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Specific Heat of Dilute Solutions of ^3He in ^4He and the ^3He -Quasiparticle Excitation Spectrum

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High-precision measurements of the constant-volume specific heat of ^3He - ^4He solutions with up to 1% ^3He are presented for the temperature range $70 \text{ mK} < T < 1 \text{ K}$. The data are inconsistent with three previously proposed ^3He -quasiparticle excitation spectra: (1) a spectrum which is purely quadratic in wave number k , (2) a spectrum which is quadratic for small k and possesses a "roton" minimum at larger k , and (3) a spectrum which as a k^4 term in addition to the k^2 term at all values of k .

Thirty years ago Landau and Pomeranchuk¹ (LP) proposed that ^3He atoms in dilute solutions of ^3He in ^4He should behave as an ideal Fermi gas with a quasiparticle excitation spectrum $\hbar^2 k^2 / 2m_3^*$. Various recent experiments²⁻⁸ on ^3He - ^4He mixtures have, however, demonstrated that this energy spectrum is not completely adequate. In order to explain the experimental results, it has been suggested that the spectrum should also contain² a term proportional to k^4 or that the spec-

trum possesses⁹⁻¹¹ a rotonlike minimum. In the latter case, the spectrum in the region of the minimum is approximated by $\Delta_3 + \hbar^2(k - k_3)^2 / 2\mu_3$. Neutron-scattering measurements,⁸ although consistent with the LP spectrum at small wave numbers k , do show deviations from the quadratic form which increase with increasing momentum. Unfortunately, because of resolution problems at the larger values of k , these measurements are inconclusive regarding the existence of a possi-

ble minimum.

Very recently Bhatt¹² has argued that the threshold for a ³He quasiparticle to emit a ⁴He roton leads to an energy spectrum which deviates from the LP form. For $k \lesssim 2 \text{ \AA}^{-1}$, his calculated spectrum has been parametrized by

$$\epsilon = (\hbar^2 k^2 / 2m_s^*) (1 - k^2 / k_0^2), \quad (1)$$

with $k_0 \approx 3.4 \text{ \AA}^{-1}$. This spectrum is consistent with the neutron, second-sound velocity (u_2),^{2,7} and normal-fluid density (ρ_n)⁵ data.

In this Letter, high-precision specific-heat results on seven very dilute solutions of ³He in ⁴He are presented. They cover the concentration range from 40 to 10 000 ppm and are in the temperature range from 70 mK to 1 K. The measured specific heat differs substantially from that of an ideal (LP) Fermi gas. The results are also clearly inconsistent with the existence of a ³He "roton" minimum described by the parameters derived¹³ from the ρ_n measurements. In addition, there are serious discrepancies with Bhatt's calculations which indicate that the behavior of these mixtures is not understood.

The measurements were made in a thin-walled copper calorimeter¹⁴ which had a volume of 40 cm³. Three graphite support tubes, a quartz filling capillary (0.005 cm i.d., 0.010 cm o.d. by 80 cm long), the electrical leads to the heater and thermometers, and a 15-cm length of 0.004-cm-diam copper wire provided the weak thermal connection to the mixing chamber of a dilution refrigerator. Because the cell was very well isolated thermally and because no heat switch was used, it was possible to cool a mixture sample, with its large heat capacity, only extremely slowly (2 mK per day for a 3000-ppm solution). This difficulty was circumvented by heat sinking the filling capillary on its way to the mixing chamber in several places and by admitting each of the mixtures to the calorimeter extremely slowly. Thus the sample was cooled to near the mixing chamber temperature before it entered the cell. When the cell was completely filled, as indicated by the sharp onset of cooling, a hydraulically operated valve located on the mixing chamber was closed, confining the sample to constant volume. The molar volume of each of the samples was very near that corresponding to vapor pressure. The heat capacity was measured with a precision of about 0.1% using the standard heat-pulse technique with temperature steps equal to 5% of the temperature.

The specific-heat data for the seven mixtures

(open circles) as well as the results¹⁴ for pure ⁴He (<0.005 ppm ³He, solid circles) are shown on a log-log plot in Fig. 1. Previous measurements¹⁵⁻¹⁷ of the specific heat of ³He-⁴He mixtures have only been carried out for ³He concentrations greater than 1% and so a detailed comparison between these and the present, considerably more precise results cannot be made; there is, however, overall consistency. For $T \lesssim 0.7$ K the data in Fig. 1 for pure ⁴He fall along a nearly straight line corresponding to the phonon contribution which is dominated by the term proportional to T^3 . The deviation from this linear behavior at higher temperatures is due to the roton term. With the addition of even very small amounts of ³He, the low-temperature specific heat is modified considerably. On the assumption that the ³He quasiparticles can be treated quantitatively as an independent excitation system, the heat capacity assigned to the ³He quasiparticles is then simply the total heat capacity minus the phonon and ⁴He-roton contributions and is for $T > T_F$ roughly equal to the classical value $\frac{3}{2}XR$. Here R is the gas constant and X is the molar concentration. Previous experimental results indicate^{6,8,18} that both the phonon and roton parts of the pure ⁴He dispersion relation are not appreciably altered by the presence of small amounts of ³He. In addition, as explicitly demonstrated in Fig. 1, the

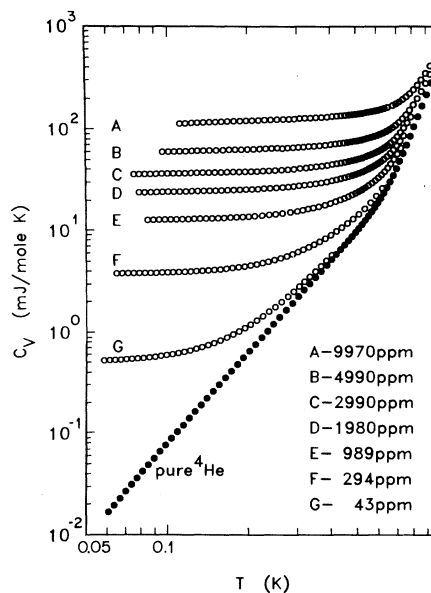


FIG. 1. Specific heat of dilute solutions of ³He in ⁴He. The ³He concentrations are those measured before admitting the samples to the calorimeter.

phonons and rotons in the mixtures make only a small contribution to the total heat capacity at low temperature. Thus over a significant tem-

perature range the specific heat per mole of solution attributed to the ^3He quasiparticles is accurately given by

$$C_V(^3\text{He}) = C_V(\text{mixture}) - C_V(^4\text{He}). \quad (2)$$

In Fig. 2, $C_V(^3\text{He})$ divided by $\frac{3}{2}XR$ is plotted as a function of T . Only points for which the phonon-plus-roton contribution was less than half of the total specific heat are shown. The error bars correspond to an estimated uncertainty in the phonon-roton term of 1%. Note that the size of the error bars decreases rapidly with decreasing temperature. The Fermi temperature is proportional to $X^{2/3}$ and for the 1% mixture is approximately 120 mK. Thus most of the data for each of the samples was obtained for $T \geq T_F$. If the LP spectrum were correct, the measured values for $C_V(^3\text{He})$ should be at most several percent lower than the limiting classical value $\frac{3}{2}XR$, and approaching that value with increasing temperature. Figure 2 clearly demonstrates that this is not the case.

If one assumes Bhatt's parametrization of his calculated spectrum, the specific heat for $T \gg T_F$ is given by

$$C_V/\frac{3}{2}XR \approx 1 + 5T/\tau + 90T^2/\tau^2, \quad (3)$$

with

$$\tau \equiv \hbar^2 k_0^2 / 2m_3^* k_B.$$

With the use of Bhatt's value for k_0 and $m_3^* = 2.3m_3$, this becomes $C_V/\frac{3}{2}XR \approx 1 + 0.12T + 0.06T^2$. Although the slope of the measured quasiparticle specific heat (Fig. 2) for $T \gg T_F$ is in reasonable agreement with the average slope determined using Eq. (3), the extrapolation of the high- T behavior to $T = 0$ does not pass through unity. To force this to occur would require assigning to the mixtures values of X as much as 6% smaller than the concentrations measured before admitting the gas samples to the calorimeter. It is difficult to estimate the size of possible heat-flush effects but it is expected to be much smaller than this discrepancy in the concentration. At lower temperatures the data at the intermediate concentrations show a small knee near 150 mK. For a spectrum of the form given by Eq. (1) this structure in the specific heat should not be present.¹⁹ Instead the behavior at high T should, with decreasing temperature, smoothly round over near T_F and become directly proportional to T .

Shown in Fig. 2 as solid curves is the specific heat²⁰ of an ideal Fermi gas (LP) computed using values of X which are, except for the most dilute

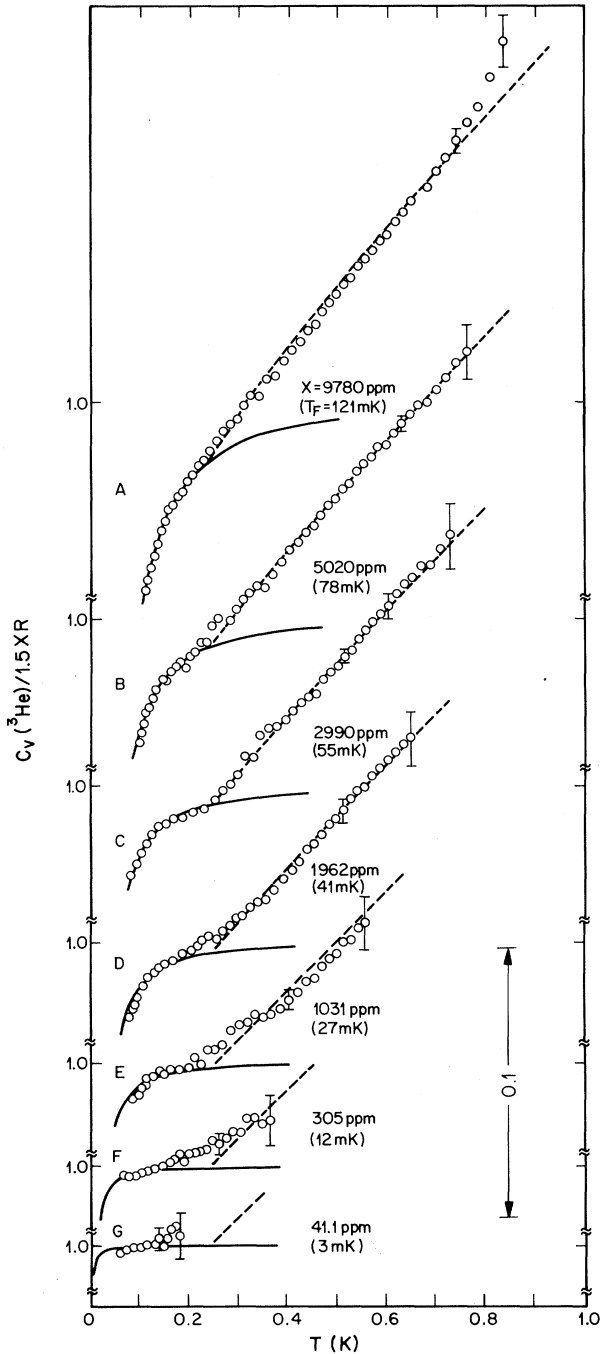


FIG. 2. Specific heat of the ^3He quasiparticles. The solid curves give the specific heat of an ideal Fermi gas (LP) computed using the concentrations given in the figure and $m_3^* = 2.28m_3$. For $T > T_0 = 0.245$ K the dashed curves correspond to $C_V^{LP} + \frac{3}{2}XRS(T - T_0)$ with $S = 0.20 \text{ K}^{-1}$.

samples, within 1% or 2% equal to the measured concentrations. These values of X given in Fig. 2 can be compared with the measured concentrations listed in Fig. 1. The curves describe the data very well at low temperatures (in agreement with the findings of Anderson *et al.*¹⁷ for $T < 100$ mK) and indicate that there is a relatively sharp onset for the deviations from the LP behavior near 0.25 K. This suggests that an activation energy may be involved. Thus an attempt was made to fit the data using an expression which is the sum of the LP²⁰ and ³He "roton"¹¹ contributions. There were three adjustable parameters: X , $A \equiv k_3^2 \mu_3^{1/2}$, and Δ_3 . This function, however, was not adequate to describe the data. For each of the samples there were considerable systematic deviations which increased in size with increasing X , and thus the best-fit parameters were quite sensitive to the exact temperature range of the fits. When all of the data shown in Fig. 2 were included in the analysis, the best-fit values of Δ_3/k_B and μ_3/m_3 ($k_3 \approx 1.9 \text{ \AA}^{-1}$ assumed) ranged from 2.4 K and 8×10^{-5} at $X \approx 1000$ ppm to 3.5 K and 9×10^{-4} at $X \approx 10000$ ppm. Note that the extremely small effective mass implies an unrealistically sharp minimum.

Much better fits were obtained using an *empirical* expression which was equal to C_V^{LP} for $T < T_0$ and equal to the sum of C_V^{LP} and $\frac{3}{2}XRS(T - T_0)$ for $T \geq T_0$. The parameters X , S , and T_0 were adjustable. This analysis yielded best-fit values of S and T_0 for the five highest-concentration samples which were nearly independent of X . The dashed curves for all of the samples in Fig. 2 were plotted using $S = 0.20 \text{ K}^{-1}$ and $T_0 = 0.245 \text{ K}$. A specific heat with a similar temperature dependence can be generated²¹ using a quasiparticle spectrum which deviates from the quadratic form only above $k_c \approx 0.8 \text{ \AA}^{-1}$ ($\epsilon/k_B \approx 2.3 \text{ K}$). For example, the purely phenomenological spectrum

$$\epsilon = \begin{cases} \hbar^2 k^2 / 2m_3^*, & k < k_c, \\ (\hbar^2 k^2 / 2m_3^*) [1 - 0.2(k - k_c)/k], & k > k_c, \end{cases} \quad (4)$$

describes the data well up to $T = 0.6 \text{ K}$. At higher temperatures, the computed specific heat is too small, which indicates that at the higher values of k ($k \gtrsim 1.5 \text{ \AA}^{-1}$) the deviations from the LP form are larger than those given by Eq. (4). Although we know of no microscopic mechanism for gen-

erating such a spectrum, we point out that it is also consistent with the neutron, ρ_n , and u_2 data. This does not, however, imply that other interpretations of the experimental results are excluded.

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