Kinetic Energy and Condensate Fraction in ³He-⁴He Mixtures

Y. Wang

The Intense Pulsed Neutron Source, Argonne National Laboratory, Argonne, Illinois 60439

P. E. Sokol

Department of Physics, The Pennsylvania State University, University Park, Pennsylvania 16802

Deep inelastic neutron scattering measurements have been carried out to measure the kinetic energy and condensate fraction in liquid ³He-⁴He mixtures. Samples with ³He concentration on the order of 10% were studied at 1.4 K and 2.3–2.5 K. The condensate fraction was found to be 0.18 ± 0.03, much larger than theoretical predictions. The measured kinetic energies of the ³He and ⁴He in the mixture remain the same as their values in the pure phase, in disagreement with current theoretical predictions.

The single particle atomic momentum distribution, $n(p)$, and the Bose condensate fraction, n_0, in liquid ⁴He are central to our understanding of the microscopic dynamics of the liquid. In particular, the appearance of a superfluid phase, which has been the subject of intensive study, is intimately linked to the concept of a broken Bose symmetry, with the condensate fraction acting as the microscopic order parameter of the Bose condensed phase [1]. There has been a great deal of work directed towards understanding the microscopic dynamics of the liquid and the condensate and recently a rather complete understanding has emerged for pure liquid ⁴He [2,3].

Less well understood, at the micrscopic level, are the effects of ³He, a fermion, on the Bose condensed ground state and the condensate fraction of liquid ³He-⁴He mixtures. At the macroscopic level, the addition of ³He modifies the characteristic signature of the Bose condensed phase: superfluidity. The addition of ³He suppresses the superfluid transition and reduces the superfluid fraction [4]. However, at the microscopic level, recent calculations [5,6] have suggested that n_0 is enhanced as the ³He concentration, x, is increased. The predicted enhancement is small, increasing n_0 from 0.10 for the pure liquid to 0.12 for $x = 0.10$. While small, the enhancement is quite significant in that it highlights the difference between the superfluid fraction, a macroscopic transport property, and the condensate fraction, the microscopic order parameter of the Bose condensed phase.

The most direct information on the momentum distribution is obtained from deep inelastic neutron scattering measurements at high momentum transfers Q. The impulse approximation (IA) [7], which is exact in the $Q \rightarrow \infty$ limit, directly relates $n(p)$ to the measured scattering. At finite Q, deviations from the IA, known as final state effects (FSE), modify the scattering from the IA prediction. At sufficiently large Q, these FSE are calculable [8] in helium and direct information on the momentum distribution and n_0 can be obtained.

In this Letter we report the first deep inelastic neutron scattering measurements of liquid ³He-⁴He mixtures in the normal and superfluid phases. We observe an enhancement of the condensate fraction, as predicted theoretically. However, our experimental value of n_0, 0.18 ± 0.03, represents an enhancement of 80% over the pure liquid, far larger than the theoretical predictions of 0.11–0.12. The single particle kinetic energies, $\langle E_{kin} \rangle$, of the ³He and ⁴He atoms in the mixture were also measured. Little change from the pure systems was observed, in disagreement with theoretical predictions of a significant increase of $\langle E_{kin} \rangle$ for the ³He atoms.

The scattering of neutrons by a condensed sample is described by the dynamic structure factor which, at high Q, can be expressed by the impulse approximation in terms of a Compton profile,

$$J_{IA}(Y) = \int_Y^\infty p n(p) dp = \frac{Q}{M} S_{IA}(Q, \omega),$$

where p is the atomic momentum and M is the mass of the scattering particle. The scaling variable Y is defined as $Y \equiv (M/Q)(\omega - \omega_r)$, where ω is the energy transfer and $\omega_r = \hbar^2 Q^2/2M$ is the recoil energy. The ability to express the scattering in terms of a single scaling variable simply reflects the single particle nature of the scattering in the IA.

The IA is strictly valid only in the infinite Q limit. At finite Q's deviations from the IA become important. These deviations distort the scattering from the IA prediction and must be taken into account if accurate information on the momentum distribution and n_0 is to be obtained. For Q's above 10–15 Å^{-1}, the scattering approximately obeys Y-scaling behavior and can be expressed in terms of a generalized Compton profile,

$$J(Y,Q) = \frac{Q}{M} S(Q, \omega),$$

where Y has the same definition as above. In terms of the generalized Compton profile, the effect of FSE on the scattering can be expressed in the form of a convolution,

$$J(Y,Q) = \int R(Y - Y', Q) J(Y') dY',$$

where $R(Y,Q)$ is the final state broadening function at a

momentum transfer Q. There have been several recent calculations [8,9] for ^{4}He which make explicit predictions for the form of $R(Y,Q)$ which are in good agreement with experiment [10]. Predictions of $R(Y,Q)$ for ^{3}He and mixtures are not available, but at the large Q's used in these measurements the hard core of the potential dominates and it is expected to be identical to the ^{4}He results [11] when density variations are taken into account.

The measurements were carried out using the PHOENIX chopper spectrometer [10] at the Intense Pulsed Neutron Source (IPNS) at Argonne National Laboratory. A momentum transfer of 23 Å^{-1}, at the helium peak, was used as in previous measurements of bulk liquid ^{4}He. Two different ^{3}He-^{4}He mixtures, both with concentrations on the order of $x = 0.10$, were measured. The samples were produced by mixing in the gas phase before condensing into a slab geometry sample cell attached to a ^{3}He refrigerator. The mixture concentrations were determined from the amounts of gas mixed, the relative scattering intensity of the ^{3}He and ^{4}He, and by mass spectroscopic analysis. All three techniques agreed within the experimental errors. Measurements were carried out for approximately two weeks for each sample and temperature studied. The long counting times were necessitated by the large neutron absorption cross section of the ^{3}He in the mixture. The scattering was measured in the normal liquid at 2.3–2.5 K and in the superfluid at 1.4 K, as determined by germanium resistance thermometry, with the superfluid transition occurring at 1.99 K for the concentrations studied.

The scattering from the normal liquid at 2.3 K for a mixture with $x = 0.10$ is shown in Fig. 1. The ^{3}He and ^{4}He scattering are separated due to their different masses, with the peak at $Y = 0$ due to the ^{4}He and the peak at $Y = -3$ due to the ^{3}He. The ^{3}He peak is offset from the IA prediction of $Y = 0$ due to the use of the ^{4}He

mass in the calculation of the Y scale. When a Y scale based on the ^{3}He mass is used the ^{3}He peak is centered at $Y = 0$. Thus, both the ^{4}He and ^{3}He scattering are consistent with the predictions of the IA in that they are symmetric and centered at $Y = 0$, when the appropriate mass is used.

The scattering from both the ^{4}He and the ^{3}He in the normal liquid phase of the mixture is nearly Gaussian, similar to the behavior observed for the corresponding pure liquids. The solid curve is a fit of a model $n(p)$, consisting of a sum of Gaussians convoluted with instrumental resolution and FSE to the data. The results indicate that the ^{4}He scattering is slightly non-Gaussian, just as in the pure liquid. By comparison, the scattering from the pure liquid is slightly broader than that of the ^{4}He in the mixture. This is reflected in the kinetic energy of the ^{4}He in the mixture being slightly lower than for the pure liquid, as discussed below. This narrower $n(p)$ and lower E_{kin} are consistent with the lower density of the mixture. However, these results are in agreement within the respective errors of the measurements, so no quantitative conclusions can be drawn.

The scattering in the superfluid phase of the mixture at 1.4 K is shown in Fig. 2. The scattering from the ^{3}He component has not changed significantly upon cooling. The scattering from the ^{4}He, on the other hand, has become much more peaked about $Y = 0$ reflecting the appearance of a condensate in the superfluid phase. A comparison of the scattering to that observed in the pure liquid shows that the buildup at small Y is much larger than for a comparable temperature in the normal liquid.

The condensate fraction can be extracted directly from the observed scattering using a model $n(p)$ which incorporates the condensate and the known singular behavior at small p. This model has been successfully applied to pure liquid ^{4}He over a wide range of temperatures and
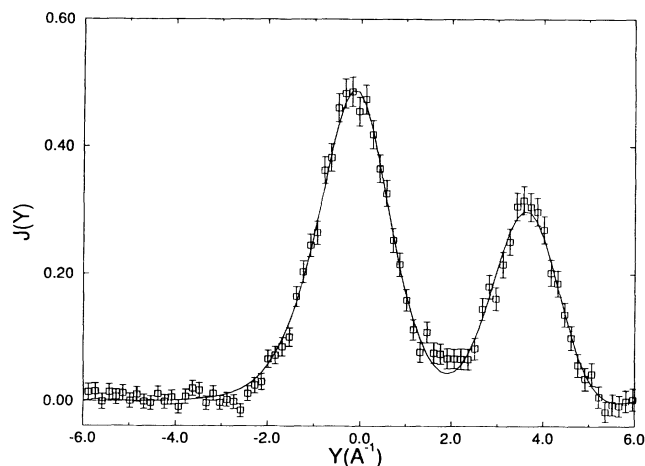

FIG. 1. The ^{4}He scattering in the normal liquid from a 9.5% mixture at 2.3 K. The solid line represents the result of fitting a model $n(p)$ consisting of a sum of Gaussians.
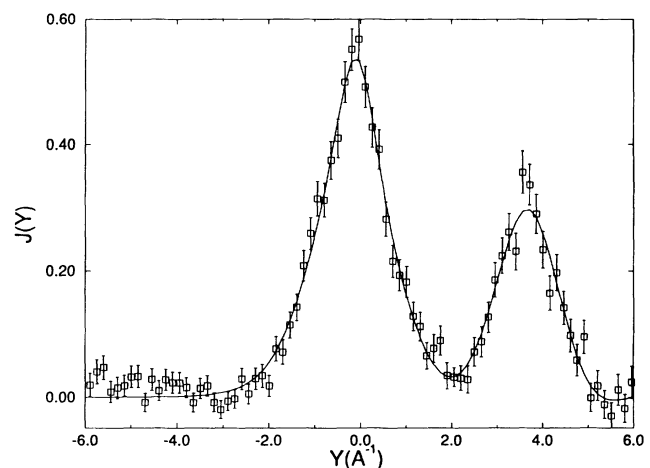

FIG. 2. The ^{4}He scattering in the superfluid in a 9.5% mixture at 1.4 K. The solid line represents the fit of the model $n(p)$ discussed in the text.

densities [12,13]. The main difference between mixtures and the pure liquid, in terms of this model, is the singular behavior of $n(p)$ at small p. The main effect of the ^{3}He on this singular behavior is through its effect on the density and speed of sound of the liquid, which are well known [14]. Analysis using other methods [15] to extract n_0 yields similar results.

A fit of the above model by the superfluid results, shown by the solid curve in Fig. 2, yields a value for the condensate fraction in the mixture of 0.18 ± 0.03. This represents an enhancement of nearly a factor of 2 over the pure liquid value [16] of 0.10. The experimental value is also much larger than the theoretical predictions, which are on the order of 0.11–0.12. Earlier results [5] using less sophisticated trial wave functions predict higher values of n_0, both in the pure liquid and in mixtures. However, if these values are scaled to the accepted value of n_0 in the pure liquid, all theoretical predictions are in reasonable agreement. Our results, along with the theoretical predictions, are shown in Fig. 3.

The origin of this large discrepancy is not clear. The experimental work was carried out at the same density as the theoretical calculations. The calculations are at $T = 0$, whereas the experiments were carried out at finite temperature. However, we expect the condensate fraction to decrease with increasing temperature which would only increase the discrepancy. An estimate of the zero temperature value of the condensate, based on the temperature dependence of the bulk liquid, gives $n_0 = 0.22$ at $T = 0$, an even bigger disagreement with the theoretical predictions.

The single particle kinetic energy per atom (E_{kin}) can be obtained directly from the second moment of the momentum distribution. As such, it provides a useful

FIG. 3. The ^{4}He condensate fraction as a function of the ^{3}He concentration. The cross symbol represents the experimental results, the triangle represents the HNC calculation [6], and the circle represents the VMC calculation [5]. The inset shows the concentration dependence of n_0 over a wider range.

characterization of $n(p)$. Alternatively, the E_{kin} can be obtained directly from the experimental data using the second moment sum rule [17]. The measured E_{kin} is 13 ± 3 K for the ^{4}He and 11 ± 3 K for the ^{3}He in the mixture of 10% ^{3}He concentration. The temperature dependence of E_{kin} is too small to resolve from our data due to the experimental uncertainty.

The E_{kin} of both the ^{4}He and the ^{3}He are, to within experimental uncertainty, the same as that of the respective pure liquids [10,16]. The ^{4}He values are consistent with the theoretical predictions which, due to the low ^{3}He concentration, are not significantly different from the pure. The ^{3}He E_{kin}, on the other hand, are nearly a factor of 2 lower than theoretical predictions of E_{kin} [5,6].

A possible explanation for this discrepancy lies in the fact that the E_{kin} in quantum liquids is primarily due to the zero point motion resulting from the confinement of an atom by its neighbors. The density of the liquid, which determines the average volume per atom, is fixed by the temperature, pressure, and concentration which are accurately known. However, the E_{kin} observed in our measurement, which is consistent with the pure phase value, suggests that the local volume per ^{3}He is the same as in pure ^{3}He. Thus, the ^{3}He atom decreases the local density of the liquid to maintain a volume per atom similar to pure liquid ^{3}He, with a corresponding increase in the local density of the ^{4}He component. This picture is consistent with the theories [18] and measurements [19] of the density of ^{3}He-^{4}He mixtures in which the mixture density is approximately the average of the pure liquid densities weighted by the concentration. The variational calculations, on the other hand, predict that the average volume per atom is the same for ^{3}He and and ^{4}He [20,21] in the mixture, a condition enforced by the trial wave functions used. Thus, the lower ^{3}He E_{kin} measured suggests that a more flexible trial wave function may be needed. We note, however, that a better trial wave function, reflecting the lower local density around a ^{3}He atom, would give a higher local density for the ^{4}He component which would not explain the increase in n_0 by the ^{4}He wave function alone.

Another possibility is that $n(p)$ for the ^{3}He atoms have very long non-Gaussian tails. In the pure liquid, the experimentally determined [22] E_{kin} of 8 K is lower than the theoretical predictions of 12 K, presumably due to the existence of exponential tails in $n(p)$ that are too small to be detected experimentally [23]. Such long tails have also been predicted in mixtures. However, to explain the discrepancy with the experimental results they would need to contain much more of the weight in $n(p)$ than they do in the pure liquid. In addition, we note that the theoretical calculations are at $T = 0$ while our measurements are well above the Fermi temperature for the mixture. In this case we expect that thermal fluctuations will smear out the discontinuity at the Fermi surface and, presumably, the exponential tails.

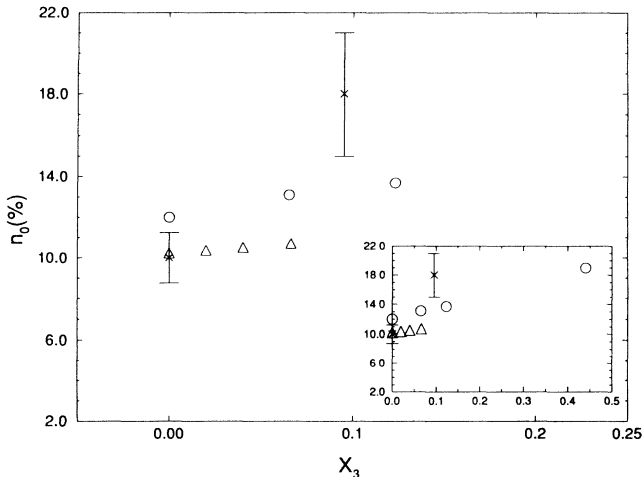In conclusion, we have measured the momentum distri-

bution of ^{3}He-^{4}He mixtures with $x = 0.1$ in both the normal liquid and superfluid phases using deep inelastic neutron scattering. The experimentally determined condensate fraction in the superfluid of 0.18 is much larger than any of the current theoretical predictions. In addition, we extracted values for the E_{kin} of the ^{3}He and ^{4}He atoms individually. The results for the ^{4}He atoms are in reasonable agreement with the theoretical predictions while those for the ^{3}He are much lower than theoretical predictions. This suggests either that the local environment of the ^{3}He in the variational calculations is too confined or that the ^{3}He $n(p)$ have extremely long tails, even at the high temperatures in these measurements.

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