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## Inelastic Neutron Scattering from a Liquid <sup>3</sup>He-<sup>4</sup>He Mixture\*

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We report inelastic-neutron-scattering measurements on liquid <sup>3</sup>He<sub>0.05</sub><sup>4</sup>He<sub>0.95</sub> in the  $Q$  region from 0.8 to 2.3 Å<sup>-1</sup> at 1.6 K. The shifts and increases in natural linewidth of the single excitation peaks relative to pure <sup>4</sup>He at the same temperature were determined by a least-squares fitting procedure. The shifts vary from a small, possibly negative, value at the roton minimum to positive values around 0.5 K at both ends of the  $Q$  range. The full widths at half-maximum of the extra broadening vary from 0.5 to 1.5 K over the same range.

Inelastic-neutron-scattering measurements on liquid <sup>4</sup>He demonstrated as early as 1958 the unique nature of the elementary excitations in that quantum liquid.<sup>1</sup> Neutron measurements on liquid <sup>3</sup>He and <sup>3</sup>He-<sup>4</sup>He mixtures, however, have been discouraged by the large ratio of absorption to scattering cross section for <sup>3</sup>He nuclei (about 5000:1 at thermal energies). Information about the elementary excitations in these liquids has therefore been obtained indirectly. The most direct measurements reported so far on <sup>3</sup>He-<sup>4</sup>He mixtures have been those recently made of zero-momentum roton pairs by Raman scattering.<sup>2</sup> The information obtained, however, is limited to the roton minimum and can be influenced by interactions between the rotons making up the pair. Fortunately improvements in the reliability and accuracy of the neutron method made in the last few years, together with the recent adoption of correlation techniques,<sup>3</sup> now make it possible to measure highly absorbing systems such as <sup>3</sup>He and its solutions. As the first part of a program to study the excitations in these fluids, we present here results for a 5% solution of <sup>3</sup>He in <sup>4</sup>He at 1.6 K. At this temperature the <sup>3</sup>He atoms form essentially a classical system, but the influence of this system on the <sup>4</sup>He excitations neverthe-

less provides unique information on the nature of the <sup>3</sup>He-<sup>4</sup>He interaction.

The measurements were made on the thermal-neutron time-of-flight system<sup>4</sup> at the CP-5 Reactor at Argonne, with a 4.05-Å beam produced by reflection from pyrographite monochromators and the correlation chopper described by Sköld.<sup>3</sup> Data were collected in fifteen independent groups of three detectors each. The mean scattering angles ranged from 30° to 108° to cover a  $Q$  range of 0.8 to 2.5 Å<sup>-1</sup> (for elastic scattering). The angles subtended by the groups of detectors at the sample ranged from 1.8° to 3.0°, depending on the spacing. The channel width for the time-of-flight analysis was 20 μsec so that 251 time channels (the number of elements in the pseudorandom sequence) equaled the rotor period of 5020 μsec. Runs were made on the empty container (44 h), liquid <sup>4</sup>He as a baseline reference (135 h), and the <sup>3</sup>He<sub>0.05</sub><sup>4</sup>He<sub>0.95</sub> mixture (636 h). The samples were contained in a holder consisting of a parallel array of Al-alloy capillary tubes, 0.55-mm internal diameter, similar to that used in an earlier measurement on liquid argon.<sup>5</sup> The volume of sample seen by the neutron beam was 1.45 cm<sup>3</sup>. The holder was attached to the tail-piece of a standard helium cryostat in which the

helium space was continuously pumped. The sample holder was connected to a reservoir bottle and a Bourdon tube to measure the vapor pressure. To ensure that the sample container was full, the  $^4\text{He}$  and the mixture were run at pressures of 20 and 25 Torr, respectively, somewhat above the standard volumes and pressures at 1.6 K. This meant that the fluid extended part way up the capillary tube connecting the sample holder to the external pressure system. The temperature profile of the sample was determined from (a) the resistance of a Ge resistor inserted in the bottom of the sample holder, (b) the pressure above the liquid helium in the pumped cryostat, and (c) the equilibrium pressure in the Bourdon tube measured after closing the reservoir bottle at various points in the process of condensing the sample from the reservoir into the sample holder. From these measurements it was determined that the temperature in the part of the sample seen by the neutron beam was within the range  $1.57 \pm 0.1$  K during the course of data collection.

Samples of the gases used in the experiments were analyzed by mass spectrometry for chemical and isotopic composition. The chemical impurities ( $\text{H}_2\text{O}$ ,  $\text{N}_2$ , and  $\text{O}_2$ ) in a typical sample totaled  $100 \pm 20$  ppm by volume of gas. The total mixture sample in the experimental system (sample holder, reservoir, and gauge) was analyzed to have 4.91%  $^3\text{He}$  in  $^4\text{He}$ , by volume. Care must be taken with liquid-vapor systems to prevent dilution of the liquid by the preferential evaporation of  $^3\text{He}$ . If we assume that the gas in the reservoir, manifold, and gauge space was enriched in  $^3\text{He}$  by the enrichment factor<sup>6</sup> appropriate for a vapor in equilibrium with a 5% solution at 1.6 K—certainly an extreme assumption—the  $^3\text{He}$  concentration in the liquid would be 4.43%.

The raw data were decorrelated to give a representation of the data as it would have been obtained with a single-slit chopper. At all succeeding stages in the data analysis the correct statistical errors applicable to the correlation technique were used, but in all other respects the data were reduced to the scattering function form  $S(Q, \omega)$  exactly as described by Copley, Price, and Rowe.<sup>7</sup> The container scattering data were corrected for attenuation by the sample and subtracted from the sample data in the regions where the container data showed any appreciable structure (primarily at zero energy transfer). The empty container run showed no structure in the region of the excitation peaks for any of the an-

gles, and no empty container correction was applied in these regions. The data at various angles were normalized to each other (and approximately to absolute cross sections) using the scattering from a 1-mm slab of vanadium as a reference. The results for the  $^4\text{He}_{0.95}^3\text{He}_{0.05}$  mixture were then corrected for the  $^3\text{He}$  absorption with a cross section of 5327 b at 2200 m sec<sup>-1</sup>, varying as  $1/v$ .

The scattering function thus derived for pure  $^4\text{He}$  was then fitted by a single Gaussian at each angle, with the Gaussian position, width, and area as fitting parameters. The range of energy transfers included in this fitting was chosen to span the well-defined peaks which were clearly exhibited at each angle. Since the data are at constant angle rather than constant  $Q$ , they were multiplied by the factor  $|J|$ , where the Jacobian  $J = 1 - \hat{k}_1 \cdot \nabla\omega/v_1$ . The mixture results were then fitted by a Lorentzian broadened by a Gaussian of width equal to that found for the pure  $^4\text{He}$ . Once again, the area, the width, and the position of the Lorentzian were left as parameters. The results of the data analysis and fitting are shown in Fig. 1 for three representative angles. The values of

$$\chi = [(F_{\text{obs}} - F_{\text{calc}})^2 / \sigma^2]^{1/2}$$

obtained from these fits were between 0.9 and 2.4, where  $\chi = 1.0$  would be considered a perfect fit. Although it is not shown in this figure, at each angle a small amount of scattering remained near zero energy transfer for the mixture data after subtracting the sample-container scattering. However, these data are of such low statistical accuracy (since the sample container correction is large) that we cannot be quantitative about them. If they are due to incoherent scattering from  $^3\text{He}$ , they imply narrower quasielastic peaks than would be expected from diffusion-constant measurements.<sup>8</sup>

From the results of the fitting procedures described, we were able to extract the energy shifts and increased widths of the neutron groups measured in the mixture. These results are shown in Fig. 2, where the error bars represent standard deviations estimated from the fitting procedure. As can be seen from Fig. 2, and from Fig. 1, the neutron groups for the mixture are everywhere broadened with respect to those in pure  $^4\text{He}$ . The energy shifts are in general to higher frequencies, although there appears to be a small negative shift in the vicinity of the roton minimum. The integral of the one-phonon scat-

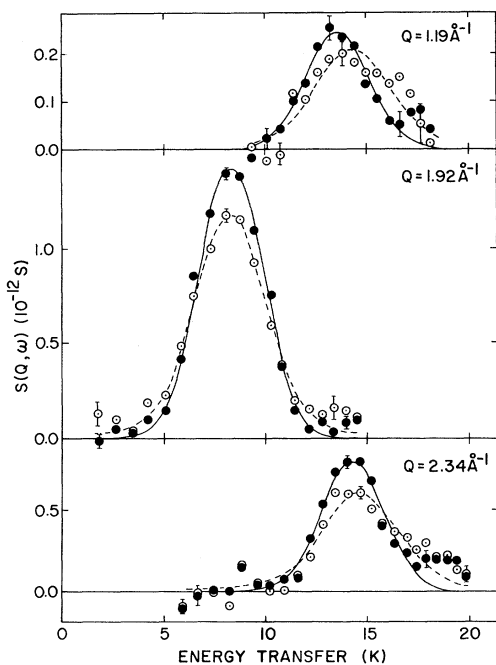


FIG. 1. Experimental data for  ${}^4\text{He}$  (solid circles) and the  ${}^4\text{He}_{0.95}{}^3\text{He}_{0.05}$  (open circles) liquids at three selected angles of scattering for a temperature of 1.6 K. The solid and broken lines represent the functions fitted to the data as described in the text. The value of wave-vector transfer  $Q$  shown is determined at the energy corresponding to the peak. The change of wave vector with energy over the peaks is less than  $0.01 \text{ \AA}^{-1}$  at the smallest angle, rising to  $0.07 \text{ \AA}^{-1}$  at the largest angle.

tering  $Z(Q)$  for both samples has been evaluated, and the measured ratio  $Z({}^4\text{He})/Z(\text{mixture}) = 1.01 \pm 0.09$  over all fifteen  $Q$  values measured. The values for  $Z(Q)$  in  ${}^4\text{He}$  are everywhere 10–20% lower than those measured by Cowley and Woods<sup>1</sup> at 1.1 K. The dispersion relation for pure  ${}^4\text{He}$  shown in Fig. 2 is in good agreement with earlier neutron-scattering results,<sup>1,9</sup> yielding a roton energy  $\Delta$  of  $8.31 \pm 0.02 \text{ K}$  at 1.6 K. Note that this is *only* the fitting error (standard deviation), and does *not* include estimates of systematic errors which would increase this error by a factor of approximately 10. However, relative measurements of  ${}^4\text{He}$  compared to the mixture do have errors of this magnitude.

The energy shift observed at the roton minimum in this experiment is  $-0.04 \pm 0.03 \text{ K}$ , while the full width of the broadening observed is  $0.89 \pm 0.09 \text{ K}$ . The results agree reasonably well with recent laser Raman studies<sup>2</sup> of the two-roton line, which for a 5% concentration of  ${}^3\text{He}$  show no observable shift and increases in the full width of

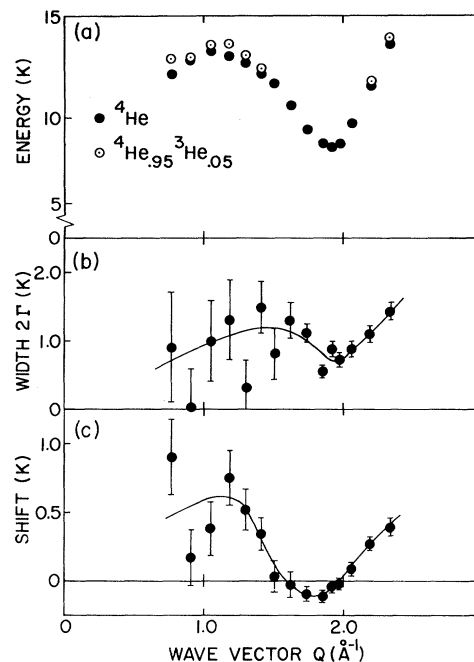


FIG. 2. (a) Energy-wave-vector dispersion relation as measured for  ${}^4\text{He}$  and  ${}^4\text{He}_{0.95}{}^3\text{He}_{0.05}$  at 1.6 K. Where no result is shown for the solution, the point is indistinguishable from the corresponding point for  ${}^4\text{He}$ . The estimated errors are everywhere less than the point size. (b) Increase in full width at half-maximum for the  ${}^4\text{He}_{0.95}{}^3\text{He}_{0.05}$  results derived from the fitting procedure described in the text. (c) Shift in excitation energy for  ${}^4\text{He}_{0.95}{}^3\text{He}_{0.05}$  compared with pure  ${}^4\text{He}$  at 1.6 K. The continuous curves in (b) and (c) are merely a guide for the eye.

a single excitation of 0.2 and 0.6 K at temperatures of 0.6 and 1.3 K, respectively. Both of these experiments are in strong disagreement with estimates of the shift made from fourth-sound-velocity, ion-mobility, and normal-fluid-fraction measurements. There have been recent theoretical estimates of the roton energy shift and width for solutions of  ${}^3\text{He}$  in  ${}^4\text{He}$ . The present results agree with the estimates of Bagchi and Ruvalds<sup>10</sup> in that the energy shift of the roton minimum is small compared with the width, but show that this is not the case for other values of the wave vector. They do not confirm the large splitting predicted by Bartley, Robinson, and Wong.<sup>11</sup> It should be noted that the decrease in density of the solution would be expected to *raise* the energies by about 0.1 K at the roton minimum on the basis of the result of Dietrich *et al.*<sup>9</sup> for  $(\rho/\Delta)\partial\Delta/\partial\rho$  at the roton minimum.

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### High-Temperature Transport Properties of Liquid He<sup>3</sup>

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The viscosity of liquid He<sup>3</sup> is shown to saturate at high temperatures like  $A+B/T$ , in good agreement with experiment; the spin-diffusion coefficient should saturate, too, like  $A' - B'/T$ , while the thermal conductivity varies linearly with  $T$ . The same formulas reduce at very low temperature to the results of the paramagnon model.

I consider the temperature dependence of the transport properties of liquid He<sup>3</sup> in its normal state (above 3 mK up to  $\sim 3$  K). It is shown that the same formula provides a good description of their temperature variations at high temperature as well as at very low temperature, where it leads back to the well-known paramagnon result. At high temperature we thus give explicit formulas for the viscosity  $\eta$ , the spin-diffusion coefficient  $D$ , and the thermal conductivity  $K$ .

Up to 500 mK, Rice,<sup>1</sup> using the paramagnon model for nearly magnetic Fermi liquids,<sup>2</sup> gave a theoretical description of  $\eta^{-1} = \text{const} \times T^2$  which appeared to fit the data quite well at very low temperature. But at high  $T$ , Rice's  $\eta^{-1}$  would behave like  $T$ . However, according to Fig. 1, where I have gathered the results of several experiments,<sup>3</sup>  $\eta^{-1}$  does not seem to follow such a prediction and looks more as if it would slowly saturate at high  $T$ .

It is shown here that this is indeed what happens. Actually the Rice theory as it stands cannot apply at high temperature since Rice used only the low-temperature form of the paramagnon propagator.

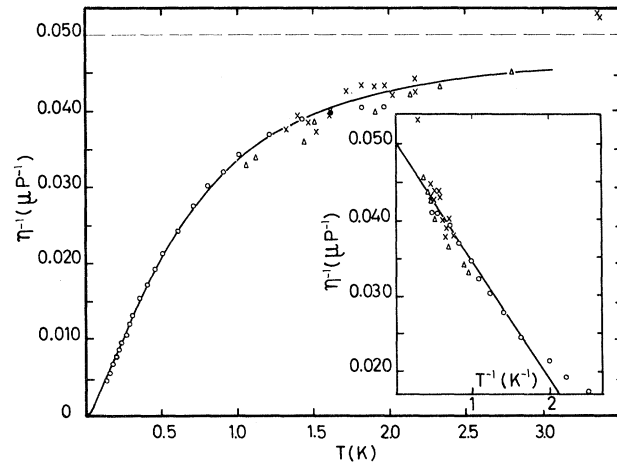


FIG. 1. Experimental values of  $\eta^{-1}$  versus  $T$ : circles and triangles, from Tables 1 and 2, respectively, of Betts *et al.* (Ref. 3); crosses, from Table II of Taylor and Dash (Ref. 3); below 0.1 K, solid curve drawn according to  $\eta T^2 = \text{const}$  found by Abel and co-workers, Refs. 3 and 10; solid curve drawn through the experimental points is a guide for the eye; dashed line, theoretical saturation obtained by extrapolation at  $T_\infty$  of the straight line of the inset. Inset, plot of the high- $T$  values for  $\eta^{-1}$  versus  $T^{-1}$ ; straight line given by the theory.