Bose-Einstein Condensation in Liquid $^4$He in Vycor

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(Received August 6, 2002; revised November 11, 2002)

We present neutron scattering measurements of Bose-Einstein condensation (BEC) in liquid $^4$He confined in Vycor. The data show clear evidence of a condensate in Vycor with a condensate fraction comparable to that of bulk superfluid $^4$He, approximately 7.5% at low temperature and SVP. The temperature dependence of $n_0(T)$ is also similar to that in the bulk with critical temperature for BEC, $T_{BEC}$, in the range $1.80 < T_{BEC} < 2.05$ K. The data are not accurate enough to show whether $T_{BEC}$ for Vycor is the same or greater than the depressed critical temperature for superfluidity, $T_c=1.95$ K.

1. INTRODUCTION

The celebrated Landau theory of liquid $^4$He attributes superfluidity to the existence of well defined phonon-roton excitations. Modern data shows that well defined phonon-roton excitations at higher wave-vectors exist only in the superfluid phase, not in normal $^4$He. In a complementary picture, superfluidity is attributed to Bose-Einstein condensation (BEC). Similarly, INS measurements verified that there is a condensate in the superfluid phase ($n_0 = 7.25\%$ at $T = 0$ K) and definitely no condensate in the normal phase. In addition, it has been shown theoretically that Bose fluids support sharp, long-lived phonon-roton excitations when there is a condensate. In bulk liquid $^4$He, both superfluidity and well defined phonon-roton excitations appear to arise from BEC and all three properties have a common onset temperature, $T_s=2.177$ K.

When liquid $^4$He is confined in porous media, the transition temperature to superfluidity, $T_c$, is suppressed below $T_s$, the superfluid density,
\( \rho_s(T) \), at low temperature can be reduced below unity, and the critical exponent describing the temperature dependence of \( \rho_s(T) \) immediately below \( T_c \) can be modified from the bulk value.\(^{12,13}\) The higher the degree of confinement, the further \( T_c \) is reduced.\(^{13,14}\) Monte Carlo simulations of \(^4\)He at reduced densities find that \( \rho_s(T) \) is significantly reduced by disorder but that \( T_c \) is little changed.\(^{15}\) Analytic calculations\(^{16}\) applicable to a dilute Bose gas at \( T = 0 \) find that both \( \rho_s(T) \) and the condensate fraction, \( n_0(T) \), are reduced by homogeneous disorder and that \( \rho_s(T) \) is reduced more than \( n_0(T) \). This result is also found\(^{17}\) in Bose fluids at higher densities using Diffusion Monte-Carlo methods. Indeed, complete suppression of superfluidity at finite condensate fraction is possible.\(^{16}\)

INS experiments show that superfluid \(^4\)He in porous media supports well defined, three-dimensional (3D) phonon-roton excitations as in bulk superfluid \(^4\)He.\(^{18-20}\) In addition superfluid \(^4\)He supports two-dimensional (2D) excitations that propagate in the liquid layers adjacent to the media walls.\(^{18-21}\) In Vycor, these 2D layer modes appear to be responsible for differences in thermodynamic and superfluid properties at low temperature from the bulk.\(^{19}\) In contrast to bulk helium, well-defined phonon-roton excitations exist above \( T_c \) in Vycor and Geltech.\(^{19,20}\) This suggests that there is a condensate above \( T_c \). This condensate could be localized\(^{22,23}\) to favorable regions in the porous media separated by regions of normal fluid so that superflow across the whole sample is not possible.

In this paper, we present direct measurements of the condensate fraction of liquid \(^4\)He in Vycor using high energy transfer inelastic neutron scattering. These measurements are very difficult because the sample volume of liquid \(^4\)He in the Vycor is very small (approximately 5 cm\(^3\)) and background subtractions for scattering from the Vycor and from the solid \(^4\)He layers adjacent to the Vycor walls are necessary. The main findings are that \( n_0 \) is not reduced by disorder at low temperature and that the temperature dependence of \( n_0(T) \) is similar in Vycor and in the bulk. The Vycor sample and the neutron scattering experiment are described in Sec. 2. The data analysis and the results are given in Sec. 3 and the discussion in Sec. 4 concludes the paper.

2. EXPERIMENTAL

Vycor, a 30% porous silica glass, was chosen because of its reduced \( T_c = 1.95 \) K and its narrow distribution of pores of 70 Å mean diameter. The Vycor was made using the \(^{11}\)B isotope instead of natural boron in order to reduce the neutron absorption by \(^{10}\)B. The Vycor sample, a slab of dimensions \( 5 \times 5 \times 2 \) cm, was inclined at 45° with respect to the incoming neutron beam in reflection geometry and mounted inside an Al sample cell
attached to a 3He sorption cryostat. A BN shielded piece of aerogel glass was mounted in the cell outside of the neutron beam to absorb any excess 4He, which could be outside the Vycor. Adsorption measurements show that Vycor fills before aerogel.

The experiment was performed using the MARI chopper spectrometer at the ISIS spallation neutron source using an incident neutron energy of 750 meV at an energy resolution of about 15 meV. Wave vectors $Q < 30 \text{ Å}^{-1}$ and energies $\omega < 700 \text{ meV}$ were recorded. To estimate the background, we measured the empty Vycor sample in the cell at $T < 5 \text{ K}$ and a partly filled (five liters of gas equivalent) Vycor at $T = 1.3 \text{ K}$. The latter filling correspond to two layers of 4He on the internal surface of the Vycor. Since the first (and possibly) the second layer on Vycor is solid, this measurement allows us to subtract the contribution coming from the solid 4He. The Vycor was then slowly filled at $T = 4.2 \text{ K}$ to a total of 15 liters of gas, monitoring the vapor pressure to ensure that no bulk liquid was present. Measurements at this filling, which corresponds to 95% filling of the Vycor, were made at temperatures of 0.5, 1.3, 1.8, 2.0, and 2.25 K, spanning the superfluid and normal phases.

As in the case of bulk liquid 4He, the observed INS data is expressed in terms of the dynamic structure factor $S(Q, \omega)$. Standard procedures were employed to correct and transform the data to the net dynamic structure factor arising from the liquid only, convoluted with the instrument resolution. This is conveniently then written in terms of the “$y$ scaling” energy transfer variable $y = (\omega - \omega_R)/v_R$ as $J(Q, y) = v_R S(Q, \omega)$ where $\omega_R = \hbar Q^2/2m$ and $v_R = \hbar Q/m$ are the free atom recoil frequency and velocity, respectively. The instrumental resolution was calculated by Monte Carlo simulation and is much narrower than $J(Q, y)$. We investigated $J(Q, y)$ in detail for wavevectors $24 \leq Q \leq 29 \text{ Å}^{-1}$ where Final State (FS) effects are small but not negligible and $J(Q, y)$ varies slowly with $Q$.

3. RESULTS

The net $J(Q, y)$ of liquid 4He in Vycor and that in the bulk from Ref. 10 are compared in Fig. 1. From the upper frame of Fig. 1, we see that $J(Q, y)$ of superfluid 4He in the bulk and in Vycor are generally the same within statistical precision. Specifically, the width of $J(Q, y)$, which is approximately proportional to the mean square atomic momentum along $Q$, $\langle k_Q^2 \rangle$, and the atomic kinetic energy, $\langle K \rangle = (3\hbar^2/2m)\langle k_Q^2 \rangle$, is the same within error. The statistical precision of the data is much higher in the bulk because a larger volume of 4He can be used and because subtractions of scattering from solid 4He layers on the Vycor walls and from the Vycor itself are not required.
Fig. 1. The observed $J(Q, y)$ of liquid $^4$He in the bulk and in Vycor. The two are the same (top frame) within statistical precision. The changes arising from BEC are shown in the middle (bulk) and lower (Vycor) frames.

The observed $J(Q, y)$ in normal and superfluid $^4$He are compared in the lower two frames of Fig. 1. The condensate contribution to $J(Q, y)$ adds intensity chiefly at $y = 0$ leading to a higher peak in $J(Q, y)$ in superfluid $^4$He. This is most clearly seen in the bulk $^4$He data. The condensate term $n_s R(Q, y)$ in $J(Q, y)$, where $R(Q, y)$ is the FS broadening function, also introduces a right-left asymmetry in the $T = 0.5$ K data. In Vycor the increased peak height of $J(Q, y)$ at $y = 0$ in the superfluid phase is less obvious because of the limited statistical precision of the data.

To analyze the data, we express $J(Q, y)$ as a convolution of the Impulse Approximation (IA) to $J(Q, y)$, $J_{IA}(y)$, and the Final State broadening function, $R(Q, y)$; $J(Q, y) = J_{IA}(y) \otimes R(Q, y)$. The IA is $J_{IA}(y) = \int d k \, n(k) \, \delta(y - k_{\bar{Q}})$ where $k_{\bar{Q}} = k \cdot \bar{Q}$ and $n(k)$ is the atomic momentum.
distribution. $J_{IA}(y)$ is also denoted the longitudinal momentum distribution. The observed data also appears convoluted with the instrument resolution function, $I(y)$, $J_{obs}(Q, y) = J(Q, y) \otimes I(y)$. In our measurements, $I(y)$ is much narrower than $J(Q, y)$ in the $Q$ range considered.\textsuperscript{10}

We represent the momentum distribution in $J_{IA}(y)$ as\textsuperscript{10}

$$n(k) = n_0[\delta(k) + f(k)] + A_1n^*(k). \tag{1}$$

Here $n_0\delta(k)$ is the condensate contribution. The term $n_0f(k)$ arises from a coupling between single particle and density excitations via the condensate and is very sharply peaked at $k = 0$ (see Fig. 1 of Ref. 10). $A_1$ is a constant and $n^*(k)$ is the momentum distribution of the atoms above the condensate ($k \neq 0$) which is normalized to unity. Normalization of $n(k)$ gives $\int dk \, n(k) = n_0[1 + I_f] + A_1 = 1$ where $I_f \equiv \int dk \, f(k) \approx 0.25$.

In bulk $^4$He, there is sufficient precision in the $J(Q, y)$ data to determine $n_0$, the shape of $n^*(k)$ in (1), and $R(Q, y)$. In Vycor, we find the data is accurate enough to determine only one fitted parameter with confidence, essentially the width or second moment of the data. We therefore elect to determine $n_g(T)$ from the second moment of $J(Q, y)$ following a method proposed originally by Sears.\textsuperscript{7} The method is the following. The second moment of $J(Q, y)$ is $M_2 = \int dy \, y^2 J(Q, y)$. Since the FS function $R(Q, y)$ is normalized to unity and its second moment is zero, $M_2$ reduces to the second moment of $J_{IA}(y)$, $M_2 = \int dy \, y^2 J_{IA}(y)$. From the definition of $J_{IA}(y)$ above, this is $M_2 = \langle k_G^2 \rangle = \int dk \, k_G^2n(k)$. Also, when there is a condensate, the first two terms of $n(k)$ in (1) are sufficiently sharply peaked at $k = 0$ that they do not contribute significantly to the second moment $\langle k_G^2 \rangle$. Specifically from (1), $\langle k_G^2 \rangle = \int dk \, k_G^2n(k) \approx A_1 \int dk \, k_G^2n^*(k)$. Thus the condensate does not contribute to the kinetic energy, $\langle K \rangle = (3\hbar^2/2m)\langle k_G^2 \rangle$, and the $\langle K \rangle$ decreases with decreasing temperature as BEC sets in. The $n_0(T)$ can be determined from this drop in $\langle K \rangle$. In the normal phase, $\langle k_G^2 \rangle_N = \int dk \, k_G^2n_N(k)$ where $n_N(k)$ is given by (1) with $n_0 = 0$ and $A_1 = 1$. Assuming that the second moment of $n^*(k)$ and $n_N(k)$ are the same, which is found to be the case in bulk liquid $^4$He,\textsuperscript{10} we have $\langle K \rangle / \langle K \rangle_N = \langle k_G^2 \rangle / \langle k_G^2 \rangle_N = A_1$. Combining this value of $A_1$ with the normalization equation for $n(k)$, $n_0[1 + I_f] + A_1 = 1$, we have

$$n_0 = \frac{1 - \langle K \rangle / \langle K \rangle_N}{I_f}. \tag{2}$$

This is the result connecting the condensate fraction, $n_0$, at a given temperature to the kinetic energy $\langle K \rangle$ at that temperature and the kinetic
energy in the normal phase $\langle K \rangle_N$. It is the expression proposed by Sears\textsuperscript{7} with the contribution from $f(k)$ ($I_f = 0.25$) included.

To determine $\langle K \rangle$ we fitted a function $J(Q, y) = J_{IA}(y) \otimes R(Q, y)$ to the data. This fitted $J_{IA}(y)$ does not contain a condensate but has Gaussian, fourth cumulant and sixth cumulant components. The fourth and sixth cumulants of $J_{IA}(y)$ and $R(Q, y)$ were set at their bulk normal liquid $^4$He values taken from Ref. 10 where $R(Q, y)$ was found to be independent of $T$. The parameter that sets the Gaussian component in $J_{IA}(y)$ (the second moment of $J(Q, y)$) was then determined by fitting this $J(Q, y)$ to the data in Vycor.

A typical fit to data is shown in Fig. 2 and the resulting “one-parameter” kinetic energies are shown in Fig. 3. The method was tested by comparing one-parameter $\langle K \rangle$ values in bulk liquid $^4$He with the accurate values found in Ref. 10. The agreement of the two is excellent in normal $^4$He. In the superfluid the approximate one-parameter method gives a marginally lower $\langle K \rangle$ value. The $\langle K \rangle$ values in Vycor and in the bulk are the same within error except perhaps at higher temperatures near and above $T_s$ where the Vycor values may be approximately 1 K higher.

![Fig. 2. J(Q, y) of liquid $^4$He in Vycor showing a fit as described in the text.](image)
The condensate fraction, \( n_0(T) \), of liquid \(^4\text{He}\) in Vycor and in the bulk, obtained from (2) using the \( \langle K \rangle \) values, and accurate values for the bulk from Ref. 10 are shown in Fig. 4. This shows that there is a condensate in superfluid \(^4\text{He}\) in Vycor of magnitude comparable to that in the bulk. The one-parameter values of \( n_0(T) \) in bulk \(^4\text{He}\) are clearly larger than the accurate values.\(^{10}\) We believe this is because fitting a function \( J(Q, y) \) that does not contain a condensate to data containing a condensate generally underestimates \( \langle K \rangle \). This effect was also noted by Mayers \textit{et al.}\(^{24}\)

An underestimate of \( \langle K \rangle \) in the superfluid leads to an overestimate of \( n_0 \) from (2). For example, if we use the accurate bulk values \( \langle K \rangle = 14.45 \pm 0.3 \) K at \( T = 0.5 \) K and \( \langle K \rangle = 16.3 \pm 0.3 \) K at \( T = 2.3 \) K shown in Fig. 3, we obtain \( n_0 = 9.1 \pm 3.0 \)% from (2). This compares with the accurate value of \( n_0 = 7.25 \)% shown in Fig. 4. Also, (2) itself overestimates \( n_0 \) somewhat because the contribution of \( f(k) \) to the \( \langle K \rangle \) is neglected and all the drop in \( \langle K \rangle \) below \( T_c \) is attributed to BEC. Thus, we have determined how much (2) overestimates \( n_0(T) \) using bulk \(^4\text{He}\) as a control. Figure 4 then shows that there is definitely a condensate in Vycor of magnitude comparable to the bulk value.
Fig. 4. Temperature dependence of the condensate fraction, $n_0$. The squares and triangles are the “one-parameter” values obtained from (2) for liquid $^4$He in Vycor and in the bulk, respectively. Circles are accurate values for the bulk from Ref. 10. The dashed and dotted lines are guides to the eye that display the expected temperature dependence of $n_0(T)$ in Vycor and the bulk, respectively.

4. DISCUSSION

Our data shows that there is a condensate in liquid $^4$He in Vycor. As in the bulk, BEC accompanies superfluidity and well defined excitations in Vycor. All measured values of $T_c$ for superfluidity in Vycor\(^{26}\) lie in the range 1.94 < $T_c$ < 2.03 K. The tortuosity, $\chi$, of a porous media may be defined in terms of the fraction \((1-\chi) = \rho_s(T=0)/\rho\) of atoms participating in superfluid flow at zero temperature.\(^{25,15}\) In Vycor, \(^{25}\) $\chi = 0.77 \pm 0.01$. Monte Carlo simulations of hard spheres in a box containing hard cylinder impurities find (using the definition of $\chi$ above) \(0.51 \leq (1-\chi) \leq 0.74\) depending chiefly on the volume excluded by the impurities.\(^{15}\) We observe no corresponding reduction of $n_0(T)$ in Vycor below the bulk value at liquid densities.

At liquid $^4$He densities, we expect\(^{16}\) disorder and confinement to reduce $\rho_s(T)$ more than $n_0(T)$. In the bulk, as a result of the strong mutual interaction and in the absence of static impurities, all the fluid flows
together and \( \rho_s(T=0)/\rho = 1 \). When quenched disorder or static impurities are introduced the \(^4\text{He}\) interacts with the static disorder and will be “held back” by the impurities in a flow measurement reducing \( \rho_s(T=0) \). In contrast, \( n_0(T) \) is already greatly reduced from unity by the interatomic interaction in the bulk, to approximately \( n_0 = 7.25\% \) at \( T = 0 \) K and SVP. The fraction of volume excluded by interatomic interaction can be characterized by the parameter \( n_a^3 = 0.21 \) where \( a = 2.2034 \) Å is the hard sphere diameter of the \(^4\text{He}\) atoms. Disorder and confinement can further reduce \( n_0(T) \) only if it has length scales comparable to the interatomic spacing and excludes volumes comparable or greater than \( n_a^3 \). Thus we expect disorder to reduce \( \rho_s(T) \) more than \( n_0(T) \), which is consistent with analytic expressions valid in the dilute limit.\(^{16}\)

General arguments,\(^{22}\) calculations,\(^{27,16,28}\) and measurements of phonon-roton excitations in Vycor and Geltech silica\(^{19,20}\) suggest that the condensate can be localized by disorder. In this event, when BEC first appears it could be localized in favorable regions in Vycor that are separated by regions of normal fluid. In this circumstance, there would be phase coherence over short length scales only in specific regions. In this situation, superfluidity would not be observed in a torsional oscillator experiment since that requires phase coherence across the whole sample. As the temperature is further reduced, the transition temperature to superfluidity, \( T_s \), may be associated with the joining (percolation) of these localized regions\(^{28}\) so that there is the phase coherence across the entire sample needed for observable superflow. This temperature, \( T_s \), would clearly lie below \( T_{\text{BEC}} \) for BEC in favorable regions. The state between \( T_s \) and \( T_{\text{BEC}} \) may be referred to as a “mixed” or “Bose glass” state\(^{29}\) in which there is localized BEC but no superflow. Precise determination of \( T_{\text{BEC}} \) is a clear goal of future experiments.

Herb and Dash\(^{30}\) have reported “superfluid onset” at temperatures higher than \( T_s \) in \(^4\text{He}\) films. While they observe superfluid onset at the usual \( T_s \), they also observe a superfluid onset at higher temperatures, within 20 mK of \( T_s \) in some cases. It is not clear whether this superflow above \( T_s \) in films is related to our suggestion of localized BEC above \( T_s \) in disordered 3D media or not. However it is an example of “mixed” phenomena above \( T_s \). Similarly, Thibault \textit{et al.}\(^{31}\) report a fountain pressure in aerogel above \( T_s \). This suggests local regions of superfluidity above \( T_s \) in aerogel.

In liquid \(^3\text{He}\) in aerogel, Bunkov \textit{et al.}\(^{32}\) report evidence for coherence in the spin dynamics of \(^3\text{He}\), of a type associated with superfluidity, at temperatures above \( T_s \). At temperatures between \( T_s \) in aerogel and \( T_c \) in bulk liquid \(^3\text{He}\), as determined in a torsional oscillator experiment for example, liquid \(^3\text{He}\) in aerogel shows evidence of coherence or broken symmetry in NMR properties. In this temperature range, liquid \(^3\text{He}\) in aerogel is not in a simple normal phase.
In summary our results show that there is definitely BEC in superfluid $^4$He in Vycor. Within precision, the magnitude of the condensate fraction $n_0(T)$ is the same in Vycor and in the bulk, as is its temperature dependence. We have not been able to determine whether the critical temperature for BEC lies above or is the same as that for superfluid flow.

ACKNOWLEDGMENTS

We thank P. S. Danielson at Corning Inc. for providing the isotopic Vycor sample, W. G. Stirling for scientific support, Oleg Petrenko for assistance on the MARI spectrometer, the cryogenic group at ISIS for technical support, and A. D. Taylor for allocating director’s discretionary time for the experiment. This work was partially supported by the National Science Foundation Grant DMR 0115663.

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