SOLUBILITY OF He^3 IN LIQUID He^4 AT 0°K†

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In an earlier publication[1] the problem of the equilibrium between the two phases in a phase-separated liquid He^3-He^4 mixture near 0°K has been discussed on the basis of the theory of impurities in liquid helium put forward by Landau and Pomeranchuk.[2,3] The theory gives the way in which the excitation spectrum of the liquid is modified by the presence of the impurity; e.g., a small concentration X of He^3 modifies the Landau (phonon + roton) excitation spectrum of liquid-helium II by adding a branch of Fermi particles, equal in number to the number of He^3 atoms, and with energy-momentum relation $\epsilon = -E_3(X) + p^2/2m_3^*$. The effective mass m_3^* has been determined from second sound measurements[4] to be 2.5 times the real mass of He^3 at the saturated vapor pressure. The binding energy E_3 is found[5] from vapor-pressure data to be rather close to L_3^0, the latent heat of vaporization per atom of pure liquid He^3 at 0°K, although it actually depends slightly on X. In a similar way the Fermi quasiparticle spectrum of pure liquid He^3 is modified by the addition of a Bose branch when He^4 is dissolved in it.[6] The thermodynamic properties of dilute He^3-He^4 mixtures can thus be calculated in a straightforward way using standard formulas of statistical mechanics. In particular, in a mixture in which phase separation has occurred, one can equate the chemical potentials of each isotope in the two phases to obtain the concentration in each phase as a function of T. It was found earlier[1] that the upper He^3-rich phase is almost pure He^3 for $T \leq 0.2$°K and this has been confirmed by experiment.[7] The theory also shows that it is possible, in principle, for He^3 to remain dissolved in He^4 up to a certain concentration, even at 0°K.[1] This is true for any solution in which He^3 is more strongly bound than it is in pure liquid He^3. The maximum solubility at 0°K, X_0, is then determined by equating the latent heat of vaporization of He^3 from the solution to that from the pure liquid:

$$-E_3(X_0) + kT_F(X_0) = -L_3, \qquad (1)$$

where kT_F is the Fermi energy of He^3 in solution (calculated for a gas of the same number density and with mass m_3^*). Determinations of $E_3(X)$ from vapor-pressure data are not at present sufficiently precise to demonstrate that this possibility is realized in nature.

A more sensitive method of determining $E_3(X)$ than from vapor pressures is from measurements of the phase-separation temperatures T_S of dilute solutions of known X. This has been done in the present work by measuring the heat capacity of the solutions under their saturated vapor pressure down to temperatures of about 0.1°K or less. The solutions were contained in a nylon calorimeter packed with small crystals of cerium magnesium nitrate used as a refrigerant and as a thermometer. The phase separation appears as a discontinuity ΔC in the specific heat (Fig. 1), as was previously observed at higher temperatures by Ouboter, Beenakker, and Taconis.[5] We found such discontinuities in solutions with $X = 0.08$, 0.10, 0.12, and 0.15 but not with $X = 0.04$, 0.06, or 0.07.

The failure to observe discontinuities in the specific heat for low concentrations can be interpreted in two ways: Either the phase-separation line was not crossed or the discontinuities were too small to be noticed. Since it can be shown that $\Delta C/C$ is approximately proportional to $T(dX/dT)_{T=T_S}$, the second possibility would indicate that the phase-separation line becomes steeper at low temperatures.

The results for the heat capacities (some of which are given in Fig. 1) also show that the Landau-Pomeranchuk theory is indeed applicable to these solutions. The specific heat in the single-phase region is very close to that of an ideal Fermi-Dirac gas, shown by the continuous curves above the discontinuities. (The values of T_F for the theoretical curves have been calculated with $m^* = 2.5m_3$ and with the volume of the solution taken from the data of Kerr.[8] The phonon and roton contributions to the specific heat are very small at the values of X and T considered.) However, this agreement cannot be interpreted to mean that the effective interaction between He^3 quasiparticles is zero or negligible. For a dilute Fermi gas of hard spheres, for instance, the specific heat at constant volume is the same as for the ideal, non-

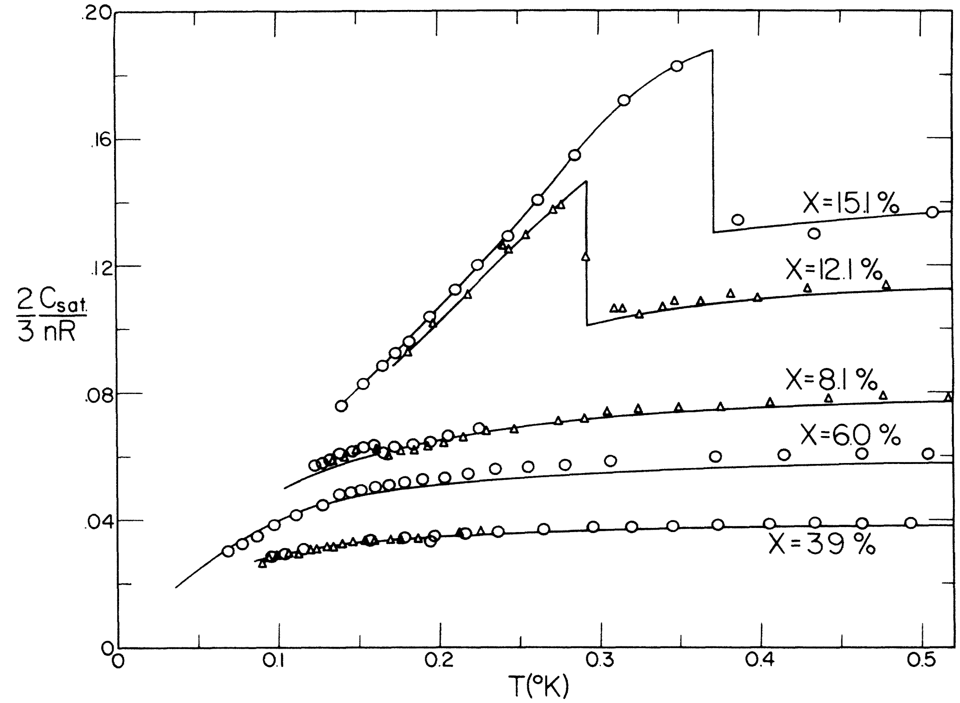

FIG. 1. The specific heat of dilute solutions of He^3 in He^4 under their own vapor pressure. The atomic concentrations X of He^3 are marked on the curves. The triangles and circles are used to distinguish between different runs. The full lines above the discontinuities represent the specific heat of an ideal Fermi-Dirac gas with effective mass $m^* = 2.5m_3$ and with the same number density as the He^3 in solution.

interacting gas, and the interaction only affects the dependence of the energy on the volume.

In Fig. 2 we have plotted our and other workers' values for T_S as a function of X, i.e., the phase-separation line. The points of Walters and Fairbank[9] have been recalculated from their nmr data with the assumption that the nuclear magnetic susceptibility of He^3 in solution is given in terms of concentration and temperature by the appropriate Fermi-Dirac expression. This fact has been verified by spin-echo measurements in our laboratory.[10]

In Fig. 2 we have plotted values of $(E_3 - L_3^0)/k$ against X, calculated from our values of $T_S(X)$, using the equality between the chemical potentials of He^3 in the upper and lower phases. Applying the quasiparticle theory for the He^3 in the lower phase and assuming the upper one is almost pure He^3, this equality can be written

$$-E_3(X) + \mu_F(T_S, X) = -L_3^0 - \int_{T=0}^{T_S} S_3^{\text{pure}}\, dT, \quad (2)$$

where μ_F is the chemical potential of an ideal Fermi-Dirac gas with $m_3^* = 2.5m_3$ and S_3^{pure}

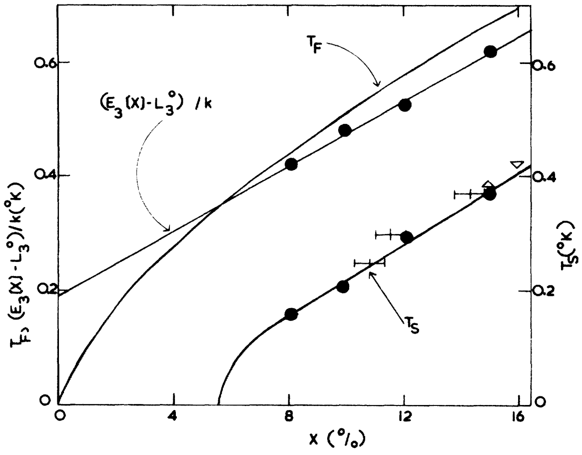

FIG. 2. Some properties of dilute solutions of He^3 in liquid He^4 as a function of X, the atomic fraction of He^3. T_S is the phase-separation temperature, T_F the Fermi temperature, and $E_3(X)$ the He^3 binding energy calculated from the present experimental values of T_S. ●, present specific-heat measurements; horizontal bars, Walters and Fairbank (see reference 9); Δ, Ouboter, Beenakker, and Taconis (see reference 5); ∇, T. R. Roberts and S. D. Sydoriak, Phys. Rev. 118, 901 (1960).

is the entropy per atom of pure He^3. As the figure shows, E_3 is a linear function of X, as was also found from vapor-pressure data,[5] and extrapolating this linear dependence gives $kT_F - E_3 = -L_3{}^0$ for $X_0 \simeq 0.06$ which, from Eq. (1), represents the limit of solubility of He^3 in He^4 at 0°K. The value of X_0 is not very sensitive to the value of $m_3{}^*$ assumed, since this affects kT_F and E_3 almost equally. Values of $E_3(X)$ and $L_3{}^0$ from vapor-pressure measurements[5] are in good agreement with the results in Fig. 2 but are less accurate. Using the linear relation between E_3 and X and Eq. (1), we have calculated the phase-separation curve which is the continuous line in Fig. 2.

It can be shown that $X_0 \neq 0$ implies that the internal energy U of the solution near 0°K should have positive curvature when plotted against X, that is, the sign of the differential heat of mixing or of $(\partial^2 U/\partial X^2)_T$ should be positive. This is in agreement with our experimental observation that, when the heating current has been switched off, the temperature continues to rise as the concentration gradients produced by the "heat flush" disappear.

It is interesting that the behavior of He^3-He^4 mixtures is qualitatively the same as the low-density mixtures of Fermi and Bose hard spheres discussed by van Leeuwen and Cohen,[11] who found that, for a sphere size and number density comparable to those for liquid helium, the fermions were soluble in the boson gas up to ~15%.

The finite solubility of He^3 in He^4 at 0°K opens the way for some interesting experiments on the properties of the He^3 (in He^4) quasiparticle gas in the semidegenerate and degenerate temperature range. For example, it should be possible to propagate zero sound in this gas at sufficiently low temperatures. The possibility of a condensation of the dissolved He^3 into a state of superfluidity or "supermobility" has been pointed out by van Leeuwen and Cohen[12] who calculated the transition temperature for a mixture of hard spheres. For $X = 0.06$ their result is unattainably small. Emery[13] has discussed the problem of real solutions and has shown that, in certain circumstances, the transition temperature might be in the accessible temperature range.

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