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Correlation range and Rayleigh linewidth of xenon near the critical point

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Accurate measurements are reported of both the correlation range and the Rayleigh linewidth of xenon near its critical point. The linewidth data agree quite well with the mode-mode coupling results, and disagree with renormalization-group dynamics calculations.

In a recent letter[1] Sorensen *et al.* reported measurements of the Rayleigh linewidth and viscosity of a binary fluid mixture very near its critical point. Their measurements were consistent with renormalization-group dynamics calculations,[2] but were not consistent with the predictions of Kawasaki's mode-mode coupling calculations[3,4] as modified by Oxtoby and Gelbart.[5]

We report here the results of a very careful study of both the long-range correlation length and the Rayleigh linewidth of the pure fluid xenon near its critical point. Since both mixtures and pure fluids belong to the same universality class[6] one would expect similar behavior for the dynamics of the order-parameter fluctuations in both cases.[2] We find, however, that our data are in good agreement with the mode-mode coupling results, and do not support the conclusions reached by Sorensen *et al.*

Both the correlation range and linewidth measurements were made at the critical density for $T > T_c$. The correlation range measurements were made using a precise differential technique and apparatus which have been described previously.[7,8] They covered the temperature range $0.0026 \leq T - T_c \leq 10$ K, which corresponds to $9.0 \times 10^{-6} \leq t \leq 3.4 \times 10^{-2}$, and are accurately represented by the expression

$$\xi = 1.93 t^{-0.62} \text{ Å} . \quad (1)$$

The linewidth measurements were made using the same apparatus with the addition of a multichannel spectrum analyzer to determine the power spectrum of the photocurrent. The analyzer was calibrated to an absolute accuracy of better than 2%, and all spectra were Lorentzian to within that accuracy. The measurements were made for two different values of the scattering wave vector $q_1 = 2.59 \times 10^4$ cm^{-1} and $q_2 = 2.24 \times 10^5$ cm^{-1} in the reduced temperature range $6.2 \times 10^{-6} \leq t \leq 3 \times 10^{-3}$.

For both linewidth and correlation range measurements made within 0.03 K of T_c, a scattering cell with an internal length of only 1.1 mm was used. This was sufficient to effectively eliminate the problems of multiple scattering and beam bending previously encountered[9] near T_c. All measurements were made with the sample in thermal equilibrium, the establishment of which required ~24 hours for temperatures near T_c. No effects due to heating of the sample by the incident beam were observed.

The predictions of both the mode-mode coupling theories[3,4,5] and the renormalization-group mode dynamics calculations,[2] for the critical part of the linewidth Γ_c, can be represented by,

$$\Gamma_c = R \frac{k_B T q^3}{\bar{\eta}} \left(\frac{K_0(q\xi)}{(q\xi)^3} R(q, \xi) \right) . \quad (2)$$

Here $\bar{\eta}$ is the macroscopic shear viscosity, $R(q, \xi)$ is a weakly nonuniversal function described by Oxtoby and Gelbart,[5] and

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

The numerical constant R has the value $(6\pi)^{-1}$ in the mode-mode coupling theories, which are very nearly identical to the decoupled-mode results of Perl and Ferrell,[10] while the renormalization-group calculations yield $R = 1.2(6\pi)^{-1}$.

We used the methods and auxiliary data tabulated by Swinney and Henry[11] to obtain Γ_c values from our measured linewidths. The temperature-dependent background linewidth Γ_B, which must be subtracted from the measured linewidths, ranged from a few percent of the total very near T_c, to ~40%, 0.9 K from T_c. Fortunately, accurate values[12] for both Γ_B and $\bar{\eta}$ are available for xenon, which allows a direct parameter-free comparison between our data and the theories. This comparison is displayed in Fig. 1, which shows the percent deviation between Γ^* and $K_0(q\xi)R(q, \xi)/(q\xi)^3$, where $\Gamma^* = \Gamma_c/(Rk_BTq^3/\bar{\eta})$, with $R = (6\pi)^{-1}$. A least-mean-squares fit with R adjustable yields $R = 1.01(6\pi)^{-1}$, which clearly disagrees with the result $1.20(6\pi)^{-1}$ predicted by the renormalization-group dynamics calculations.[2] Since the combined errors in our measurements and the viscosity measurements are less than five percent, the disagreement is certainly real.

Clearly the deviations between the data and the

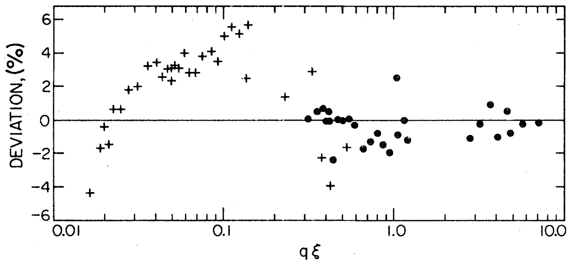

FIG. 1. Percentage deviation between the measured and mode-coupling theoretical values for the reduced linewidth Γ^* as a function of $q\xi$. The crosses correspond to $q=2.59\times10^4$ cm^{-1}, and the circles correspond to $q=2.24\times10^5$ cm^{-1}. See text for discussion.

theory are systematic for $q\xi \leq 0.1$; however, we have observed similar deviations with both sample cells, after completely realigning the optics, and thus the effect is almost certainly not attributable to systematic errors in our measurements. The negative deviations observed at very small $q\xi$ become more pronounced as $q\xi$ decreases, but are quite probably attributable to small errors in the background linewidth Γ_B, which becomes a large fraction of the measured linewidth under these conditions. An additional source of slight systematic deviations could exist in that the theoretical expression of Eq. (2), neglects the effects associated with a non-Ornstein-Zernike form of the correlation function,[13] and also neglects so called "vertex corrections.[14]" Neither effect should be more than 2–3%, however.

Theory also predicts that in the limit $q\xi \gg 1$, $\Gamma_c \sim q^z$, with $z \simeq 3.06$; however, our data at the scattering wave vector q_1 extend only to $q\xi = 0.53$, and thus we cannot test this prediction.

The fact that our data and those of Sorensen *et al.*[1] for two different systems of the same universality class, do not agree, seems to us much more serious than the failure of the renormalization-group dynamics calculations to describe our data exactly, since the result for the prefactor R might be changed by further calculations, but any such calculations would presumably apply equally to mixtures and pure fluids. After completing these measurements, however, we learned that Burstyn, Sengers, and Esfandiari[15] have made linewidth measurements on the same binary mixture studied by Sorensen *et al.*[1] with results very similar to our own, and thus the conflict is presumably between experiment and theory and not between the two different critical systems.

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