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PHASE SEPARATION AND THE SUPERFLUID TRANSITION IN LIQUID He^3-He^4 MIXTURES[*]

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The phase-separation and superfluid-transition temperatures of He^3-He^4 liquid mixtures have been investigated as functions of He^3 concentration under conditions of saturated vapor pressure by means of dielectric-constant and thermal-equilibrium time-constant measurements. The λ line, which divides the normal and superfluid phases in the temperature-concentration plane, is found to intersect the phase-separation curve at its peak.

Phase separation and the onset of superfluidity are two phase transitions known to occur in liquid He^3-He^4 mixtures. Although previous work[1] has established the general features of the phase diagram of these solutions, the detailed relationship between phase separation and superfluidity has been left unresolved.

In the present experiment, the shape of the phase-separation curve was determined by means of a dielectric-constant measurement, while the superfluid transition temperatures were obtained by observation of thermal-equilibrium time constants.

The sample chamber is shown in Fig. 1. It consisted of a copper cell of volume ~1 cc containing a small coaxial capacitor (~0.013 cc volume) at the top and bottom end. These capacitors formed part of the resonant tank circuits of two tunnel-diode oscillators, the heat-dissipating portions of which were sinked

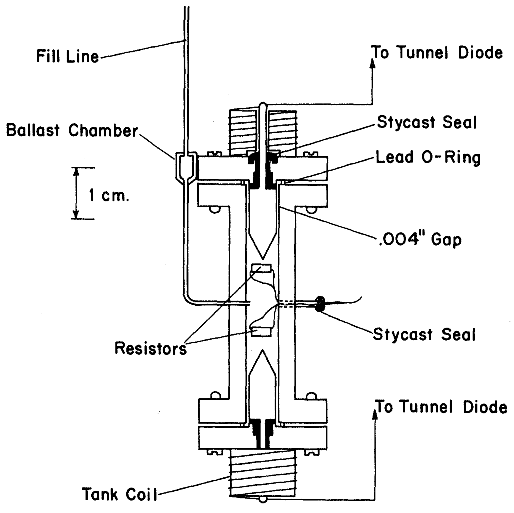

FIG. 1. Sample chamber.

to the main He^4 bath. The resonant frequency of each oscillator was ~13 MHz, with a short-term stability of ~5 Hz.

Two resistance thermometers were placed in the sample chamber. They were calibrated against the T_{62} He^3 vapor-pressure scale of Roberts, Sherman, and Sydoriak.[2] A ballast chamber of ~0.05-cc volume was used to provide an easily controlled liquid-vapor interface slightly above the interior of the sample cell.

The sample was cooled below 1.1°K by means of a conventional He^3 refrigerator. The temperature was controlled to allow warming and cooling rates of several millidegrees Kelvin per hour. These slow rates were employed to insure temperature and concentration equilibrium in the sample. Nonequilibrium effects were observed when the warming rate was increased to several millidegrees per minute.

During a typical run, the oscillator frequencies were monitored by external frequency counters. As the sample chamber was filled by condensation, a large drop in frequency (~200 kHz) was observed. This served to calibrate the relative sensitivities of the capacitors. Absolute concentration sensitivity was determined by applying the Clausius-Mossotti relation to the known molar volumes of He^3-He^4 solutions,[3] assuming that the polarizabilities of He^3 and He^4 are the same. It was found that for the range of interest, the response of the oscillators was nearly linear in He^3 molar concentration, with only small corrections necessary for changes in $\partial V_m/\partial X$ with X and in V_m with T, where V_m is the molar volume of the mixture, X the molar concentration of He^3, and T the absolute temperature.

As the full cavity was cooled, the frequencies of both oscillators decreased slightly with temperature, reflecting small changes in molar density of the homogeneous mixture. When the phase separation temperature was reached, a dramatic change took place. The frequency of one oscillator varied rapidly but continuously in one direction, while the other jumped to a value corresponding to the concentration on the other branch of the phase separation curve and then varied continuously in the opposite direction. This behavior made "homing in" on the peak of the phase-separation curve very easy; the frequency jumps for a number of different mixtures were observed until the absence of a jump indicated that the critical concentration corresponding to the peak had been reached.

Phase-separation data are shown in Figs. 2(a) and 2(b). Measurements relative to the peak can be resolved to ±0.05 mole% in X and ±0.1 mdeg K in T. However, the absolute location of the peak is not known to better than ±0.5 mole% and ±3 mdeg K. Furthermore, uncertainties in frequency calibration introduce

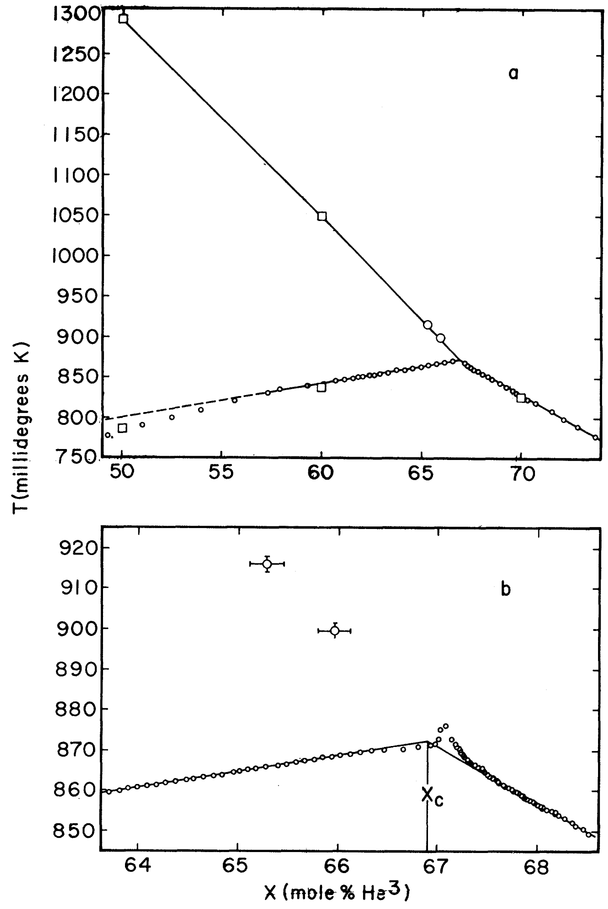

FIG. 2. (a) Phase diagram of He^3-He^4 liquid mixtures. The small circles are phase separation points obtained from dielectric-constant data. The large circles are λ temperatures from thermal-equilibrium time-constant measurements. The squares are points given by T. R. Roberts and S. G. Sydoriak, Phys. Fluids 3, 6 (1960), based on their sound-attenuation measurements, corrected to the T_{62} scale. The dashed line is an extension of the straight line determined by points near the peak. (b) Details near the peak of the He^3-He^4 liquid phase-separation curve. The small circles show the apparent He^3 concentrations in the lower and upper capacitors during the warm-up. The large circles are λ transition points. The error bars represent uncertainties relative to the peak. The absolute location of the peak of the curve is 66.9 ± 0.5 mole% He^3 and 872 ± 3 mdeg K.

possible errors of a few percent in the absolute values of the phase-separation curve slopes. The structure seen as the points leave the curve in Fig. 2(b) is also observed for concentrations farther removed from the peak, and so is thought to be associated with the phase boundary leaving the sample chamber rather than with the actual shape of the curve.

An important feature is evident in these two figures. The two branches of the phase-separation curve have a linear X-vs-T dependence as the peak is approached, with no evidence of rounding as would be expected near a normal binary-mixture critical point. It appears that the two branches intersect in a cusp.

The possibility that the λ transition might play a role in this unusual behavior led to the second phase of this experiment. The onset of superfluidity was observed for several mixtures whose He^3 concentrations were near the critical concentration by means of a thermal-equilibrium time-constant measurement. A short electrical pulse of ~10-msec duration was applied to the lower thermometer in the chamber. Such a pulse caused the sample to warm on the order of $\frac{1}{2}$ to 1 mdeg K. The thermal response of the system was measured with the upper thermometer. The time constant associated with the thermal response for a normal 65.3% mixture is plotted as a function of temperature in Fig. 3. As the temperature was lowered through the superfluid transition, a sharp drop in the response time, owing to the increased thermal conductivity, was noted. After phase separation began and the interface between the He^3-rich and He^4-rich phases fell below the upper resistor, the time constant rose again, indicating that the upper (He^3-rich) phase was not superfluid. By this method, and by observing that a mixture known to be slightly to the right of the critical concentration X_C showed no change in thermal time constant, it was concluded that within 0.2% of the peak, the He^3-rich branch does not exhibit superfluidity. Points on the λ line near the peak are shown in Figs. 2(a) and 2(b). These points are consistent with earlier sound-attenuation data of Roberts and Sydoriak[4] and indicate that the λ line intersects the peak of the phase-separation curve. Thus a second-order transition line terminates and becomes first order, still, however, separating normal fluid from superfluid in the X-T plane.

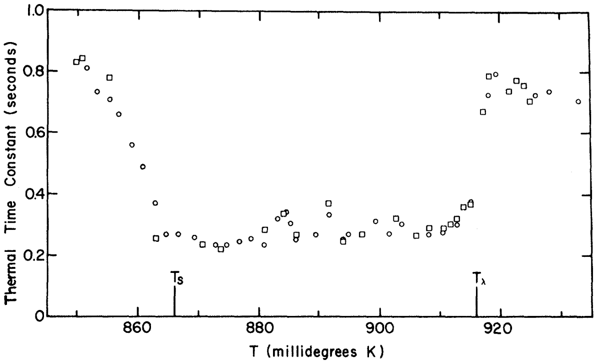

FIG. 3. Thermal-equilibrium time constant as a function of temperature for a nominal 65.3-mole% He^3 mixture. Squares are points taken during the cool down. Circles are warm-up points.

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