Rayleigh-Linewidth Measurements near the Critical Point of a Binary Fluid

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We report careful measurements of the Rayleigh linewidth of light scattered from a critical binary fluid. Laser heating and multiple-scattering effects are accounted for. With use of independent data for ξ and the shear viscosity, a parameter-free fit is obtained with theory in the region $55 > k\xi > 0.32$. The renormalization-group–modified mode theory is shown to be necessary; however, we find a value of the dynamical exponent $z = 2.992 \pm 0.014$ instead of 3.065.

In this Letter we report new and careful measurements of the Rayleigh linewidth of light scattered from the binary fluid 3-methylpentane + nitroethane near the critical point. The need for this experiment has arisen on two fronts. Theoretically, the predictions of the mode theories,[1-5] dynamic droplet model,[6] and renormalization-group mode (RG mode) theory,[7] while similar, all differ in important aspects. To properly test these theoretical predictions, two other independent experimental determinations must be made. First, static light-scattering measurements must be made to yield values for ν and ξ_0 in the relation for the correlation length $\xi = \xi_0 t^{-\nu}$, where $t = (T - T_c)/T_c$. Second, accurate measurements of *both* the macroscopic shear viscosity and the background viscosity must be made. With this information a parameter-free test of the linewidth theories may be made (for a review see Swinney and Henry[8]).

Experimentally, we have found in previous work[9,10] that a number of problems may arise when making linewidth measurements very near T_c. Excellent temperature stability is necessary. In our experiment we have achieved stability of ± 40 μK/day and reduced gradients to $\leqslant 40$ μK/cm. The effect of heating by the incident laser beam, which has been ignored by previous workers, must be considered. We have measured the heating effects in our system using a technique described in another paper.[10] We have found ~ 0.13 mK/mW of heating by the incident beam. We used this knowledge to adjust the incident power so that the heating effect was small compared to $T - T_c$. Finally, by careful visual observation we were able to determine T_c itself with a precision of ± 50 μK.

Although the indices of refraction of the component fluids are closely matched, multiple scattering becomes an important consideration near T_c. We have determined the relative magnitude of the double- to single-scattered light intensities, I_2/I_1, by measuring the depolarization ratio of the scattered light and applying a second-order analysis method.[9,10] We found at $\theta_{\rm scat} = 90°$ a range of values from $I_2/I_1 = 9\%$ at $T - T_c = 1$ mK to 2.9% at 18 mK. With extrapolation to $T - T_c \ll 1$ mK, theoretical[11] considerations indicate $I_2/I_1 \simeq 15\%$. These ratios would be smaller at smaller $\theta_{\rm scat}$. This low level of multiple scattering combined with our knowledge of the similarity between the single- and double-scattered linewidths[9] led us to conclude that multiple-scattering effects were minimal.

Autocorrelation measurements were performed and the scattered-light correlation function was fitted by a two-cumulant fit. We found the ratio of the first two cumulants to be $K_2/K_1^2 \simeq 0.03$, indicating exponentiality. When $T - T_c \leqslant 1$ mK, $K_2/K_1^2 \simeq 0.05$ but was still within our experimental error. The fluid system was always agitated several hours before a run to insure against gravitational gradients. 72 acceptable Rayleigh linewidth determinations were made at six different k ($\vec{k}$ being the scattering wave vector) in the range 0.15 ± 0.07 mK $\leqslant T - T_c \leqslant 263 \pm 1$ mK ($5\times 10^{-7} \leqslant t \leqslant 8.8 \times 10^{-4}$).

Our first analysis concerns the dynamic exponent z in the ralation $\Gamma \propto k^z$, where Γ is the linewidth. Recent RG calculations[7] have indicated $z = 3.065$ when $k\xi \gg 1$. To test this we have least-squares fitted the forty data points for which $55 \geqslant k\xi \geqslant 10$ with this relation ($\xi_0 = 2.275$ Å and $\nu = 0.625 \pm 0.003$ as determined by Chang *et al.*[12]). We found $z = 2.995 \pm 0.018$ with a standard error (S.E.) of fit of 3.9%. The errors quoted in this paper represent one standard deviation. We also performed this fit to the 32 data points for which 0.15 ± 0.07 mK $\leq T - T_c \leq 0.38 \pm 0.07$ mK and found $z = 2.988 \pm 0.019$, with an S.E. of 3.5%. Fits to the data for which $k\xi \geq 20$ and $k\xi \geq 30$ showed no trend for z to increase to 3.065. We conclude that our data indicate $z = 2.992 \pm 0.014$, more than three standard deviations from $z = 3.065$.

We fitted all the data with the various theoretical forms. The dynamic-droplet-model prediction for the Rayleigh linewidth is[6]

$$\Gamma_{\rm DM} = (\gamma k_{\rm B} T / 6\pi\eta_b\xi) k^2 (1 + \xi^2 k^2)^{1/2}, \quad (1)$$

where $k_{\rm B}$ is Boltzmann's constant, T is the absolute temperature, and η_b is the *background* viscosity. γ is an extra factor to be described below.

The mode theoretical form for Γ is[1-4]

$$\Gamma_M = (\gamma k_{\rm B} T / 6\pi\eta\xi^3) K_0(k\xi)\, R(k\xi), \quad (2)$$

where

$$K_0(x) = \tfrac{3}{4}[1 + x^2 + (x^3 - x^{-1}) \arctan x]. \quad (3)$$

η is now the macroscopic viscosity and $R(x)$ is the nonlocal viscosity factor best described by Oxtoby and Gelbart.[4] Because (2) gives essentially identical results as the decoupled-mode theory,[5] we shall not consider that form explictly here.

The viscosity data of Stein, Allegra, and Allen[13] and Tsai and McIntyre[14] for this fluid system indicate essentially identical critically divergent parts. The background viscosity measurements, however, differ, the data of Tsai and McIntyre being 4.1% smaller. This is not surprising since we expect the critical part to be universal and the background to be system or preparation dependent. Thus, it is important to make independent viscosity measurements on our particular fluid system to insure against systematic errors that might result from using either set of data.

Sixteen viscosity data points in the region 0.25 K $\leq T - T_c \leq 13$ K were obtained with a calibrated Poiseuille flow viscometer with an accuracy of 1%.[15] We found our data to be systematically (12 ± 1)% larger than those found by Tsai and McIntyre (7.2% larger than the data of Stein, Allegra, and Allen) including the region where the divergence of the viscosity is apparent. Subtraction of a nonanomalous background contribution from each data set showed that the critically divergent parts were essentially equal to 1%; equality expected by universality. All three viscosity measurements indicate that while the background parts may vary slightly, the critical parts are identical. In fitting our linewidth data, we used our viscosity measurements on our system extrapolated into the $T - T_c \leq 0.25$ K region by the functional form for the universal critical part as determined by Tsai and McIntyre.

The measured values in our linewidth fits were Γ, η, and k. The adjustable parameters were ν, ξ_0, and γ. If ν and ξ_0 agreed with the values from previous accurate static intensity measurements, we considered the fit successful.

We fitted the dynamic droplet model (1) to all 72 data points using $\eta_b = 0.409\,[1 - 0.009\,(T - T_c)]$ cP. We found $\nu = 0.667 \pm 0.023$, $\xi_0 = 1.77 \pm 0.30$ Å, and $\gamma = 1.03 \pm 0.01$ with an S.E. of 4.55%. A residuals plot is given in Fig. 1 and the fit is seen to be free of residuals. The errors in ν and ξ_0 represent one standard deviation when both ν and ξ_0 vary. This error estimate is the most realistic because ν and ξ_0 are highly correlated. Neither ν nor ξ_0 agree with the static light-scattering results of Chang *et al.*,[12] who found $\nu = 0.625 \pm 0.003$ and $\xi_0 = 2.275$ Å. However, it is interesting to note that since we used the nondivergent background viscosity in Eq. (1), we might expect that $\nu_{\rm DM} = \nu + \chi_\eta = 0.04$ (Ref. 7) is the viscosity exponent. Therefore, $\nu = 0.627$. Thus, despite its

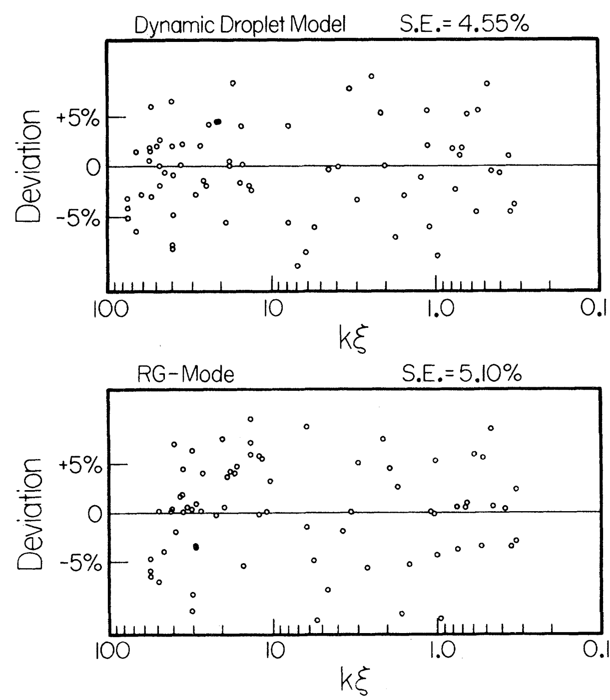

FIG. 1. Percentage deviations between the measured Rayleigh-linewidth and the theoretical forms.

problems[16] the droplet model does yield a good fit.

We next fitted the mode theoretical prediction (2) to the data. The function $R(x)$ is a nonuniversal function dependent on the background Rayleigh linewidth, which for our fluid system is quite small. Our data having experimental errors of ~4% were not precise enough to detect the $2\frac{1}{2}\%$ change in $R(x)$ from the no-background to large-background limits.[4] We therefore considered $R(x)$ in the no-background limit.

The functional form of the shear viscosity used will have an effect on the linewidth fits. We used two of the functional forms successfully used by Tsai and McIntyre.[14] First, using their fit to the power-law form $\ln\eta = A\ln t + Bt + C$, we found $\nu = 0.627 \pm 0.018$, $\xi_0 = 2.29 \pm 0.30$ Å, and $\gamma = 1.19 \pm 0.01$ with an S.E. of 5.1%. Using their fit to the logarithmic form $\eta = A'\ln t + B't + C'$ we found $\nu = 0.615 \pm 0.019$, $\xi_0 = 2.50 \pm 0.36$ Å, and $\gamma = 1.18 \pm 0.01$ with an S.E. of 5.1%.

We now examine and discuss the mode-theory results. The factor $\gamma = 1.18-1.19$ agrees very well with the RG prediction of $\gamma = 1.20$.[7] This represents the first definitive observation of this predicted factor. Next, if we take the average for the two fits, we find $\nu = 0.621 \pm 0.015$ and $\xi_0 = 2.40 \pm 0.25$ Å. These values are in excellent agreement with the static light-scattering results of Chang *et al.*,[12] who found $\nu = 0.625 \pm 0.003$ and $\xi_0 = 2.27$ Å. If we were to consider only the power-law viscosity fit, which is the expected RG dependence,[7] the agreement is excellent. A residuals plot of this fit is also given in Fig. 1. These results imply that, given the values of ν and ξ_0 found from accurate static measurements of the light-scattered intensity and the viscosity measured by us to 1%, the RG mode theoretical form, Eq. (2), with $\gamma = 1.19 \pm 0.01$ accurately describes our linewidth data with residuals indistinguishable from those shown in the lower half of Fig. 1. Because of this excellent agreement using the experimentally determined macroscopic viscosity, we can claim that a *parameter-free* fit of the linewidth data has been achieved.

Our value of the dynamic exponent $z = 2.992 \pm 0.014$, however, is 3 standard deviations from the RG prediction of 3.065. Two possible experimental explanations may be advanced. First, multiple scattering may affect the determination of z. Theoretically,[9] $\Gamma_2 \propto k^3$. If $I_2/I_1 \simeq 15\%$ for $k\xi \gg 1$ as indicated by our measurements, a simple weighted-average argument suggests an observed $z = 3.005$ which was detectable but not seen, eliminating that argument. A more likely explanation is that $k\xi$ is not yet large enough ($k\xi \lesssim 55$) to be in the region where $z = 3.065$.[7]

In conclusion, we have performed careful Rayleigh-linewidth measurements on the binary fluid system, 3-methylpentane + nitroethane, in the critical region. We found that the RG mode theory describes the data free of all parameters using independently determined values for ν, ξ_0, and the macroscopic viscosity. In particular, use of the experimentally determined viscosity was shown to be necessary to detect, for the first time, the extra factor of 1.2 indicated by RG. The dynamic exponent,[17] however, was found to be $z = 2.992 \pm 0.014$ contrary to the predicted $z = 3.065$.

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Dissipation near the Critical Point of a Two-Dimensional Superfluid

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We show that in a nonequilibrium, two-dimensional superfluid near the critical point, the energy for dissipative fluctuations is greatly reduced as a result of screening by the gas of bound vortex pairs, leading to an enhanced decay of the superfluid velocity. We also show that the superfluid density is reduced by a velocity-dependent amount as T_c is approached from below.

^{4}He films of few monolayers in thickness are of considerable interest in view of current progress in the understanding of critical phenomena in the two-dimensional X-Y model.[1,2] Experiments by Telschow and Hallock[3] and Bishop and Reppy[4] on the low-temperature properties of few-monolayer He films on a Mylar substrate suggest that dissipation of the two-dimensional (2-D) superflow in the onset transition region is much larger than the dissipation of dilute three-dimensional-type superfluid in Vycor substrates.

The purpose of this Letter is to point out that this qualitative difference between the dissipation for two-dimensional versus three-dimensional systems can be understood in terms of the fact that the critical fluctuations in a 2-D superfluid may be represented by the thermodynamics of a gas of vortex pairs interacting through long-range logarithmic potentials.

The decay of the nonequilibrium current-carrying state of a superfluid in three dimensions proceeds via the nucleation of vortex rings of a certain critical size.[5] In two dimensions the analogous mechanism is that of nucleation of vortex pairs with a critical separation normal to the superflow. We show that for a two-dimensional superfluid near the critical point the energy for creation of vortex pairs is greatly reduced as a result of screening by the gas of bound vortex pairs in a way similar to the 2-D Coulomb gas. It is this screening which reduces the free-energy barrier for nucleation of vortex pairs of critical size and hence increases the dissipation of two-dimensional superflow. We also show that the average number of vortex pairs whose size is below the critical distance in the metastable state is an increasing function of superfluid velocity, so that the superfluid density is reduced by a velocity-dependent amount as the critical temperature is approached from below.

We consider a 2-D superfluid[6] in a metastable state characterized by a superfluid velocity V_s. In the spirit of the Vinen theory[7] as elaborated by Iordanskii[8] and Langer and Fisher[9] we assume that the excitation of a vortex pair with mean separation $y>y_0$ perpendicular to the direction of superflow will reduce V_s by an amount h/mA, with A the dimensions of the film. The mechanism behind this decay process is analogous to the droplet model of homogeneous nucleation, i.e., fluctuation vortex pairs with distances $y<y_0$ tend to coalesce whereas those pairs generated at distances beyond y_0 will tend to expand indefinitely and crash against the boundaries, thereby providing a channel for flow dissipation. The critical distance y_0 is determined by considering

