

## Phonon-Roton Modes and Localized Bose-Einstein Condensation in Liquid Helium under Pressure in Nanoporous Media

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We show, using inelastic neutron scattering, that liquid helium in porous media, two gelsils and MCM-41, supports a phonon-roton mode up to a pressure of 36–37 bars only. Modes having the highest energy (“maxons”) broaden and become unobservable at the lowest pressures ( $p \approx 26$  bars) while rotons survive to the highest pressure. By comparing with the superfluid density observed by Yamamoto and co-workers in gelsil, we propose that there is a Bose glass phase containing islands of BEC surrounding the superfluid phase.

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The localization of Bose-Einstein condensation (BEC) and more complex ordered states to isolated islands by disorder is of great current interest in condensed matter physics [1–8]. The onset of this localization is usually associated with a superfluid-normal liquid or a superconducting-insulator transition. An example is Josephson junction arrays [9,10]. In the superconducting state there is connected BEC and phase coherence extended across the whole array. The superconductor-insulator (S-I) transition is associated with a crossover from extended phase coherence to BEC localized to the individual superconducting elements each having an independent phase. Similarly, in granular metal films [11] a S-I transition takes place when there is sufficient disorder to break up the superconducting state into isolated superconducting islands. In high temperature superconductors, recent scanning tunneling microscope measurements have revealed isolated islands of material having an energy gap characteristic of the superconducting state [12,13]. These islands are observed at temperatures above the superconducting state, i.e., above  $T_C$  and up to a temperature  $T^*$ . Similarly in optical lattices, BEC can be localized by the lattice potential [14] or by the potential plus disorder [15]. Lattice models illustrate how sufficient disorder can separate superconducting and anti-ferromagnetic states into independent islands leading to an extended to localized state crossover [16,17]. The localized state is generally referred to as a Bose glass [18,19].

The simplest and most accessible example of Bosons in disorder is liquid helium immersed in porous media. Superfluidity in these “dirty Bose systems” has been extensively investigated [20–24] to determine the superfluid to normal liquid transition temperature,  $T_C$ , and the critical exponents. In neutron scattering studies, we have observed the phonon-roton (P-R) modes of liquid  $^4\text{He}$  at saturated vapor pressure (SVP) ( $p \approx 0$ ) in several porous media [25–30]. These measurements showed that well-defined P-R modes exist both in the superfluid phase and at temperatures above the superfluid phase, i.e., above  $T_C$ . This was

observed first in Vycor [27,29] ( $T_C = 2.05$  K), then in 25 Å mean pore diameter (MPD) gelsil [28] ( $T_C = 0.7$  to 1.3 K) [22,23] and in 44 Å MPD gelsil [30] ( $T_C = 1.92$  K). Modes were observed up to the  $T_\lambda$ , the superfluid-normal transition temperature in bulk liquid helium ( $T_\lambda = 2.17$  K at SVP). Since well-defined P-R modes exist because there is BEC, this suggests that there is BEC above  $T_C$ . This was interpreted as BEC localized to individual islands separated by normal liquid so that there was no phase coherence across the sample, i.e., a Bose glass phase [27–32] at temperatures  $T_C < T < T_\lambda$ .

In this Letter we present measurements of the P-R modes of liquid  $^4\text{He}$  under pressure up to 57 bars in three porous media, 25 Å MPD gelsil, 34 Å MPD gelsil, and 45 Å MCM-41. The aim is to determine the domain of existence of well-defined P-R modes in pressure as well as temperature. At low temperatures (e.g.,  $T = 0.07$  K) we observe well-defined P-R modes at wave vectors in the roton region up to 36–37 bars but not above this pressure. At  $p = 31.2$  bars we observe well-defined P-R modes up to  $T = 1.4$ – $1.5$  K but not above this temperature. Since these pressures and temperatures lie above the superfluid phase, as determined in torsional oscillator measurements by Yamamoto *et al.* [23] in 25 Å gelsil, we conclude that there is a phase containing localized BEC surrounding the superfluid phase at all pressures and temperatures. We again interpret this as a Bose glass phase containing separated islands of BEC that are large enough to support well-defined P-R modes in a sea of normal liquid as at SVP [27–32]. The possibility of a Bose glass phase completely surrounding ordered phases has been predicted [17,33]. A preliminary report of this work was presented [34] at the conference Reports of Progress on Many Body Theory 14. Yamamoto *et al.* [35] have proposed a similar Bose glass phase at higher pressure based on specific heat measurements.

The existence of P-R modes and superfluidity in bulk liquid helium at higher pressures is also of great interest

[36–38]. From the loss of P-R modes observed here, modes in the long wavelength phonon region and in the roton region only are predicted in bulk helium above 30 bars.

The present measurements were performed on the IN6 time-of-flight neutron spectrometer at the Institut Laue-Langevin, France. An incident neutron wavelength of 4.15 Å was used. The gelsil samples of 50% porosity, manufactured by 4F International Co., consisted of two monolithic cylinders each of diameter 1 cm and height 1.5 cm, placed one on top of the other in a cylindrical Al sample cell. The gap between the gelsil cylinders and the cell walls was less than 0.5 mm. The 47 Å MPD MCM-41 was the same as used previously [39] to establish negative pressures. The same cell was used for all three samples and was cooled with a circulating  $^3\text{He}$  cryostat for the 34 Å gelsil and the MCM-41 and by a dilution fridge for the 25 Å gelsil. The neutron scattering measurements were similar to those conducted previously [26–29,40].

To set the stage for our results, a schematic phase diagram of liquid  $^4\text{He}$  in 25 Å MPD gelsil is shown in Fig. 1. The superfluid phase observed by Yamamoto *et al.* [23] is depicted by a solid background (extended BEC) at low temperature and pressure. Surrounding the superfluid phase is the Bose glass phase containing islands of BEC. We observe well-defined P-R modes at all wave vectors investigated ( $0.3 \leq Q \leq 2.3 \text{ \AA}^{-1}$ ) throughout both these phases up to 25 bars.

To introduce the pressure dependence above 25 bars, we show the P-R mode energy dispersion curve,  $\omega_Q$ , versus

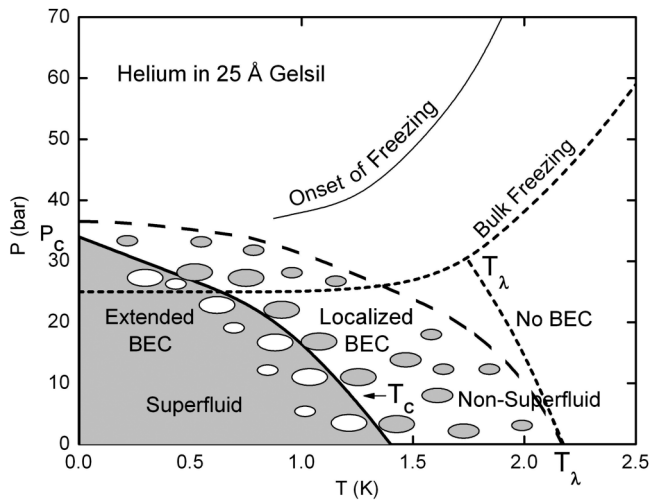


FIG. 1. Schematic phase diagram of  $^4\text{He}$  in 25 Å mean pore diameter (MPD) gelsil. The superfluid phase observed by Yamamoto *et al.* [23] is depicted by the solid region bounded by a solid line ( $T_c$ ). The Bose glass phase containing islands of BEC surrounding the superfluid phase is indicated by dark patches and is bounded by a long dashed line. A solid line marks the onset of freezing [24]. Short dashed lines show the phase boundaries in bulk  $^4\text{He}$ .

wave vector  $Q$  at SVP ( $p \approx 0$ ) and at  $p = 31.4$  bars in Fig. 2. The roton energy is indicated by  $\Delta$  (i.e.,  $\Delta = \omega_Q$  for  $Q$  at the roton minimum). At SVP in bulk superfluid  $^4\text{He}$  a continuous, sharply defined P-R mode is observed [41,42] from low  $Q$  out to  $Q = 3.6 \text{ \AA}^{-1}$ . This is also the case in porous media in the  $Q$  range investigated ( $0.3 \leq Q \leq 2.3 \text{ \AA}^{-1}$ ) [26,28,30]. As pressure increases, the roton energy,  $\Delta$ , decreases [40,43] while the mode energies,  $\omega_Q$ , at lower  $Q$  ( $Q \leq 1.7 \text{ \AA}^{-1}$ ) increase. At approximately 20 bars, the mode energy  $\omega_Q$  at  $Q \approx 1.1 \text{ \AA}^{-1}$  (the maxon region) reaches [39,43]  $2\Delta$  and becomes limited by  $2\Delta$ . The  $\omega_Q$  cannot exceed  $2\Delta$ . If  $\omega_Q > 2\Delta$  the single P-R mode lies in the 2 P-R energy band and can decay to two rotons [44]. For example, as seen in Fig. 2, the  $\omega_Q$  at SVP comes up to and is limited by  $2\Delta$  at higher  $Q$ . All the response at  $\omega$  above  $2\Delta$  is broad [45].

At  $p \geq 26$  bars we no longer observe a well-defined P-R mode in the present porous media in the maxon region (see also Ref. [40]). At 31.2 bars we observe a well-defined P-R mode for  $1.6 \leq Q \leq 2.3 \text{ \AA}^{-1}$  only where  $\omega_Q \leq 2\Delta$  as shown in Fig. 2. At 31.2 bar the mode energy  $\omega_Q$  would lie above  $2\Delta$  for wave vectors  $0.5 \leq Q \leq 1.6 \text{ \AA}^{-1}$  and all the P-R response is broad. We expect a well-defined sound mode in the long wavelength region,  $Q \leq 0.5 \text{ \AA}^{-1}$  but its intensity at  $p = 31.2$  bars is apparently too small to be observed in the present small pore media. Thus, for kinematical reasons unrelated to BEC, the high energy P-R modes broaden and disappear first at the lowest pressures. We now focus on the remaining modes.

Figure 3 shows the temperature dependence of the dynamic structure factor  $S(Q, \omega)$  at  $Q = 2.1 \text{ \AA}^{-1}$  and  $p = 31.2$  bars in 25 and 34 Å MPD gelsil. There are two peaks

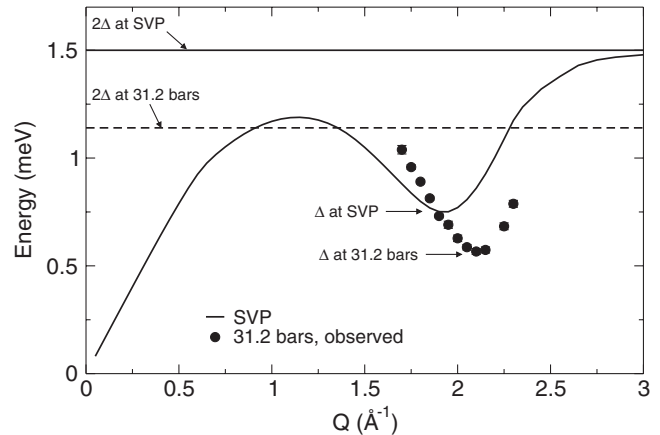


FIG. 2. The solid lines show the phonon-roton (P-R) energy  $\omega_Q$  dispersion curve and twice the roton energy ( $2\Delta$ ) at saturated vapor pressure (SVP) ( $p \approx 0$ ). The solid dots show the present observed P-R dispersion curve at  $p = 31.2$  bars in 34 Å MPD gelsil and the dashed line shows the corresponding  $2\Delta$ . At  $p = 31.2$  bars, P-R modes are observed at wave vectors in the range  $1.6 \leq Q \leq 2.4 \text{ \AA}^{-1}$  only where  $\omega_Q \leq 2\Delta$ .

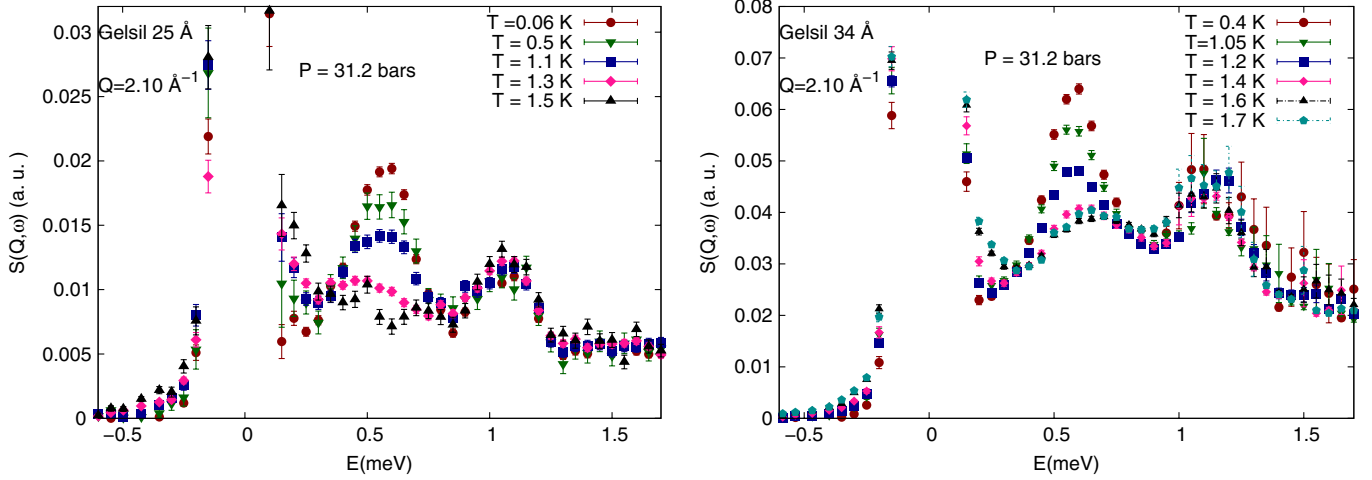


FIG. 3 (color online). Temperature dependence of  $S(Q, \omega)$  of  $^4\text{He}$  in 25 and 34 Å MPD gelsil at  $Q = 2.1 \text{ \AA}^{-1}$  and  $p = 31.2$  bars. There is substantial elastic scattering from the gelsil centered at  $\omega = 0$ . The height of the roton mode peak at  $\omega \approx 0.6$  meV decreases with increasing temperature until a well-defined mode is no longer observed at  $T \gtrsim 1.4$  K. The peak at  $\omega \approx 1.1$  meV arises from phonons in the bulk solid  $^4\text{He}$  that lies between the gelsil sample and the cell wall.

of interest in  $S(Q, \omega)$ , one at energy  $\omega \sim 0.6$  meV arising from rotons in the liquid in the gelsil and one at  $\omega \sim 1.1$  meV arising from phonons in the bulk solid helium around the gelsil sample. As temperature is increased, the roton peak broadens until at  $T = 1.4$  K a well-defined roton is no longer observed. However, the roton does survive up to  $T \approx 1.4$  K, which is well above the  $T_C \approx 0.2$  K of the superfluid phase observed by Yamamoto *et al.* shown in Fig. 1. Thus there is a temperature range ( $T_C \leq T \leq 1.4$  K) at 31.2 bars in which the liquid supports a roton but is not superfluid, as is the case in the temperature range  $T_C \leq T \leq T_\lambda$  at SVP. We identify this as the range of localized BEC.

Figure 4 shows the pressure dependence of  $S(Q, \omega)$  in the roton region at low temperature. The roton is again at energy  $\omega \approx 0.6$  meV and the roton energy decreases with increasing pressure. The peak at  $\omega \approx 1.1$  meV again arises from phonons in the solid around the gelsil and their energy increases with increasing pressure. The P-R mode broadens and is no longer observed at pressures above 37.8 bars. Indeed two samples were grown at 37.8 bars, one which shows a weak P-R mode and the other no mode. We believe that there is a small difference between these pressures and that 37.8 bars represents a cutoff pressure for the existence of rotons in 25 Å MPD gelsil. The pressures shown in Fig. 4 are the pressures at which the solid around the sample was formed at 2.1 K. This pressure 37.8 bars translates to a pressure of 36.3–36.8 bars when the cell is cooled to  $T = 0.07$  K. The pressure 36.3 bars is above the QPT pressure  $p_c = 34$  bars observed by Yamamoto *et al.*. Also since a roton is observed above  $p_c$  there must be liquid in the gelsil above  $p_c$ .

At low temperature, P-R modes in the roton region disappear at approximately the same pressure in all three porous media. Specifically, modes in the roton region are

observed up to but not beyond the pressures and temperatures indicated by the long dashed line in Fig. 5 in the present two gelsils. In the larger pore MCM-41, they are observed up to a somewhat higher temperature at  $p = 31.2$  bars. Also, the P-R modes that have the highest energy,  $\omega_Q$ , broaden and disappear at the lowest pressures ( $p \approx 26$  bars) in all three porous media. The loss of the higher energy P-R modes at  $p \geq 26$  bars is expected to be universal [40,43,44] and to hold for bulk helium.

In summary, we propose that liquid helium in 25 Å MPD gelsil has a Bose glass phase surrounding the superfluid

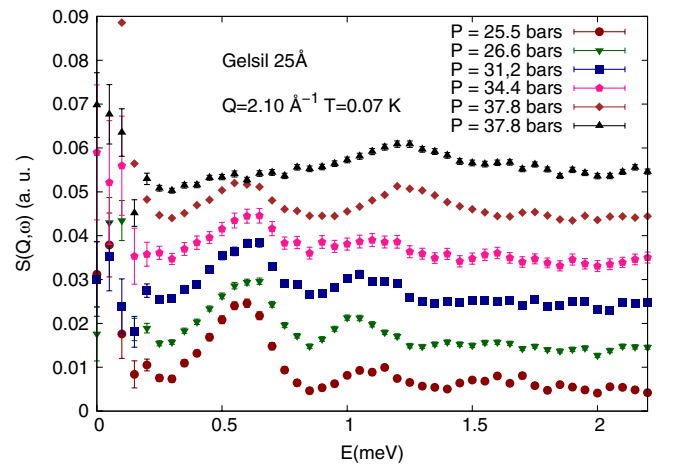


FIG. 4 (color online). Pressure dependence of  $S(Q, \omega)$  at  $Q = 2.1 \text{ \AA}^{-1}$  of  $^4\text{He}$  at  $T = 0.07$  K in 25 Å MPD gelsil. The P-R mode at  $\omega \approx 0.65$  meV is no longer observed at pressures  $p = 37.8$  bars and above. The peak at  $\omega \approx 1.1$  meV arises from phonons in the bulk solid  $^4\text{He}$  around the gelsil sample. The pressure 37.8 bars is the pressure at which the cell was filled at  $T = 2.1$  K and blocked. The corresponding pressure at  $T = 0.07$  K is estimated to be  $p = 36.3\text{--}36.8$  bars.

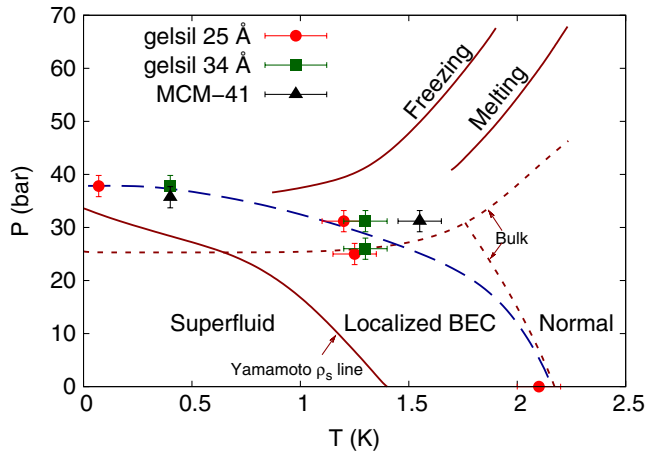


FIG. 5 (color online). Well-defined phonon-roton (P-R) modes are observed at pressures and temperatures up to, but not above, the points with error bars joined by a long dashed line in the present porous media. Otherwise the lines are as described in Fig. 1 for  $^4\text{He}$  in 25 Å MPD gelsil.

phase at all pressure and temperature. This Bose glass phase consists of islands of BEC separated by normal liquid. The evidence for the islands of BEC is the direct observation of well-defined P-R modes in the Bose glass phase. We have shown previously [27–32] that P-R modes and a Bose glass phase exist at SVP at temperatures  $T_C < T < T_\lambda$  (in Vycor, 25 and 44 Å gelsil). BEC is depleted by pressure [36,46] as well as temperature. Thus at higher pressure as well as higher temperature, BEC probably exists only in certain regions in porous media where the density is lowest, for example, in the larger pores.

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