig. 1 for $\epsilon \lesssim 0.007$. We compared the scattered intensities in the coexisting superfluid phase with the modified Ornstein-Zernicke expression for the correlation function obtained from the Blume-Emery-Griffiths[12] model by Furman and Blume.[7] Their Fourier transform $C(\vec{q}, \epsilon)$ of the two-point correlation function, which is proportional to the scattering intensity, has the form $C(\vec{q}, \epsilon) \approx (aq^2 + b\epsilon)/(aq^2 + c\epsilon^2)$, where a, b, and c are constants given by the model. We find that our data can be well represented by this expression. The fit to our data at four values of q shown in Fig. 1 yields $a/c = 1.6 \times 10^{-16}$ and $aq^2 \ll b\epsilon$ for $\epsilon > 10^{-3}$.

Therefore, for $\epsilon > 10^{-3}$, we have[19]

$$C(\vec{q}, \epsilon) \approx \frac{\text{const}}{q}\left(\frac{q/\epsilon}{1.6 \times 10^{-16} q^2/\epsilon^2 + 1}\right). \qquad (2)$$

If we *define* the critical exponents η and ν by writing $C(\vec{q}, \epsilon) = (1/q^{2-\eta}) F(q/\epsilon^{\nu})$,[10] we find that our experimental results are consistent with the critical exponents $\nu_-' = 1$ and $\eta_-' = 1$, which together with $\gamma_-' = 1$ fulfill the scaling relationship $\gamma = (2 - \eta)\nu$. Thus the correlation length ξ_-' for concentration fluctuations in the superfluid phase (*not* the superfluid correlation length) is given by $\xi = 1.3\epsilon^{-1}$ Å. The tricritical exponents for ^{3}He-^{4}He mixtures below T_t are thus $\beta = \gamma_\pm' = \nu_-' = \eta_-' = 1.0$ consistent with tricritical scaling. Note that the Furman-Blume form is consistent with simple scaling only in the region $aq^2 \ll b\epsilon$.

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*Now at Department of Geology and Geophysics, Yale University, New Haven, Connecticut 06520.

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[19]To allow for some modification of this expression, which still leads to $C(0,\epsilon) \propto \epsilon^{-1}$, we set $C(\vec{q},\epsilon) \propto \epsilon^{-1}/(mq^n/\epsilon^2 + 1)$. The best fit was obtained for $m = 1.6 \times 10^{-16}$ and $n = 2.0$.