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DECOMPOSITION OF ^{3}HE-^{4}HE MIXTURES NEAR THE TRICRITICAL POINT

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Light scattering measurements on decomposing ^{3}He-^{4}He mixtures are reported for temperatures near the tricritical point. The results deviate from those obtained for common critical systems. This may indicate that for ^{3}He-^{4}He a different separation mechanism is relevant.

I. INTRODUCTION

A binary mixture decomposes spontaneously when quenched into the domain of instability in the phase separation regime. This process of "spinodal decomposition" has been studied in detail for mixtures with an ordinary critical point. In the spinodal region concentration fluctuations should grow preferentially at a particular wave vector k_m, as predicted by the linear theory of Cahn/1/ for the initial stages of decomposition; extended theories have taken into account non-linear terms and hydrodynamic and gravitational effects, leading to a coarse graining of the fluctuations at later stages, in quite good agreement with experimental observations in organic mixtures/2-4/.

We have investigated the decomposition of ^{3}He-^{4}He mixtures, which in the vicinity of the tricritical point are described by two order parameters and a set of critical exponents completely different from those at ordinary critical points, from which consequences for the decomposition process might be expected. Experimental studies of the same system have been reported earlier by Hoffer et al./5/; theoretical predictions for ^{3}He-^{4}He sofar are restricted to a linear theory by Hohenberg and Nelson/6/.

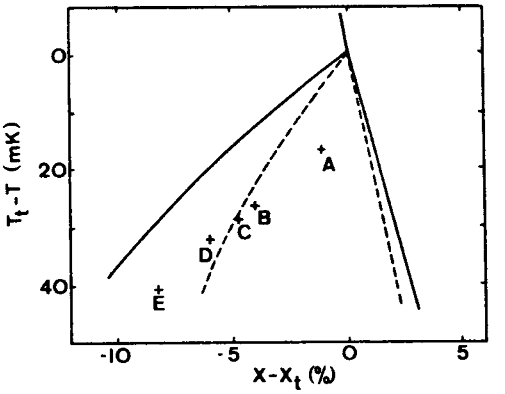


Fig.1: Solid Lines: Phase diagram of ^{3}He-^{4}He mixtures near T_t(=0.860K at P=0.2 bar). The symbols A to E denote states prepared by quenching a superfluid ^{3}He-^{4}He mixture from the coexistence curve at P_i=0.7 bar to a final pressure P_f=0.2 bar. In each of the quenches A to E a halo was observed. The central area between the dashed lines is the spinodal regime after Hohenberg and Nelson/6/.

II. EXPERIMENTAL

The sample, 1 cm³ of a tricritical ^{3}He-^{4}He mixture, was quenched into the instability regime by a pressure jump technique/4,5/: After an equilibrium state in the one phase region or on the coexistence curve had been prepared at an initial pressure P_i, this pressure was quickly released through the supply capillary to a final value P_f, leaving the system in a state inside the miscibility gap. The pressure relaxation, measured directly in the cell, took ∿0.2 sec for a typical quench from P_i=0.7 to P_f=0.2 bar. Each state prepared by such a quench (five of them shown in Fig.1) could be well localized in the phase diagram knowing the initial temperature, the pressure dependence of the phase diagram and the temperature rise during the adiabatic decompression.

In the decomposition process following the quench the intensity of light (λ=632.8 nm) scattered from the developing concentration fluctuations was recorded at different angles with a set of photomultipliers; in some instances the scattering pattern was also photographed.

II. RESULTS

A phenomenon which is generally considered as a hallmark of spinodal decomposition was observed also in ^{3}He-^{4}He mixtures near the tricritical point: For a wide range of quenches into the miscibility gap a halo in the scattered light intensity appeared[1], which collapsed to a disc at a rate depending on the quench conditions, a typical time scale being 1 sec.[2] As indicated in Fig.1, halos appear not only inside, but also well beyond the spinodal regime estimated by Hohenberg and Nelson/6/. On the other hand the timescale of our measurements, expressed in units of $\tau = D\xi^{-2}t$ (where D is the diffusion coefficient and ξ the correlation length/3,7/) corresponds to $\tau \geq 10^6$, which for the ordinary binary mixtures is the region of late decompositional stages. Therefore it is not surprising that the linear Hohenberg-Nelson theory, developed for the very early stages of tricritical decomposition, is not applicable here.

The existence of a contracting halo, reflecting the coarse graining of dominant concentration fluctuations, at first glance appears to be in accord with the experiments on spinodal decomposition in ordinary mixtures. A more detailed analysis shows, however, that pronounced deviations from the usual behavior occur in the ^{3}He-^{4}He system near T_t:

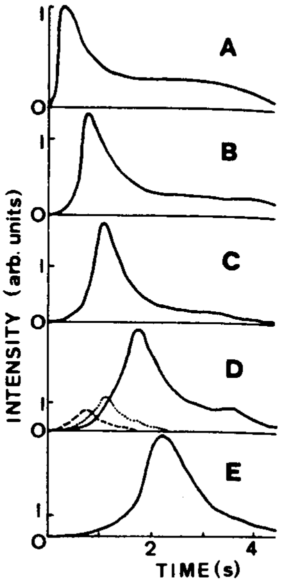
i) For "off-critical" quenches - where the proportion of the coexisting phases during decomposition deviates distinctly from unity (i.e. for most of the quenches marked in Fig.1) - a universal description is usually obtained using scaled variables of reduced time $\tau = D\xi^{-2}t$ and wave vector $q = k_m\xi$ (k_m is the scattering vector characterizing the maximum of the halo intensity)/4/. If this type of scaling were obeyed in ^{3}He-^{4}He, one would expect a drastic critical slowing down of the halo contraction proportional to $(T_t-T)^{-3}$ as the tricritical point is approached[3]. The experiment, as shown in Fig.2, reveals just the opposite, namely a speeding up as one proceeds towards T_t from quench E to A.

ii) The variation of k_m with time, as already noticed by Hoffer et al./5/, was found to be given by $k_m \propto t^{-1}$ during the later stages of halo contraction, whereas in the other critical systems always $k_m \propto t^{-1/3}$ was obtained for off-critical quenches/4/.

iii) A close inspection of the scattering pattern shows that the halo is not completely symmetric around the primary beam; rather, in the horizontal scattering plane the peak at k_m appears to be sharper than in the vertical direction.

This last observation leads us to the assertion that for the decomposition process investigated here gravity is important - although for a coarse graining stage at $k_m \sim 10^4$ cm^{-1} this might be unexpected. (For most experiments on the ordinary binary mixtures the influence of gravity could be neglected.) If the growth of droplets of the minority phase is determined by their motion in the gravitational field, the droplet distances in horizontal and vertical direction might well be correlated differently, thus giving rise to asymmetric scattering. Also the lack of conventional scaling would not be surprising, because the growth rate in the gravitationally dominated regime should not only depend on temperature, but also on the proportion of the minority and majority phase. Indeed, quenches at different proportions and constant temperature showed that for an increasing fraction of the minority phase the contraction of the halo develops faster.

Fig.2:
Scattered light intensity (in the horizontal scattering plane) for the quenches marked in Fig.1. For clarity the intensity at several scattering angles ϕ is shown only in quench D(—ϕ=2.3°; ···4.4°; ---5.1°); the appearance of a halo can be inferred from the crossing of the curves for the various angles. The speeding up of the halo contraction on approaching the tricritical point is reflected in the shift of the intensity maxima towards earlier times as $T \to T_t$.



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1) With the decomposition starting in the temperature range $T_t-T>50$ mK no halos, but only contracting disc-like patterns were seen from the very beginning of the separation process.

2) Halos have been observed by Hoffer et al./6/, however, in contrast to here, not for quenches from the superfluid phase. This difference may be due to much smaller quench depths P_i-P_f in our experiment.

3) Although the power laws for D and ξ given in /7/ are not likely to hold for $(T_t-T) \geq 10$ mK, the argument should qualitatively remain correct.

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