Dynamics of liquid $^4$He in Vycor

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We have measured the dynamic structure factor of liquid $^4$He in Vycor using neutron inelastic scattering. Well-defined phonon-roton (p-r) excitations are observed in the superfluid phase for all wave vectors $0 \leq Q \leq 2.15$ Å$^{-1}$. The p-r energies and lifetimes at low temperature ($T = 0.5$ K) and their temperature dependence are the same as in bulk liquid $^4$He. However, the weight of the single p-r component does not scale with the superfluid fraction $\rho_s(T)/\rho$ as it does in the bulk. In particular, we observe a p-r excitation between $T = 1.952$ K, where $\rho_s(T) = 0$, and $T = 2.172$ K of the bulk. This suggests, if the p-r excitation intensity scales with the Bose condensate, that there is a separation of the Bose-Einstein condensation temperature and the superfluid transition temperature $T_c$ of $^4$He in Vycor. We also observe a two-dimensional layer mode near the roton wave vector. Its dispersion is consistent with specific heat and $\rho_s(T)$ measurements and with layer modes observed on graphite surfaces.

Liquid $^4$He immersed in Vycor is a readily accessible example of bosons in disorder and confinement. The specific heat, superfluid density and other thermodynamic properties of this "dirty Bose system" have been extensively investigated\cite{1,2,3} to reveal the impact of disorder and finite length scales on excitations and phase transitions\cite{4}. Understanding gained in helium can be transferred to other examples of bosons in disorder such as flux lines in superconductors\cite{5}, granular metal films\cite{6}, Cooper pairs in Josephson junction arrays\cite{7} and (possibly) Cooper pairs in high-$T_c$ materials if pairing occurs above the Bose-Einstein condensation (BEC) temperature that leads to superconductivity\cite{8}. Direct measurement\cite{1,2,3} and simulation\cite{4,8} of excitations of liquid $^4$He in disorder, however, have only recently begun. In this Letter, we present neutron scattering measurements of the dynamic structure factor $S(Q,\omega)$ of liquid $^4$He in 30% porous Vycor over a wide wave-vector range, $0.3 \leq Q \leq 2.15$ Å$^{-1}$, and temperature range, $0.5 \leq T \leq 2.31$ K. We present the first observation of phonons in Vycor, evidence for a two-dimensional (2D) layer mode, and evidence that the superfluid transition temperature $T_c$ and the BEC temperature of liquid $^4$He in Vycor may not be the same.

The present Vycor sample, a cylinder of 9.7 mm diameter and 40 mm height, was synthesized in the usual way except that natural boron was replaced by $^{11}$B (99.95% purity). Natural boron has a large absorption cross-section for neutrons while $^{11}$B does not. Small-angle neutron scattering measurements on the present sample showed that it has the same static structure factor $S(Q)$ as standard Vycor plates made with natural boron\cite{9}. The sample was fitted into a tightly machined cylindrical aluminum sample holder of 1.5 mm wall thickness and 100 mm height. The sample was fully filled with helium with a compartment of bulk $^4$He above the Vycor for reference measurements. The Vycor and bulk $^4$He compartments were separated by a Cd spacer. The sample cell was mounted in a $^3$He cryostat, where the thermometers were calibrated against the $^3$He vapor pressure in the sample cell in a separate run. The temperature was regulated within $\pm 0.02$ K. The measurements were made on the IN6 time-of-flight spectrometer at the Institut Laue Langevin, using an incident neutron energy of 3.83 meV and an energy resolution (FWHM) of about 110 $\mu$eV.

Figure 1(a) shows $S(Q,\omega)$ of liquid $^4$He in Vycor for a wave vector $Q$ in the phonon region, $Q = 0.35$ Å$^{-1}$. At $T = 0.5$ K, $S(Q,\omega)$ is confined almost entirely to a single peak arising from creation of single phonons in the liquid by the neutrons. Multiphonon creation is small at these wave vectors. The peak width is set by the instrument resolution width. As $T$ is increased to $T = 2.31$ K, the single phonon peak broadens, but there is still a well defined peak in the normal phase. This shows that liquid $^4$He in Vycor supports a well defined sound mode in both the superfluid ($T = 0.5$ K) and normal ($T = 2.31$ K) phases.
K) phase, as in bulk liquid $^4$He. This is the first direct observation of phonons in Vycor by neutron scattering.

Figure 1(b) shows $S(Q, \omega)$ at several temperatures for $Q = 1.7$ Å$^{-1}$, a wave vector between the maxon $(Q = 1.1$ Å$^{-1})$ and the roton $(Q = 1.95$ Å$^{-1})$ regions. There is a well defined, single phonon-roton (p-r) excitation in $S(Q, \omega)$ at the lowest temperature, $T = 0.5$ K. As $T$ increases, the p-r peak broadens and the integrated intensity in the peak decreases. At $T = 2.31$ K, there is no discernible peak in $S(Q, \omega)$. However, at $T = 1.99$ K, which is above $T_c = 1.952$ K [7] (where $\rho_S = 0$) but below the bulk value for $T_\lambda = 2.172$ K, there is still a thermally broadened peak. Woods and Svensson [18] proposed that for wave vectors at the maxon and higher $(Q \gtrsim 1.1$ Å$^{-1})$, the weight of the characteristic maxon-roton excitation in $S(Q, \omega)$ of bulk liquid $^4$He scaled as the superfluid fraction, $\rho_S(T)/\rho$. There is no mode for $Q \gtrsim 1.0$ Å$^{-1}$ in normal bulk liquid $^4$He where $\rho_S(T) = 0$. We return to this point at the end of this Letter.

Figure 2(a) shows $S(Q, \omega)$ at the roton wave vector $Q = 1.95$ Å$^{-1}$ and $T = 0.5$ K. The intense peak at $\omega = 0.74$ meV arises from exciting the p-r mode at the roton wave vector. We call this the 3D roton. The p-r energies $\omega_Q$ for $0.3 \leq Q \leq 2.15$ Å$^{-1}$, obtained from $S(Q, \omega)$ at many $Q$ values such as shown in Figs. 1 and 2(a), are displayed in Fig. 3. The phonon-roton energies in Vycor are the same as in the bulk (perhaps marginally lower for $Q \gtrsim 1.9$ Å$^{-1}$), within the present experimental precision $(\pm 5\mu$V).

Returning to Fig. 2(a), we see that there is additional intensity at energies below the 3D roton peak in Vycor that is not seen in bulk liquid $^4$He [4,21]. This additional intensity is small, approximately 8% of the main 3D p-r integrated intensity at the roton. We emphasize that $S(Q, \omega)$ of liquid $^4$He in both Vycor and bulk shown in Fig. 2(a) are from the present measurements. We observe the additional intensity in Vycor for $Q \gtrsim 1.7$ Å$^{-1}$ only. The intensity of the new mode is shown in Fig. 2(b) with its energy dispersion as an insert. For $Q \lesssim 1.7$ Å$^{-1}$, the intensity in the new mode either becomes too weak to be observed, or the mode energy lies sufficiently close to the 3D p-r mode that it cannot be resolved from the p-r peak.

Since all measurements were made on fully filled Vycor, we cannot identify the region of the liquid from which the additional scattering originates. However, we interpret the additional intensity as a 2D layer mode propagating in the liquid layers adjacent to the two solid $^4$He layers on the Vycor surfaces. The layer mode energy near its “roton” minimum is well described by $\omega(Q) = \Delta_{2D} + (Q - Q_{2D})^2/2\mu$ with $\Delta_{2D} = 0.55 \pm 0.01$ meV, $Q_{2D} = 1.94 \pm 0.01$ Å$^{-1}$ and $\mu = 0.13 \pm 0.01$ $m_4$ ($m_4$ is the $^4$He atomic mass). The present “gap energy” $\Delta_{2D}$ is consistent with the gap energy $0.54 \pm 0.03$ meV of 2D rotons on graphite surfaces observed by Tomlinson et al. [21] and with that of 0.6 meV observed by Lauter et al. [22] on graphite surfaces. The difference between the 3D roton energy $\Delta = 0.742$ meV and $\Delta_{2D}$ is consistent with the differences predicted originally by Padmore [23] for 2D rotons and calculated more recently by Clements et al. [24]. The calculations by Clements et al. also suggest that the layer mode intensity is small at lower $Q$ as found here. The present gap energy $\Delta_{2D} = 0.55 \pm 0.01$ meV is consistent with the gap energy of 0.53 meV obtained by Brewer et al. [25] for the layer mode contribution to the specific heat in Vycor. Kiewiet et al. [26] found that the superfluid density $\rho_S(T)$ in Vycor for $T \lesssim 1.4$ K is well described if the normal density arises from exciting “one-dimensional” phonons and a roton-like mode having a roton gap of 0.50 meV. This interpretation is consistent with the phonons observed here, which will propagate predominantly along the pores, and with the present 2D layer mode. For $T \lesssim 1.4$ K, the 3D roton energy $\Delta = 8.62$ K is too high for 3D rotons to be excited. Thus, the interpretation of Kiewiet et al. [26] is consistent with the phonons and the 2D layer mode that we observe here. The additional intensity shown in Fig. 2(a) is similar to that observed by Dimeo et al. [3] at an energy of 0.3-0.5 meV at the roton wave vector, although their intensity is two times greater (20% of the 3D roton). However, they could not determine the mode energy with precision.

To determine how the weight of the p-r mode component in $S(Q, \omega)$ scales with temperature, we have fitted the following model to the data,

$$\chi''(Q, \omega) = f_S(T)\chi''_{g}(Q, \omega) + f_N(T)\chi''_{N}(Q, \omega),$$

where $S(Q, \omega) = [1 - \exp(-\hbar\omega/k_BT)]^{-1}\chi''(Q, \omega)$. At $T = 0.5$ K, $\chi''_{g}(Q, \omega)$ is the total observed $\chi''(Q, \omega)$, corresponding predominantly to the single p-r peak [see Fig. 1(b)]. $\chi''_{N}(Q, \omega)$ is the total $\chi''(Q, \omega)$ observed at $T = 2.31$ K, which contains no p-r peak at all. The $f_S(T)$ is a free parameter that we obtain by fitting Eq. (1) to $\chi''(Q, \omega)$ observed at temperatures between $T = 0.5$ and 2.31 K, and $f_N(T) = 1 - f_S(T)$. In the fit the p-r mode energy and width in $\chi''_{N}(Q, \omega)$ was allowed to vary with temperature. Clearly, the model requires $f_S(T) = 1$ at $T = 0.5$ K and $f_S(T) = 0$ at $T = 2.31$ K. The fitted fraction $f_S(T)$ represents, approximately, the fraction of the total $S(Q, \omega)$ taken up by the single excitation peak.

Figure 4 shows the fitted values of $f_S(T)$ compared with the superfluid fraction $\rho_S(T)/\rho$ in Vycor and in bulk liquid $^4$He. The fraction $f_S(T)$ in Vycor clearly does not scale with $\rho_S(T)/\rho$ in Vycor. At low temperatures, both $f_S(T)$ and $\rho_S/\rho$ are approximately unity, while at higher $T$, $f_S(T)$ is still large whereas $\rho_S(T)/\rho \approx 0$. In searching for possible causes, we note that there is some bulk liquid $^4$He between the Vycor sample and the sample cell walls. The fraction of such bulk liquid to liquid in Vycor pores is estimated to be at most 10%. This small fraction could not account for the large deviation of $f_S(T)$ from $\rho_S(T)/\rho$ in Vycor. We would essentially need all of the liquid to be bulk liquid to explain the scaling in Fig. 4. Also, since $\chi''_{N}(Q, \omega)$ is expected to be largely independent of $T$ for $Q > T_\lambda$ (as it is in bulk liquid $^4$He), the
fitted \( f_S(T) \) should not be sensitive to the temperature at which \( \chi^\prime\prime_N(Q, \omega) \) is defined.

The central finding is therefore that the weight of the single phonon-roton excitation peak in \( S(Q, \omega) \) at higher \( Q \) values does not scale with \( \rho_S(T)/\rho \) in Vycor. The deviation is large and there remains a p-r peak in \( S(Q, \omega) \) above \( T_c \) where \( \rho_S(T) = 0 \). Thus the apparent scaling of peak weight with \( \rho_S(T) \) in bulk \(^4\)He is not universal and does not extend to confined geometries. As noted, Glyde and Griffin (GG)\(^{29}\) proposed that the sharp excitation in \( S(Q, \omega) \) at \( Q \gtrsim 1 \AA^{-1} \) arises because there is a condensate and that the weight should scale approximately as the condensate fraction, \( n_0(T) \). The excitation weight in \( S(Q, \omega) \) in Vycor might still scale with \( n_0(T) \) (as in bulk \(^4\)He) if \( n_0(T) \) in Vycor were similar to bulk \(^4\)He and particularly if \( n_0(T) \) were finite between \( T_c = 1.952 \) K and \( T_\lambda = 2.172 \) K. We therefore arrive at the interesting conclusion: either the GG proposal is incorrect or there is a (possibly localized) condensate in \(^4\)He in Vycor between \( T_c \) and \( T_\lambda \). If the latter is true, then we are observing the separation of the BEC temperature from the superfluid transition temperature (\( T_\nu \)) by disorder or confinement\(^{23}\). This intriguing possibility needs to be clarified by further measurements of \( S(Q, \omega) \) between \( T_c \) and \( T_\lambda \) and direct measurements of \( n_0 \).

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FIG. 1. Dynamic structure factor of liquid $^4$He in Vycor at temperatures as shown. (a) $Q = 0.35$ Å$^{-1}$ (phonon region). The lines are guides to the eye. (b) $Q = 1.7$ Å$^{-1}$ (between the maxon and roton regions). Symbols are $^4$He in Vycor, with errors smaller than the symbol size. The lines show bulk data for comparison.
FIG. 2. (a) $S(Q, \omega)$ of liquid $^4$He at $T = 0.5$ K in Vycor at the roton wave vector (solid circles and line). The dashed line is the component of $S(Q, \omega)$ arising from exciting the 3D roton in the liquid (peak height $H = 8.0$ meV$^{-1}$). The intensity at energies below the roton peak is attributed to a 2D layer mode propagating in the liquid layers adjacent to the Vycor walls. The open circles are the corresponding $S(Q, \omega)$ observed in bulk $^4$He (present measurements). (b) Integrated intensity in the 2D layer mode versus wave vector. At $Q = 1.95$ Å$^{-1}$, the integrated intensity of the 2D mode is 8% of the 3D roton intensity in fully filled Vycor. The inset shows the energies of the 2D layer mode (solid circles and line) and the 3D roton (open circles) in Vycor as well as the 3D bulk roton energy (dotted line).
FIG. 3. Phonon-roton energy dispersion curve of liquid $^4$He in Vycor (open circles) and in bulk $^4$He (line). The error bars are much smaller than the symbol size.

FIG. 4. Fraction $f_S(T)$ of the total $S(Q, \omega)$ that is taken up by the low-temperature component $S_S(Q, \omega)$ (chiefly the single-excitation component) as a function of temperature for Vycor [see Eq. (1)]. The $f_S(T)$ in Vycor does not scale with the superfluid fraction $\rho_S(T)/\rho$ of Vycor (dotted line; Ref. [1]). The dashed line is $\rho_S(T)/\rho$ for bulk $^4$He [1].