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LA-UR -81-942

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SUBMITTED TO: LT-16
August 19-25, 1981
Los Angeles, CA

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UNITED STATES
DEPARTMENT OF ENERGY
CONTRACT W-7405-ENG-36

QUANTITATIVE MEASUREMENTS OF LIGHT SCATTERING IN DECOMPOSING ^3He-^4He LIQUID MIXTURES*

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Measurements are presented of the angular distribution of light scattering by decomposing ^3He-^4He mixtures. It is found that the structure factor has a scaling form $S(\kappa,t) = \kappa_m^{-3} F(\kappa/\kappa_m)$ where κ_m^{-1} is related to the size of a growing domain and $F(\kappa)$ is time independent during the early stages of phase separation.

INTRODUCTION

We have made quantitative measurements of the angular distribution of light scattering by ^3He-^4He mixtures pressure quenched into the miscibility gap near the tricritical point. An earlier study[1] in ^3He-^4He showed that laser light scattered by the decomposing mixture is in the form of a collapsing ring similar to that observed in organic binary mixtures[2,3]. Recent Monte Carlo studies of Marro, Lebowitz, and Kalos[4] (MLK) have shown that the (normalized) structure factor $S(\kappa,t)$ exhibits a scaling behavior in the homogeneous form

$$S(\kappa,t) = \kappa_m^{-3}(t)\, F(\kappa/\kappa_m(t)) \qquad (1)$$

where $\kappa_m^{-1}(t)$ is a measure of the time dependent domain size, t is reduced time, and $F(\kappa/\kappa_m)$ is independent of time. Experiments in isobutyric acid water (IBW) and ^3He-^4He mixtures[5] have confirmed validity of the scaling[1]. In this paper we show that a similar scaling form is valid for ^3He-^4He liquid mixtures. We also discuss some of the characteristic similarities and differences in the angular distribution of light scattering in ^3He-^4He and organic binary mixtures.

EXPERIMENTAL

The experiments were carried out in a ^3He crystal with optical windows for optical access. Pressurization was accomplished by using the bellows that forced the bottom of the sample space. In this way the concentration of the mixture was not affected by the pressurization and decompression phases. A single five transducer with a resolution of 10^{-3} was used to monitor the rapid decompression of the mixture. Germanium thermometers located near the top and the bottom (inside the sample space) monitored the initial temperature and the slight temperature rise that accompanied decompression and phase separation.

The angular distribution of the scattered laser light (λ = 6328 Å) was determined using a Reticon photodiode array of 512 elements, the signals from which were stored in a transient recorder. Due to the large number of elements in the photodiode array, a continuous distribution of intensity could be easily obtained. Because each of the photodiode elements is rectangular (width = 13 μm, height = 2.5 mm), the array was masked off to expose a wedge-shaped sensing area, thus preserving good resolution at low scattering angles. The output signals were subsequently plotted, smoothed, and appropriately corrected to yield the angular distribution of scattered light intensity $I(\kappa,t)$. We have not as yet measured $I(0,t)$ and thus cannot correct the data for effects of multiple scattering. However, because of the small scattering intensities, we expect these effects to be small, even for our relatively low [illegible] (1.[illegible] cm).

RESULTS AND DISCUSSION

We now discuss the light scattering measurement made during the early stages of phase separation of an initially homogeneous [illegible] fluid mixture at a [illegible] = 0.[illegible] K and a [illegible] 0.918 K and a [illegible] pressure of 5.[illegible] atm. Forward light scattering was observed until the pressure in the cell dropped to the [illegible] atm when a bright and hazy defined ring of light suddenly appeared [illegible] visually observed on a screen[1]. This is consistent with the fact that light scattering is expected [illegible] reduced after the spinodal boundary is crossed. After its sudden appearance, the ring [illegible] collapsed to the central beam within several minutes, which is about [illegible] times faster than that is observed in organic binary mixtures. The intensity distribution of the [illegible] scattered light recorded by the photodiode array reveals a [illegible] which progressively increase in intensity before the [illegible] the ring gradually collapses to the central laser beam. It is interesting to point out that the time evolution of the angular distribution of scattered light in ^3He-^4He has a striking resemblance to results from a Monte Carlo study of tricritical spinodal decomposition in a [illegible]

metamagnet[5], where, for the case of the conserved order parameter, it is seen that the intensity distributions are more sharply peaked than those observed in organic binary mixtures.

In the cases of I-W and L-W, the maximum intensity I_m in the scattered light increased by almost three orders of magnitude before the ring finally collapsed to the central beam. In ^3He-^4He, the corresponding increase in I_m is only about a factor of 30. The wave vector κ_m ($\kappa = [4\pi/\lambda]\sin\theta/2$, where θ is the scattering angle) corresponding to I_m was found to decrease in a similar manner to that observed in I-W and L-W, i.e., $\kappa_m \propto t^{-\phi}$, where ϕ gradually changes from ~1/3 at early times to ~1 at later times.

In Fig. 1 we have plotted the results of our measurements in terms of $\kappa_m^3\, I(\kappa,t)$ as a function of κ/κ_m for various times t. Whereas MLF found that Eq. (1) was best satisfied when K was taken as the first moment, k_1, of the structure factor, we follow Ref. 7 and identify this parameter with κ_m. Note the sharpness of the peaks in the scaled curves (Fig. 1). The overall shape of the curves is quite similar to that presented by MLF (see Fig. 2 of Ref. 4). It is obvious from Fig. 1 that the function $F(\kappa/\kappa_m)$ is reasonably independent of time, at least for values of $\kappa/\kappa_m \geq 1$. To the left of the peak in Fig. 1, the data show a slight trend with time, suggesting that at the earliest times scaling does not hold, [illegible]

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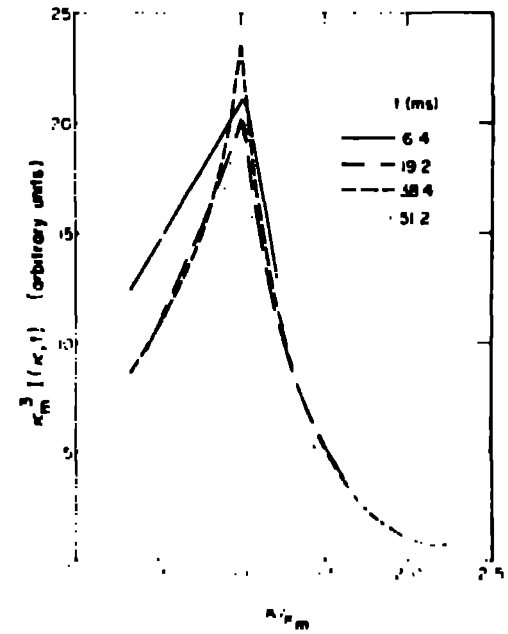

Fig. 1. Smoothed intensities of scattered laser light, scaled by κ_m^3, vs κ/κ_m for a mixture of liquid ^3He-^4He of composition X = 0.672 and initial temperature 0.816 K, following a pressure quench [illegible]. With the [illegible]

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*Supported in part by the U.S. Department of Energy and by the National Aeronautics and Space Administration Working Group for Physics and Chemistry Experiments (NASA PACE).