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PRELIMINARY OBSERVATIONS OF LIGHT SCATTERING FROM THE $^3\text{He}-^4\text{He}$ MIXTURE NEAR ITS CONSOLUTE CRITICAL POINT

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Résumé. — Nous présentons les résultats préliminaires sur l'opalescence critique que nous avons observée dans les mélanges $^3\text{He}-^4\text{He}$ au voisinage du sommet de la courbe de coexistence.

Abstract. — Critical opalescence ce has been observed in $^3\text{He}-^4\text{He}$ mixtures in the vicinity of the top of the miscibility gap. Preliminary results are reported.

We have observed critical opalescence in a mixture of $^3\text{He}-^4\text{He}$ at equilibrium near the critical point at the top of the miscibility gap. We were motivated to study this system by the unique features of its critical point which is supposed to occur at the conjunction of two phase transitions, the superfluid transition along the lambda line and the demixing that leads to two-phase equilibrium across a miscibility gap that vanishes at the consolute temperature of about 0.87 °K [1].

If this system is analyzed by assuming the same scaling relations that are applied around conventional critical points, some remarkable properties of the critical opalescence are predicted. The critical behavior of the coexistence curve and the concentration dependence of the chemical potential difference $\mu_3^P-\mu_4^P$ are known experimentally [2], [3], [4]. The data yield the critical exponent values $\delta = 2$, $\gamma' \approx 1$ and $\beta = 1$ where the usual notation has been used [5]. The observations of the specific heat [3] also suggest $\alpha = -1$. With the help of the scaling relations [5]:

$$v = \frac{\beta(\delta + 1)}{3}$$
$$\eta = \frac{2 - 3(\delta - 1)}{(\delta + 1)}$$

or $v = \gamma(2 - \eta)$, we find $\gamma \approx 1$ and $\eta \approx 1$. According to the theories of Gor’kov and Pitaevskii [6], and Ganguly and Griffin [7], the light scattering intensity should begin to diverge near the critical temperature as the temperature $T$ approaches the critical temperature $T_\text{c}$ as $e^{-\gamma'v} = e^{-1}$ where $\varepsilon = |T - T_\text{c}|/T_\text{c}$ assuming that the scattering vector amplitude $(K)$ is sufficiently small that $(K\xi) \ll 1$, where $\xi$ is the correlation length. As $\varepsilon \to 0$ we would expect the correlation length to diverge also as $e^{-\gamma} = e^{-1}$ to suppress the finite angle scattering intensity roughly in accord with the Ornstein-Zernicke denominator $1 + (K\xi)^2$ as modified by the corrections to the two particle correlation function represented by the exponent $\gamma$ [5]. These effects are ordinarily small with $\gamma \approx 0.08$; however in this case the scaling relations gave $\gamma \approx 1$ implying a more substantial problem.

We report here measurements of the relative intensity of the light scattered at 90° in a temperature range of $\pm 0.030$ °K about the critical temperature $T \approx 0.871$ °K by a mixture of 0.671 $^3\text{He}$ mole-fraction $^4\text{He}$. The temperature was controlled to $\pm 1$ μK, as measured by a germanium resistance thermometer in the liquid. Equilibration times appear to be about 45 minutes. The accompanying figure is a composite of the data from three different runs that each span the entire temperature range, showing the scattered intensity as a function of thermometer resistance.

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The relative intensity of scattered light is shown along with a sketch of the coexistence curve and lambda line for the $^3\text{He}-^4\text{He}$ mixture. The right hand ordinate is the mole-fraction of $^3\text{He}$. The left hand ordinate is relative intensity. The abscissa is the resistance of the thermometer. This resistance varies approximately linearly with temperature in this small temperature range, i.e. within $\pm 0.003$ °K in the 0.06 °K range.
A distinct, reproducible, asymmetric, truncated or rounded peak about 20 mK wide was observed in the vicinity of the critical point. An approximate sketch of the coexistence curve and \( \lambda \) line are superimposed on the plot of the scattering data with a dashed line indicating the expected composition of the mixture and a shaded area to indicate its uncertainty. The temperature scales for the phase diagram and the scattering data are still uncertain with respect to each other by about \( \pm 0.005 \) °K. The scattering in the lower phase, well below its vanishing temperature, increases smoothly with temperature. At temperatures that are in approximate correspondence with the one phase superfluid region there is a mild decrease with temperature, and above, at a temperature that appears to be close to the \( \lambda \) temperature the intensity again begins to fall sharply. The magnitude of the scattering power at 90° scattering angle agrees within roughly a factor of two with values expected from theory [7], using best known values of the experimental parameters at temperatures \( \sim 10 \) mK away from the critical point along the coexistence curve. Further experiments are underway.

Scattered intensity changes under non-equilibrium conditions were striking. Extremely small rates of decrease in temperature (\( \lesssim 10^{-6} \) °K/s) substantially enhanced the scattering. Similarly, an increase in temperature temporarily depressed the scattering intensities.

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