Excitations of Metastable Superfluid ^{4}He at Pressures Up to 40 Bars

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Neutron scattering measurements of the fundamental excitations of liquid ^{4}He confined in 44 Å pore diameter gelsil glass at pressures up to 40 bars in the wave vector range $0.4 < Q < 2.3$ Å^{-1} are reported. Above 25.3 bars and at low temperature ($T = 0.4$ K) the characteristic phonon-roton mode of superfluid ^{4}He is no longer observed as a well-defined mode in the phonon-maxon region ($0.4 < Q < 1.6$ Å^{-1}). Modes at wave vectors $Q > 1.6$ Å^{-1}, especially the rotons, are observed up to complete solidification of all the liquid at a pressure of $\sim$40 bars where the roton vanishes. At and above a pressure of 35.1 bars, Bragg peaks are observed, indicating coexistence of liquid and solid in the pores at pressures $35 \lesssim P \lesssim 40$ bars.

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We present the first measurements of the phonon-roton (PR) excitations of superfluid ^{4}He at pressures p above the bulk liquid-solid freezing pressure, $p = 25.3$ bars. The liquid is confined in a porous media, a 44 Å pore diameter gelsil (Geltech), in which ^{4}He remains liquid up to $p \sim$ 40 bars before freezing is complete. We compare our data with measurements of the superfluid-normal liquid and liquid-solid phase boundaries made using ultrasonic methods in the same gelsil sample by Daughton and Mulders [1] (see Fig. 1).

Liquid ^{4}He in porous media (PM) is an ideal disordered Bose system in which Bose-Einstein condensation (BEC), excitations, and superfluidity are all modified and their interdependence can be explored. In PM, the superfluid-normal transition temperature T_c is suppressed to temperatures below the bulk value T_λ [2–4] [e.g., $T_\lambda =$ 2.17 K and $T_c = 1.92$ K in the present gelsil at saturated vapor pressure (SVP)]. In addition, both T_c in PM and T_λ in the bulk are further lowered if pressure is applied (see Fig. 1). Also in PM, as a result of confinement, the liquid phase exists up to higher pressures before freezing (up to 35–45 bars depending on the pore diameter and structure of the PM [5,6]). For example, $T_c = 1.25$ K at $p =$ 43 bars in Vycor [2] (70 Å diameter pores). Yamamoto *et al.* have recently reported that T_c goes to zero at $p \sim$ 35 bars in 25 Å pore diameter gelsil glass [7]. Importantly, this means there is a normal liquid phase between the superfluid and the solid phase at low T in 25 Å gelsil, suggesting that there is a "quantum" superfluid-normal transition or quantum phase transition (QPT).

In Landau's theory of superfluidity, superflow exists because superfluid ^{4}He supports only sharply defined PR modes [8]. Specifically, there can be no low-lying excitations to which a moving superfluid can decay and lose energy. The PR excitations of superfluid ^{4}He have this character in the bulk and at SVP in PM [9–11]. Thermal excitation of the PR modes constitutes the normal fluid. It is not known how this picture translates to higher p where T_c is very low. In this region, the normal component may arise from quantum fluctuations. The loss of superfluidity may be related to loss of well-defined PR excitations.

In an alternate picture, superfluidity follows directly from the phase coherence of the condensate. The existence of well-defined PR modes and particularly the absence of low-lying single particle modes are also attributed to the existence of a condensate [12,13]. The condensate fraction, n_0, is predicted to drop from 7.25%

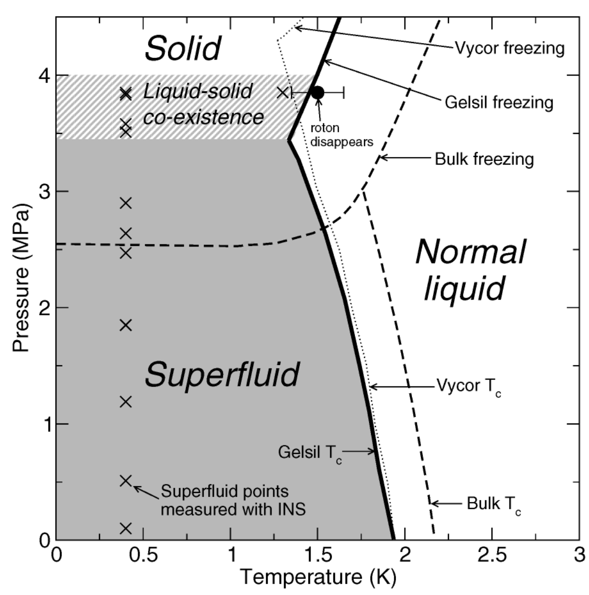

FIG. 1. Phase diagram of ^{4}He confined in 44 Å porous gelsil. Dashed line is the bulk system, bold solid line is ^{4}He confined in the current gelsil sample measured using ultrasonic techniques [1]. Dotted line is ^{4}He in Vycor from Ref. [2]. The liquid-solid coexistence region (gray stripes) shows that freezing occurs over a p range of $\sim$5.5 bars, reflecting the distribution of pore sizes. The filled circle is the temperature at which the roton disappears at 38.5 bars. Crosses show the superfluid points measured with neutrons.

at SVP [14] to 2% at 25 bars and to less than 1% at 40 bars [15,16]. It is not known whether the above ideas all remain true in the metastable liquid when n_0 becomes extremely small [17,18], and whether effects of a very small condensate can be observed in the PR excitations. Also, in PM, the "normal" liquid above T_c but below T_λ has been shown to contain localized BEC (local phase coherence) [10,11,19].

For these reasons, we investigated PR excitations in 44 Å gelsil where modes are readily observed at SVP and T_c lies substantially below T_λ ($T_c \sim 1.3$ K at 35–40 bars). We find indeed that the PR mode at wave vectors $0.4 < Q < 1.6$ Å^{-1} disappears at pressures above 25.3 bars. Modes are observed at higher wave vectors ($Q > 1.6$ Å^{-1}) only (see Fig. 2). Since the liquid is still superfluid, the dynamic response is apparently at higher energy beyond the range observed here (up to 4 meV), with no low-lying excitations. At low temperature ($T = 0.4$ K), the roton is clearly observable at all pressures up to 38.5 bars. At and above $p = 35.1$ bars, in addition to the liquid signal, Bragg peaks characteristic of a hcp crystalline powder structure are observed. Solidification therefore takes place over a p range ~35–40 bars, reflecting the pore size distribution of the gelsil. The roton completely disappears at total solidification ($p \sim 40$ bars). At the liquid-solid transition, the roton energy is finite (~0.55 meV) with no evidence of "soft mode" behavior.

The measurement was performed on the IN6 time-of-flight spectrometer at the Institut Laue-Langevin, France. Incident wavelengths of 4.62 Å and 4.17 Å were used, the latter measured using Bragg scattering from an α-quartz crystalline powder. The gelsil, manufactured by 4F International Co., consists of two monolithic cylinders placed one on top of the other in a cylindrical Al sample cell, connected to an external gas handling system by a capillary and cooled with a continuously circulating ^{3}He cryostat to a temperature of $T = 0.40 \pm 0.02$ K.

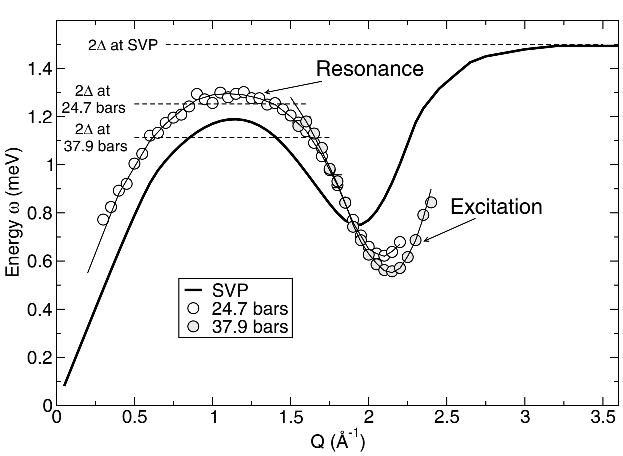

FIG. 2. Dispersion of the PR excitations, showing the change in the dispersion as a function of p. Δ is the roton energy. Solid lines through points at 24.7 and 37.9 bars are guides to the eye.

We have measured $S(Q, \omega)$ at the pressures indicated in the phase diagram in Fig. 1. For $p < 25.3$ bars, liquid exists in the gelsil pores and in the volume between the gelsil and the cell wall. At $p < 25.3$ bars, p can be measured accurately with an external pressure transducer. For $p > 25.3$ bars, helium freezes in the capillary so we used the "blocked capillary method" to acquire the metastable state. The cell and cryostat were warmed to $T = 3$ K, and the pressure of the ^{4}He was raised to a value estimated to lead to the desired pressure at $T = 0.4$ K by consulting the phase diagram. The system was allowed to equilibrate in this state for ten minutes. The cell was then cooled, while keeping the capillary warmer than the cell to delay freezing of ^{4}He in the capillary for as long as possible. Once the capillary blocks, the volume and amount of helium in the cell are constant. Essentially we navigate along the isopycnal line. As T is reduced, bulk ^{4}He in the empty volume surrounding the gelsil solidifies, but the ^{4}He within the pores remains liquid.

Since the capillary is blocked, an alternative measure of p is required. At all $p < 38.5$ bars, we observe rotons in the liquid in the gelsil. Starting at $p = 35.1$ bars, we also see Bragg peaks (Fig. 3), indicating that a solid with a hcp structure begins to form in the pores. This solid exhibits the expected excitations [20,21]. The angle of the Bragg peaks, and their change as a function of p, provide a direct measure of the lattice spacings, which gives the density of the solid and therefore the applied pressure on the liquid [22]. The coexistence of solid and liquid in the pores suggests that solidification takes place over a p range reflecting the pore size distribution of the gelsil.

While the pressures obtained from the Bragg peaks are reliable, they are not sufficiently accurate (±4 bars). We therefore determined the pressure (up to 38.5 bars) from the roton energy by extrapolating the observed energies from SVP to 25 bars to high pressures and placing our observed roton energies on the extrapolation to obtain the pressure (see Fig. 4). At higher pressures, the roton disappears so that at $p \sim 40$ bars, only solid remains. At $p > 40$ bars, in addition to the Bragg peaks, we see a broad peak in the diffraction spectrum at higher angles indicative of an amorphous solid, perhaps located in the smallest pores.

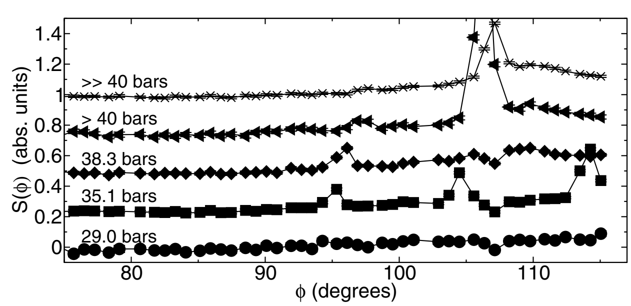

FIG. 3. Diffraction spectra [$S(\phi)$ as a function of angle ϕ] showing the variation of the Bragg peak positions of the solid helium in the pores as a function of p.

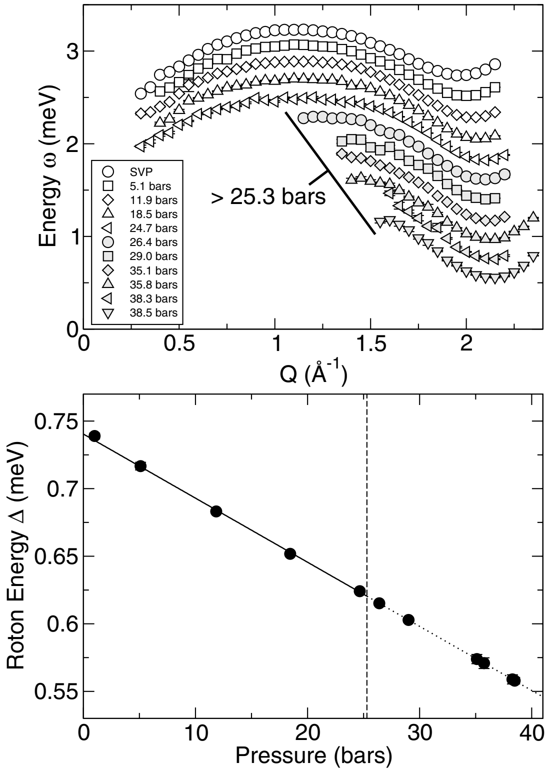

FIG. 4. Top: p dependence of the dispersion of the excitations. Open symbols, liquid ^{4}He at $p < 25.3$ bars; shaded symbols, metastable liquid ^{4}He. Data at each p is offset by 0.2 meV for clarity. Bottom: p dependence of the roton energy Δ. Here, an extrapolation of Δ at $p < 25.3$ bars (solid line) has been used to calibrate the pressure above 25.3 bars (dotted line). Dashed line shows the solidification pressure.

The p dependence of $S(Q, \omega)$ is shown in Fig. 5. The intensity of the roton excitation increases with pressure up to 25.3 bars where solidification of the bulk liquid between the gelsil and the cell wall occurs. The intensity then drops since the amount of liquid has decreased. The intensity in the phonon mode ($Q = 0.7$ Å^{-1}) disappears quite abruptly at between $p = 24.7$ and 26.4 bars. Throughout the entire metastable p range the roton ($1.6 < Q < 2.2$ Å^{-1}) is the only excitation visible. Figure 4 shows the dispersion of the observable excitations. Sharp PR modes are observed at higher wave vectors ($Q > 1.6$ Å^{-1}) only.

The roton energy at each pressure was determined by making a parabolic fit of the Landau expression $\omega_Q = \Delta + \hbar^2((Q - Q_R)^2)/2\mu_R$ [8] around the roton minimum, where Q_R is the roton wave vector and μ_R is its effective mass. The resulting p dependence of Δ is shown in Fig. 4. The lowest value we have measured is $\Delta = 0.558 \pm 0.006$ meV at $p = 38.5$ bars. Above this pressure, the roton becomes too weak to model reliably. We have also measured the temperature dependence of the excitations at $p = 38.5$ bars, at temperatures $T = 0.4$, 1.3, and 1.5 K (see Fig. 6). The roton is substantially weakened at $T =$ 1.3 K, and at $T = 1.5$ K, the roton is not observed. Since a well-defined mode exists where there is BEC [10,11,19], this indicates BEC at $T = 0.4$ K but none at $T \gtrsim 1.3$ K at 38.5 bars.

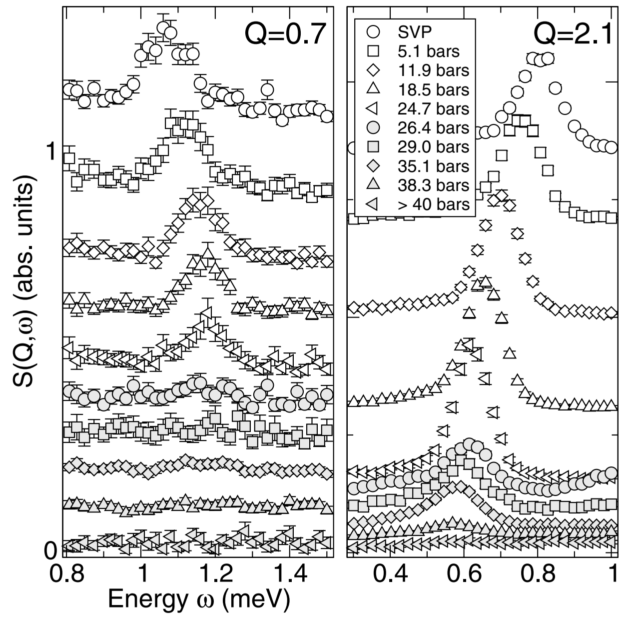

FIG. 5. p dependence of $S(Q, \omega)$ of ^{4}He confined in gelsil, at a phonon Q (left) and the roton Q (right). Open symbols, $p <$ 25.3 bars; shaded symbols: $p > 25.3$ bars. At $p = 26.4$ bars there is a phonon excitation at energy >1 meV arising from a single crystal formed between the gelsil slabs and the cell wall.

Three processes can contribute to the loss of a sharply defined, single PR mode for $0.4 < Q < 1.6$ Å^{-1} at $p >$ 25 bars. The first is transfer of intensity from the mode to the two PR band. The total single PR response function consists of the sharp PR mode plus a resonance in the two PR band, as in all systems where there is one-two excitation interaction [23–25]. When the single mode energy ω_Q lies well below the two PR band ($\omega_Q \ll 2\Delta$), the intensity is chiefly in the sharp mode. The two PR band starts at 2Δ and ω_Q cannot exceed 2Δ. As ω_Q approaches 2Δ, weight is transferred from the mode to the resonance. For example, at SVP, as Q increases beyond $Q \sim 2.8$ Å^{-1} [where $\omega_Q = 2\Delta$ (see Fig. 2)], weight is increasingly transferred from the mode to the resonance until the

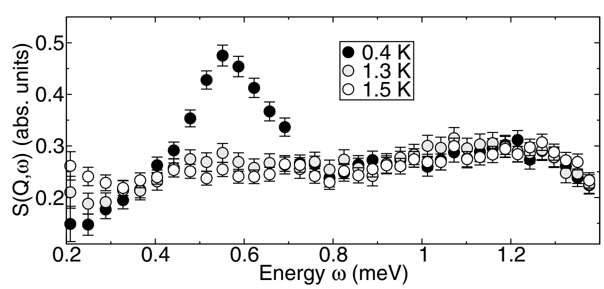

FIG. 6. Temperature dependence of the roton excitation at $p = 38.5$ bars, showing its disappearance at around 1.5 K.

mode disappears entirely at 3.6 Å^{-1}, the end point of the PR mode spectrum [26–28]. As p increases, Δ decreases and the maxon energy ω_Q increases. At 20 bars, the maxon $\omega_Q \simeq 2\Delta$. As p increases, intensity is transferred from the mode to the resonance until at $p \sim 25$ bars, only the resonance is observed [25] (see Fig. 2). Above 25 bars, only very broad intensity (a broad resonance) is observed around the maxon region. A sharp mode is observed again only for $Q > 1.6$ Å^{-1} where ω_Q lies below 2Δ (see Fig. 2). Second, there are one to two PR interference terms in $S(Q, \omega)$ [29–32]. These terms also transfer intensity from the single, sharp mode to the two PR band in the maxon region. However, they change sign and transfer intensity from the two PR band to the sharp, single mode in the roton region. This process appears to contribute since the intensity in the single roton mode increases with increasing p. Third, for PR modes with high energy (close to 2Δ), the total mode intensity of both the sharp component and the component in the two PR band, appears to be proportional to the condensate fraction [26,28]. At higher p, n_0 becomes very small and we expect reduced PR intensity when $\omega_Q \simeq 2\Delta$. These major changes in the PR mode may eventually be responsible for the loss of superfluidity at high enough pressure.

Metastable liquid ^{4}He at high pressures is currently a topic of great interest in the general study of metastable liquids [33]. Werner *et al.* [34] have reported pressurizing liquid ^{4}He up to 164 bars using compression by ultrasound. Nozières [35] has predicted that the condensate fraction will vanish eventually in the high pressure liquid and that there will therefore be a normal liquid phase before crystallization takes place. At present, immersion of the liquid in porous media is the only means of creating metastable helium with a long enough lifetime for study by neutron scattering. In future experiments, we plan to investigate the excitations in smaller pore diameter gelsil where T_c will be lower.

In summary, at pressures above 25.3 bars, the PR dispersion curve of superfluid ^{4}He in 44 Å gelsil disappears from $S(Q, \omega)$ as a well-defined mode in the phonon-maxon region, $0.4 < Q < 1.6$ Å^{-1}. The superfluid continues to support a sound mode at long wavelengths and a well-defined PR mode is observed for $Q > 1.6$ Å^{-1}, particularly at the roton Q. The roton is observed at all pressures up to solidification. No roton is observed in the solid phase. The existence of a well-defined roton at low T suggests the existence of BEC and superfluidity in 44 Å gelsil up to the solidification pressure. As temperature is increased at $p = 38.5$ bars, the roton disappears at $\sim$1.5 K, suggesting the disappearance of BEC (and superfluidity) consistent with the measured phase diagram in Fig. 1.

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[1] D. R. Daughton, Ph.D. thesis, University of Delaware, 2003.
[2] L. Cao, D. F. Brewer, C. Girit, E. N. Smith, and J. D. Reppy, Phys. Rev. B **33**, 106 (1986).
[3] M. H. W. Chan, K. I. Blum, S. Q. Murphy, G. K. S. Wong, and J. D. Reppy, Phys. Rev. Lett. **61**, 1950 (1988).
[4] J. Reppy, J. Low Temp. Phys. **87**, 205 (1992).
[5] D. N. Bittner and E. D. Adams, J. Low Temp. Phys. **97**, 519 (1994).
[6] E. B. Molz and J. R. Beamish, J. Low Temp. Phys. **101**, 1055 (1995).
[7] H. Yamamoto, H. Nakashima, Y. Shibayama, and K. Shirahama, Phys. Rev. Lett. **93**, 075302 (2004).
[8] L. Landau, Zh. Eksp. Teor. Fiz. **19**, 636 (1949).
[9] F. Albergamo, J. Bossy, P. Averbuch, H. Schober, and H. R. Glyde, Phys. Rev. Lett. **92**, 235301 (2004).
[10] O. Plantevin, H. R. Glyde, B. Fåk, J. Bossy *et al.*, Phys. Rev. B **65**, 224505 (2002).
[11] O. Plantevin, B. Fåk, H. R. Glyde, N. Mulders *et al.*, Phys. Rev. B **63**, 224508 (2001).
[12] N. N. Bogoliubov, J. Phys. (USSR) **11**, 23 (1947).
[13] J. Gavoret and P. Nozières, Ann. Phys. (N.Y.) **28**, 349 (1964).
[14] H. R. Glyde, R. T. Azuah, and W. G. Stirling, Phys. Rev. B **62**, 14337 (2000).
[15] S. Moroni and M. Boninsegni, J. Low Temp. Phys. **136**, 129 (2004).
[16] J. Boronat and J. Casulleras, Phys. Rev. B **49**, 8920 (1994).
[17] Y. A. Nepomnyashchii and E. A. Pashitskii, Sov. Phys. JETP **71**, 98 (1990).
[18] E. A. Pashitskii, S. V. Mashkevich, and S. I. Vilchynskyy, Phys. Rev. Lett. **89**, 075301 (2002).
[19] H. R. Glyde, O. Plantevin, B. Fåk, G. Coddens *et al.*, Phys. Rev. Lett. **84**, 2646 (2001).
[20] R. A. Reese, S. K. Sinha, T. O. Brun, and C. R. Tilford, Phys. Rev. A **3**, 1688 (1971).
[21] E. B. Osgood, V. J. Minkiewicz, T. A. Kitchens, and G. Shirane, Phys. Rev. A **5**, 1537 (1972).
[22] J. S. Dugdale and J. P. Franck, Philos. Trans. R. Soc. London A **257**, 1 (1964).
[23] L. Pitaevskii, Sov. Phys. JETP **9**, 830 (1959).
[24] A. Zawadowski, J. Ruvalds, and J. Solana, Phys. Rev. A **5**, 399 (1972).
[25] J. V. Pearce and H. R. Glyde, J. Low Temp. Phys. (to be published).
[26] B. Fåk and J. Bossy, J. Low Temp. Phys. **112**, 1 (1998).
[27] J. V. Pearce, R. T. Azuah, B. Fåk, A. R. Sakhel *et al.*, J. Phys. Condens. Matter **13**, 4421 (2001).
[28] A. R. Sakhel and H. R. Glyde, Phys. Rev. B (to be published).
[29] H. Horner, Phys. Rev. Lett. **29**, 556 (1972).
[30] E. C. Svensson, P. Martel, V. F. Sears, and A. D. B. Woods, Can. J. Phys. **54**, 2178 (1976).
[31] N. R. Werthamer, Phys. Rev. Lett. **28**, 1102 (1972).
[32] H. R. Glyde, Can. J. Phys. **52**, 2281 (1974)
[33] S. Balibar and F. Caupin, J. Phys. Condens. Matter **15**, S75 (2003).
[34] F. Werner, G. Beaume, A. Hobeika, S. Nascimbène *et al.*, J. Low Temp. Phys. **136**, 93 (2004).
[35] P. Nozières, J. Low Temp. Phys. (to be published).