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Anisotropic decomposition of ^{3}He-^{4}He mixtures

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The late stage of phase separation in ^{3}He-^{4}He mixtures, quenched into the miscibility gap below the tricritical point, has been studied by light scattering. The scaled structure factor $F(k/k_m)$, which in general exhibits a universal shape, becomes nonuniversal as a pronounced vertical-horizontal asymmetry of the scattered intensity appears. This result indicates the influence of gravity on the decomposition process at that stage.

In the phase diagram of most binary mixtures a miscibility gap is observed, i.e., a region where the homogeneous mixture cannot exist in thermodynamic equilibrium. From an unstable state within the miscibility gap, reached by a rapid quench in temperature or pressure, the mixture decomposes into two stable phases located on the edges of this region. Several mechanisms dominating during different stages of the decomposition process have been considered[1,2]: at an early stage "hill-up" diffusion (for spinodal decomposition) or nucleation, subsequently cluster growth, hydrodynamic processes, and separation by gravity. Experimentally, the structure factor $S(k,t)$ of decomposing liquid binary mixtures has been studied by means of light scattering.[3,4] The time dependence of the structure factor maximum at a wave vector k_m, resulting from the sequence of the various decomposition mechanisms, has been measured and found to be in quite good agreement with theoretical predictions. The effect of gravity, though, on the experimental results has not been taken into account so far.

The experiment reported here was carried out to investigate the stage of coarse graining and cluster growth in decomposing ^{3}He-^{4}He mixtures below the tricritical point. This system, which is extremely clean and well defined, is described by *two* order parameters, the concentration difference of the two coexisting phases and, in addition, the superfluid density. This fact leads to the pronounced asymmetry and the angular top of the coexistence curve (see phase diagram in the insert of Fig. 2), in contrast to the ordinary critical systems investigated earlier.[3,4]

The quenched states were prepared by decompression of the ^{3}He-^{4}He sample, as described previously.[5,6] The ensuing phase separation process was observed by means of the scattering of light from a He-Ne laser ($\lambda = 632.8$ nm), with an optical scanner serving to resolve the angular distribution of the scattered intensity. The quenches, in general, started from a state on the superfluid branch of the coexistence curve, so that the equilibrium concentration was determined by the initial sample pressure and temperature. The time domain we have studied here is $t \geq 0.5$ sec after starting decompression, from which we estimate for the reduced time $\tau = D\xi^{-2}t$ values of $\tau > 10^6$ (D = diffusion coefficient, ξ = correlation length). Besides, the range of the wave vectors k_m in our measurements, as shown below, is $k_m \leq 10^4$ cm^{-1}, which in reduced units yields $k_m\xi < 10^{-3}$.[7] These two estimates strongly suggest that our data were taken in the region of late decompositional stages and in the hydrodynamic limit where distances between clusters are much greater than the interfacial thickness.

When the sample was quenched, intense light scattering was observed, mostly concentrated in a narrow, contracting halo. As already noted by Hoffer *et al.*[5,8] the normalized structure factor[1] $\tilde{S}(k,t)$ of ^{3}He-^{4}He at this stage, derived from the peaked intensity distribution, can be well represented by a universal function $F(k/k_m)$ in the scaling form $\tilde{S}(k,t) = k_m^{-3}(t)F(k/k_m)$. Both the appearance of a halo and the scaling behavior are in good agreement with results from decomposition experiments in the critical region of *ordinary* binary mixtures like the isobutyric acid-water and lutidine-water systems.[3,4]

A careful analysis of our scattering data showed, however, that the scaling function F is not really time independent, but changes during the late stages of the decomposition process, thus violating the universality discussed above. Moreover, $F(k/k_m)$ apparently becomes *anisotropic* around the direction of the primary laser beam, since the angular distribution of the scattered intensity and with it the structure factor $S(k)$ are found to depend on the azimuthal scattering angle.

As an example, Fig. 1 shows a photograph of the scattered light which demonstrates this anisotropy. The halo obviously no longer has circular symmetry as at earlier stages of decomposition, but is sharper in the horizontal as compared to the vertical direction. In a more quantitative way the anisotropy appears in Fig. 2, where structure factors $S(k)$ derived from simultaneous scans in horizontal and vertical direc-

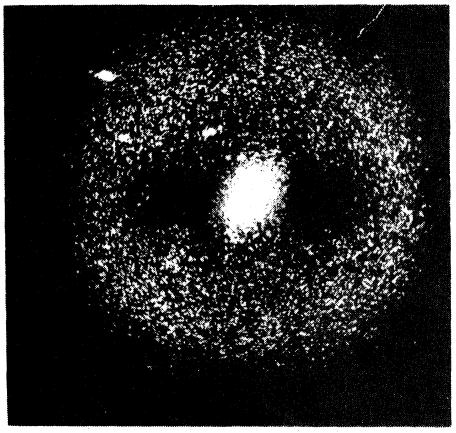


FIG. 1. Far-field scattering pattern from a ^{3}He-^{4}He mixture undergoing phase separation. The mixture was adiabatically quenched from a state on the superfluid coexistence curve at $T=0.847$ K, $p=0.422$ bar to a pressure of 0.050 bar. The pattern was recorded at a time $t=2.0$ sec after starting decompression (which is long compared to the pressure relaxation time of 0.2 sec). The central spot is due to the unscattered laser beam. The outer diameter of the halo corresponds to a wave vector of $k=7.6\times10^3$ cm^{-1}.

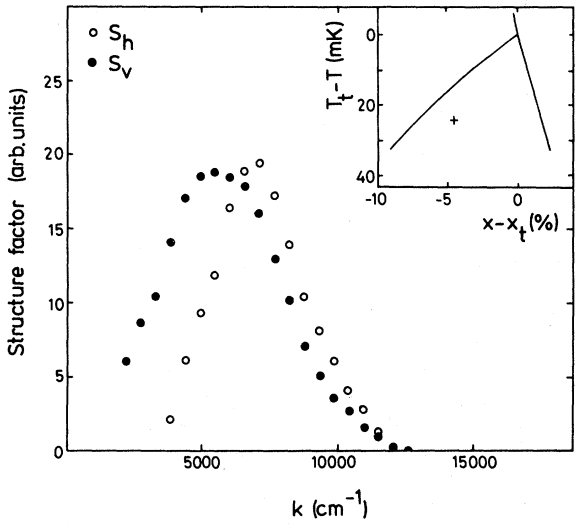


FIG. 2. Structure factors S_h and S_v, determined from the scattered intensity in the horizontal and the vertical scattering plane, respectively, at $t=1.3$ sec after starting decompression from $T=0.830$ K and $p=0.724$ to 0.310 bar. The quenched state is indicated in the insert (+). The structure factor is directly represented by the scattered intensity because multiple scattering effects, estimated from the transmissivity of the sample, are negligible at this stage. The error bars are smaller than the symbol size.

tions are plotted. Clearly the structure factor S_v from the vertical scan displays a wider peak than S_h for horizontal scattering. This behavior is observed not only for the quenched state shown in the insert of Fig. 2, but for a wide range in the central region of the miscibility gap.

A characteristic quantity for the description of the spatial growth of the new phases is the wave vector k_m, where the maximum of the scattered intensity appears. In a crude picture k_m represents some average distance between clusters of the minority phase. (Note, however, that for our observations these clusters will be connected since the volume fraction of the developing ^{3}He-rich minority phase lies well above the percolation limit of about 0.16.) In the process of coarse graining k_m decreases; the detailed dependence of k_m on time, like $k_m \propto t^{-1/3}$ or $k_m \propto t^{-1}$, has been used in the earlier experiments to distinguish between the various decomposition mechanisms mentioned above.[3,4] For the ^{3}He-^{4}He mixture investigated here, the development of k_m is shown in Fig. 3, again for horizontal and vertical optical scans. At early times scattering is isotropic and hence the values of $k_{m,h}$ and $k_{m,v}$ agree, but as decomposition proceeds $k_{m,v}$ becomes distinctly smaller than $k_{m,h}$, as suggested already by a comparison of S_h and S_v in Fig. 2. Apparently because of the anisotropy a unique dependence of k_m on time cannot be derived from Fig. 3.

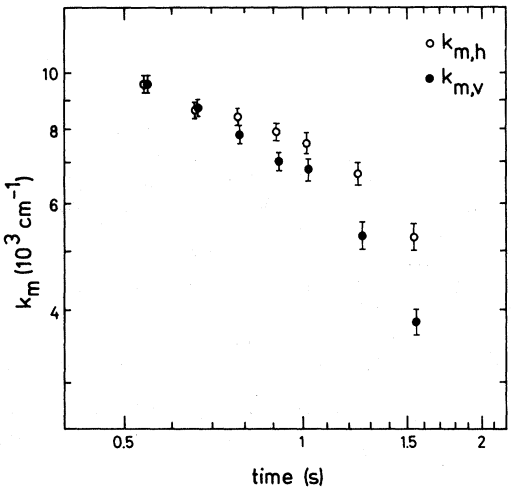
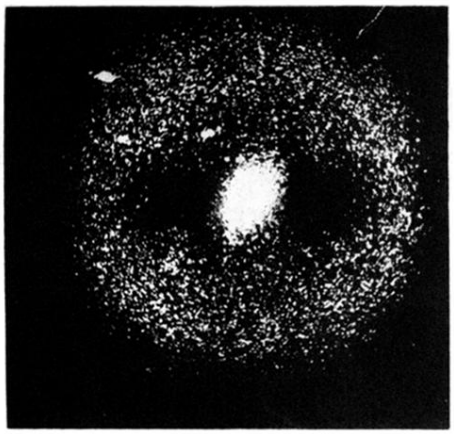


FIG. 3. Positions of the maximum intensity, $k_{m,h}$ and $k_{m,v}$, measured in the horizontal and the vertical scattering plane, for the same quench conditions as in Fig. 2. Because of the anisotropic scattering a unique time dependence of k_m which could be compared with previously proposed models for decomposition, cannot be derived.



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