Second Sound in Metastable and Phase-Separated ^{3}He-^{4}He Mixtures

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Propagation of second sound has been used to study superfluid ^{3}He-^{4}He mixtures quenched into nonequilibrium states inside the miscibility gap. From the results for the second-sound velocity we conclude that the superfluid density in the metastable state is well described by extrapolation from equilibrium values. The attenuation of the second-sound amplitude displays a sharp increase as nucleation sets in at the cloud point, and during the subsequent coarse-graining process it varies in proportion to the total surface area of the droplets.

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Phase separation of a supercooled gas or a quenched binary mixture is a common, yet rather complex phenomenon. A detailed theoretical treatment is rendered difficult by the fact that several interrelated processes become important at the various stages of the decomposition process.[1] Experimentally, a number of studies of phase-separating binary liquid mixtures have been carried out in recent years, using, in particular, light scattering as an efficient technique to probe the development of nucleating droplets up to the completion of phase separation.[2] In nearly all of these experiments the physical quantity which has been exclusively studied is the concentration, which plays the role of an order parameter. The question arises as to how other characteristic quantities are influenced when the system is quenched from equilibrium via a metastable region to an unstable state. Although it appears reasonable to extrapolate known data from the equilibrium regime of the phase diagram into the miscibility gap (i.e., the nonequilibrium region between the two branches of the coexistence curve), experimental tests to verify such an assumption hardly exist. Here we report results for phase-separating ^{3}He-^{4}He mixtures, which provide a unique system to investigate this question, since not only are these mixtures very pure and well defined, but they are, in addition, characterized in the ^{4}He-rich superfluid phase by a second-order parameter, the superfluid density. The behavior of this quantity in a nonequilibrium state, studied by means of second-sound propagation, is the subject of this Letter.

In order to prepare the mixture in a proper state inside the miscibility gap, we used the pressure-quench technique with an experimental setup as described earlier.[3,4] A schematic path in the phase diagram plotted on a reduced temperature scale is shown in the inset of Fig. 1. The temperature of the tricritical point T_t is 0.867 K at saturated vapor pressure and decreases with pressure according to $\partial T_t/\partial p = -0.033$ K/bar. The quench starts on the superfluid branch of the coexistence curve (A), and the system remains in a homogeneous, metastable state (B), until nucleation rapidly sets in as the boundary of metastability, the so-called cloud point, is reached. Local decomposition into ^{3}He-rich droplets (C^+) in a superfluid ^{4}He-rich background phase (C^-) is then essentially completed within a few milliseconds. Subsequently, the system "slides down" along the coexistence

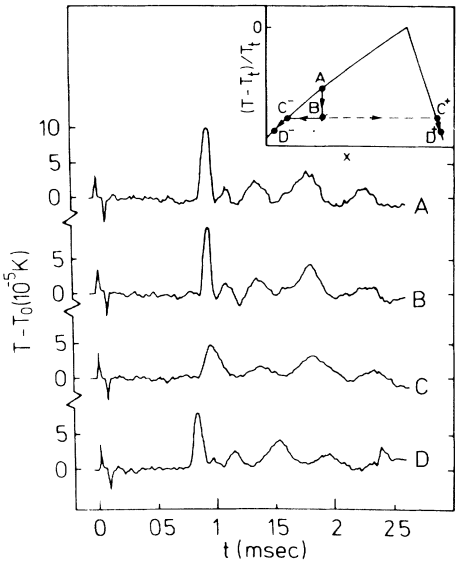

FIG. 1. Second-sound signal in a superfluid ^{3}He-^{4}He mixture at various stages of the decomposition process. The traces, taken in subsequent identical quenches, refer to the following conditions, also marked in the schematic phase diagram shown in the inset (the dotted curve symbolizes the cloud line): A, in the homogeneous superfluid phase on the coexistence curve, 23 mK below the tricritical temperature T_t, at $p = 880$ mbar; B, in the metastable regime just before reaching the cloud point where nucleation sets in; C, after nucleation, when the system has undergone decomposition on a local scale, consisting of ^{3}He-rich normal droplets (^{3}He concentration $x^+ = 0.697$) in a ^{4}He-rich superfluid background ($x^- = 0.602$); D, at the end of the decomposition process, when the droplets have accumulated in the upper phase, and the lower, superfluid phase is homogeneous again ($T_t - T = 35$ mK, $p = 510$ mbar).

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curve until the pressure quench is terminated at points D^+ and D^-. There the late stages of decomposition, coarsening and macroscopic phase separation by gravity, are also completed. The typical time scale for the whole phase-separation process is a few seconds.

Second-sound pulses (duration $t_{II}=0.1$ ms, energy 2 μJ) were generated by heating a nickel film, and detected with a carbon bolometer located on the opposite side of the sample cell at a distance of 0.66 cm. In addition, the decomposition process was monitored optically by measuring the attenuation of a laser beam transmitted through the cell. This method allows a good characterization of a mixture undergoing phase separation.[3]

Figure 1 shows examples of second-sound signals for the various conditions (A-D) described above. The traces display a sharp rise about 1 ms after the application of the heat pulse, corresponding to the transit time of the second-sound pulse. It is obvious that well-defined second-sound signals are obtained not only in the superfluid phase on the coexistence curve (traces A and D), but also in the metastable state B and in the heterogeneous mixture consisting of normal droplets (average diameter in this case 1.0 μm) in a superfluid background phase (C).

A comparison shows that the time of flight changes only slightly during the first stages. The same is then true for the superfluid density ρ_s, since the second-sound velocity v_{II} is related to ρ_s by $v_{II}^2=(\rho_s/\rho_n)f(x,T,S)$.[5] (Here ρ_n is the normal fluid density and f is a function of the concentration, temperature, and entropy of the mixture.) In contrast to the velocity, the amplitude of the second-sound pulse changes significantly in the heterogeneous mixture. This becomes even more obvious from Fig. 2, where we have plotted results for the velocity v_{II} and the amplitude A_{II} of the second-sound pulse for a large number of identical quenches, measured at various times t after the beginning of each respective quench. The bottom trace in Fig. 2 represents the optical transmissivity of the sample and is used to identify the state of the mixture: In the metastable regime the transmissivity is constant and within our resolution equivalent to the equilibrium value. The onset of nucleation is reflected in a sharp drop of the transmitted light intensity, with the subsequent slow decrease and eventual recovery of the signal being a signature of the later stages and completion of the phase-separation process. From these transmissivity data, together with angularly resolved light scattering, the number density and average diameter of the droplets in a heterogeneous mixture can be determined.[3,6,7]

We now turn to the second-sound results and consider first the *metastable regime*. In this case the heat pulse propagates in a homogeneous medium. The velocity v_{II} in this range is constant within 1% along the thermodynamic path given by the adiabatic quench, as displayed in Fig. 2(a) and, more clearly, on an expanded scale in Fig. 3(a) for another run at a different temperature. The results agree very well with an extrapolation of data in the vicinity of, but outside, the miscibility gap,[8] from which one expects the second-sound velocity to remain constant within 1% up to the largest supersaturation $\delta x/\Delta x\simeq 0.15$ reached under the present conditions. (Here $\Delta x=x^+-x^-$ is the width of the miscibility gap and δx is the deviation from the equilibrium concentration on the superfluid branch of the coexistence curve.) Our data therefore demonstrate that not only does the superfluid density exist in a superfluid mixture, but that, moreover, it closely follows the behavior extrapolated from equilibrium conditions, as already anticipated in earlier theoretical work by Hohenberg and Nelson.[9]

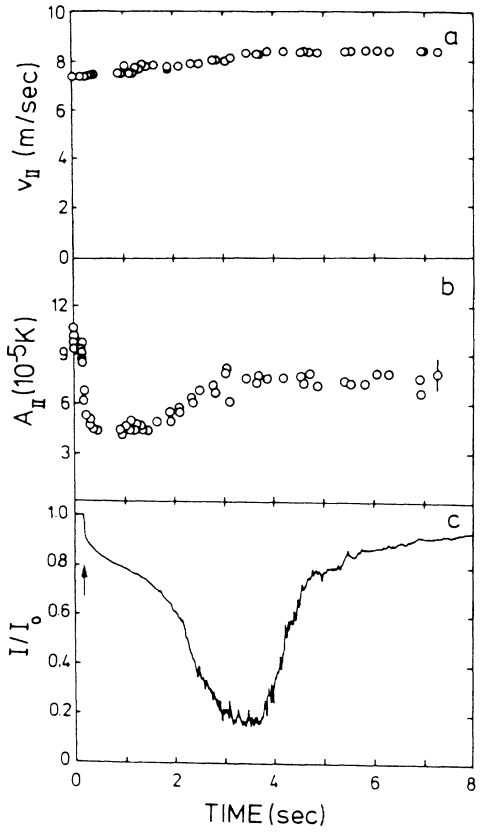

FIG. 2. (a) Velocity v_{II} and (b) amplitude A_{II} of second-sound pulses propagating through a ^{3}He-^{4}He mixture at various times after the start of the quench. The quench conditions were the same as in Fig. 1. For comparison, trace (c) shows the optical transmissivity of the sample, I/I_0, at a wavelength $\lambda=633$ nm. The onset of nucleation is marked by an arrow. The pressure quench is essentially complete after 1 s, so that the variation of the signals at the later times is due to droplet growth at nearly constant pressure.

One might ask whether the metastability of the system, although it apparently does not influence the absolute value of ρ_s, could give rise to an increased damping of second sound due to a possible enhancement of concentration fluctuations. However, the data for the

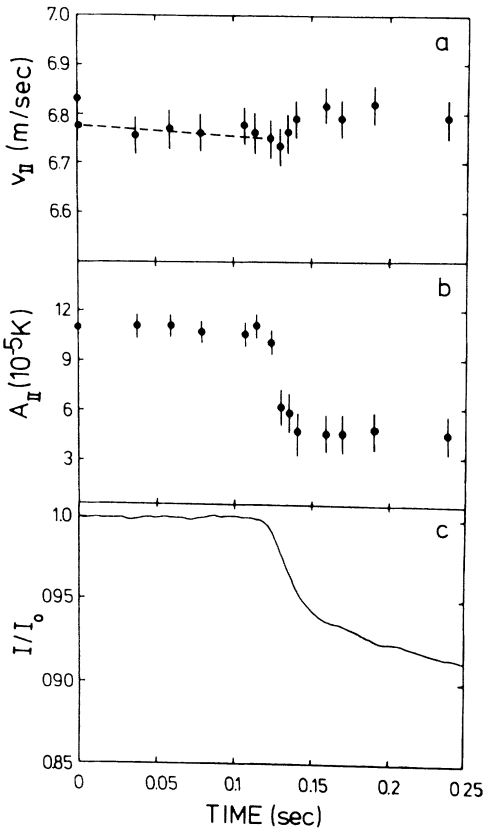

FIG. 3. Velocity v_{II} and amplitude A_{II} of second sound, and optical transmissivity in the vicinity of the cloud point. The quench parameters were the same as in Fig. 1, except for the initial and final temperatures on the coexistence curve, which were $T_i - T = 17$ and 29 mK, respectively. The dashed line in (a) shows the behavior of the velocity expected from an extrapolation of data outside the miscibility gap into the metastable state.

second-sound amplitude plotted in Figs. 2(b) and 3(b) show no signature of an anomalous attenuation up to the onset of nucleation. Thus, with respect to superfluidity, the metastable regime of the mixture does not differ from the stable equilibrium states.

A completely new situation is encountered in the heterogeneous mixture *after the onset of nucleation.* As shown earlier,[3] local decomposition develops in ^{3}He-^{4}He on a scale of only a few msec [indicated by the width of the step in the extinction signals of Figs. 2(c) and 3(c)]. Subsequently, the concentrations of droplets and background phase are close to the equilibrium concentrations on the two branches of the coexistence curve at the respective pressure and temperature. The propagation of a heat pulse is in this case no longer adequately described by ordinary second sound, but has to be modified due to the presence of the nonsuperfluid droplets. The situation resembles sound in a porous medium, however with somewhat different boundary conditions at the droplet interfaces.

To our knowledge a theory for the propagation of entropy waves for such a heterogeneous system does not yet exist. For a rough estimate we have applied a relation for the velocity of first sound in a porous medium[10]

$$v = P^{1/4} v_0 . \quad (1)$$

(Here P is the volume fraction of the pores and v_0 is the sound velocity in the bulk material without pores.) In our case, we set $P = 1 - V$, where V is the volume fraction of the minority phase. A comparison with the data yields qualitative, but not really good agreement: at $t = 0.2$ s in Fig. 3, for example, the volume fraction as inferred from optical extinction data and the phase diagram is 20%, and the second-sound velocity in the homogeneous mixture at the given pressure and temperature on the coexistence curve is 7.6 m/s.[8] According to Eq. (1) one therefore expects for the heterogeneous mixture at $t = 0.2$ s a velocity of 7.2 m/s, whereas the measured value is 6.8 m/s. Towards the end of the decomposition process the nonsuperfluid volume fraction vanishes due to macroscopic phase separation, and the second-sound velocity for the homogeneous system, as given by Ahlers,[8] is recovered.

As already mentioned, the amplitude of the heat pulse is much more strongly affected by the presence of the nonsuperfluid droplets than the velocity. At first sight one might simply attribute this to additional attenuation due to scattering or absorption of the pulse by the droplets. A more careful analysis reveals, however, that although the pulse amplitude drops, its width simultaneously increases, so that the total area of the pulse remains constant during the entire coarse-graining process up to the completion of phase separation.

We have considered two possible causes for the pulse broadening: (i) A spreading of the time of flight can occur due to different paths through the droplet configuration. The contribution of this effect should be small, however, because the dominant Fourier components of the heat pulse have wavelengths around 0.1 mm, a length scale on which the mixture essentially appears homogeneous (the average size and distance of the droplets is 2 orders of magnitude smaller). (ii) A broadening also results when part of the pulse energy is coupled to the droplet system, which due to the involved time constant gives rise to a reduction in the slope of the leading edge of the pulse, as is apparent in Fig. 1, trace C; the same is true for the flow of energy from the droplets back to the superfluid phase at the trailing edge of the pulse. The measured decrease in the pulse height turns out to be directly proportional to the surface area of the droplets, as shown in Fig. 4. The quantity $\Delta\alpha_{II}$ is defined as $\Delta\alpha_{II} = \alpha_{II}(t) - \alpha_{II,hom} = -\ln[A(t)/A_{hom}]/L$, where $A(t)$ and A_{hom} are the pulse amplitude at time t and at the end of the phase-separation process, respectively, and L is the thickness of the sample. The data are in close agreement with the solid line, which represents the droplets' surface area as determined from light-

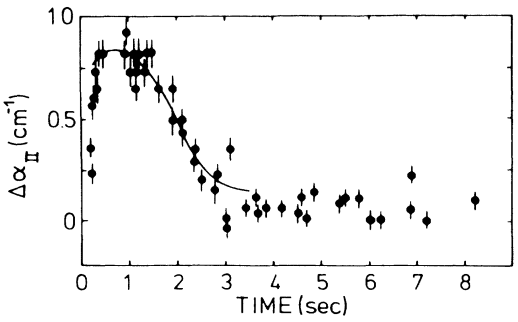

FIG. 4. Attenuation of the second-sound amplitude in the decomposing ^{3}He-^{4}He mixture of Fig. 2. $\Delta\alpha_{II}=\alpha_{II}(t)-\alpha_{II,hom}$ is the difference of the attenuation coefficients of the heterogeneous mixture, $\alpha_{II}(t)$, and of the homogeneous mixture at the end of the decomposition process, $\alpha_{II,hom}$. The solid line represents a dependence $\Delta\alpha_{II}\sim S_{tot}$, where S_{tot} is the total surface area of the droplets per unit volume, as determined from light-scattering data.

scattering data. This suggests that only a surface layer of the droplets is involved in the process which reduces the pulse amplitude.

In summary, the propagation of second sound in a superfluid ^{3}He-^{4}He mixture has been used as a novel method to gain information about nonequilibrium states. Our data for the velocity of the second-sound pulse have shown for the first time that the superfluid density in the metastable range inside the miscibility gap follows the behavior predicted by an extrapolation from the stable region of the phase diagram. The attenuation of the second-sound amplitude, on the other hand, clearly reflects the nucleation and phase-separation process. For a more-detailed analysis of the latter phenomenon a theoretical description for the propagation of a heat pulse in a superfluid phase with finely dispersed non-superfluid droplets is highly desirable.

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