

**Excitations in liquid  $^4\text{He}$  in Geltech silica and localized Bose condensation**

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We have measured the elementary excitations of liquid  $^4\text{He}$  confined in Geltech silica for temperatures between 35 mK and 2.5 K using inelastic neutron scattering techniques. Geltech is a 40–50% porous media. The pore volume diameters are concentrated at 25 Å with at most a few percent of the volume consisting of larger pores of diameter less than 70 Å. We observe well-defined phonon–roton excitations at all wave vectors  $Q$  investigated ( $0.4 \leq Q \leq 2.1 \text{ \AA}^{-1}$ ). Also observed are two-dimensional layer modes propagating in the liquid layers adjacent to the media walls. These are identified as layer modes by measuring the dynamical structure factor of liquid  $^4\text{He}$  as a function of filling of the Geltech. At partial fillings, the energies and lifetimes of the three-dimensional phonon–roton mode differ somewhat from the bulk superfluid  $^4\text{He}$  values. At full filling they are the same within current precision. Most interestingly, we observe a well-defined phonon–roton excitation above the superfluid-normal transition  $T_c = 0.725 \text{ K}$  as determined in a torsional oscillator measurement. Observation of a well defined maxon–roton above  $T_c$  in Geltech suggests, as in the case of Vycor, that there is localized Bose–Einstein condensation with phase coherence on short length scales above the macroscopic superfluid transition temperature.

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**I. INTRODUCTION**

The dynamics of fluids, polymers, and macromolecules in confined geometries is a topic of great current interest.<sup>1</sup> Liquid  $^4\text{He}$  confined in porous media such as Vycor and aerogel is a prototype example of a Bose quantum liquid in confinement and disorder.<sup>2–6</sup> While the superfluid and thermodynamic properties of liquid  $^4\text{He}$  in a wide variety of porous media have been extensively investigated,<sup>2,5</sup> measurements of the dynamics by inelastic neutron scattering have only recently begun.<sup>7</sup> To date, measurements of the excitations in aerogel of 95% and 87% porosity,<sup>8–13</sup> in Vycor,<sup>13–15</sup> and in xerogel<sup>16</sup> have been reported. A brief report on Geltech silica has appeared.<sup>17</sup> The aim of the measurements is to reveal how confinement and disorder modifies the characteristic phonon–roton (p–r) excitations from the bulk superfluid values, whether it creates new excitations and, where possible, relate changes to observed thermodynamic properties.

The basic finding to date is that, within current precision, the energies and lifetimes of the three-dimensional (3D)

phonon–roton excitations of superfluid  $^4\text{He}$  in Vycor and aerogel are the same as in the bulk both at low temperature and as a function of temperature.<sup>7,12,13,15</sup> In addition, superfluid  $^4\text{He}$  in Vycor<sup>14,15</sup> and aerogel<sup>12,13</sup> supports a two-dimensional (2D) mode that propagates in the liquid layers adjacent to the media walls. This new mode is clearly observed at wave vectors in the roton region,<sup>12,15</sup> and appears to be the same as the 2D mode observed in superfluid  $^4\text{He}$  films on surfaces<sup>7,18,19</sup> and inferred from specific heat<sup>20,21</sup> and superfluid density<sup>22</sup> measurements in Vycor, for example.

An interesting observation<sup>15</sup> in Vycor is that the weight of the 3D phonon–roton excitation in the total dynamic structure factor,  $S(Q, \omega)$ , does not scale with the superfluid density,  $\rho_s(T)$ . There is weight in the p–r excitation above the superfluid–normal transition temperature,  $T_c$ , as determined by torsional oscillator experiments,<sup>4,23</sup> for example. This suggests<sup>13,15</sup> that there is localized Bose–Einstein condensation (BEC) above  $T_c$  where  $\rho_s(T) = 0$ . Specifically, bulk superfluid  $^4\text{He}$  at low temperature supports a uniquely sharp

density (phonon–roton) excitation up to wave vectors of  $3.6 \text{ \AA}^{-1}$ . As temperature is increased, the excitation broadens and above the superfluid to normal transition ( $T_\lambda = 2.172 \text{ K}$  at SVP), normal  $^4\text{He}$  does not support a well-defined density excitation at wave vectors higher than the phonon region ( $Q \geq 0.7 \text{ \AA}^{-1}$ ). This is clear at the maxon wave vector ( $Q = 1.1 \text{ \AA}^{-1}$ ) and at wave vectors beyond the roton ( $Q \geq 2.5 \text{ \AA}^{-1}$ ) where normal  $^4\text{He}$  supports only a broad response centered at high energy near the free atom recoil energy characteristic of scattering from weakly interacting particle–hole excitations. An interesting finding is that  $^4\text{He}$  in Vycor does support a well defined, though thermally broadened, p–r excitation at higher wave vectors ( $Q \geq 0.7 \text{ \AA}^{-1}$ ) above  $T_c$ . Since well defined p–r excitations are associated with the existence of Bose–Einstein condensation,<sup>24,25</sup> this finding suggests there is localized BEC in Vycor above the superfluid–normal transition temperature  $T_c = 1.95 \text{ K}$  observed in a macroscopic measurement of superfluidity.<sup>4,23</sup> That is, neutron scattering is a local probe and could reveal “localized” BEC (local phase coherence) via the excitations that does not extend across the whole sample. Phase coherence across the whole sample as observed in torsional oscillator experiments may require a lower temperature.

In this paper we present measurements of excitations in Geltech silica, a porous media similar to aerogel consisting of interconnected pore volumes of predominantly  $25 \text{ \AA}$  diameter. The aim is to investigate excitations in smaller pore media where the effects of confinement and disorder would be greater. In particular, in Geltech silica at 88% filling the transition from normal to superfluid  $^4\text{He}$  observed in a torsional oscillator measurement<sup>26</sup> is reduced to  $T_c = 0.725 \text{ K}$  reflecting the smaller pore diameters. We find that liquid  $^4\text{He}$  in Geltech indeed supports phonon–roton excitations above the superfluid–normal transition temperature  $T_c = 0.725 \text{ K}$ . This reproduces the finding in Vycor noted above and provides further support for the concept of localized BEC above  $T_c$  confined within pores. The energies and lifetimes of the phonon–roton excitations are similar to those in bulk superfluid  $^4\text{He}$  with some small differences observed at partial fillings. We also observe the 2D layer modes found in Vycor and aerogel at wave vectors in the roton region. In addition we observe a relatively weak, nearly energy-independent inelastic response at all wave vectors that appears first just after one  $^4\text{He}$  layer is complete, which corresponds to approximately 50% filling. The intensity of this response does not increase beyond 70% filling and is independent of temperature up to  $T = 2.5 \text{ K}$ .

In Sec. II we describe the experiment. The results at low temperature as a function of filling of the Geltech and as a function of temperature are presented in Sec. III. The interpretation of the results is discussed in Sec. IV.

## II. EXPERIMENT

Geltech is a porous silica glass having nominally  $25 \text{ \AA}$  pore diameters produced using the sol–gel technique by the Geltech Corporation.<sup>27</sup> Our sample consisted of two cylindrical rods of Geltech of approximately  $10 \text{ mm}$  diameter and  $20$

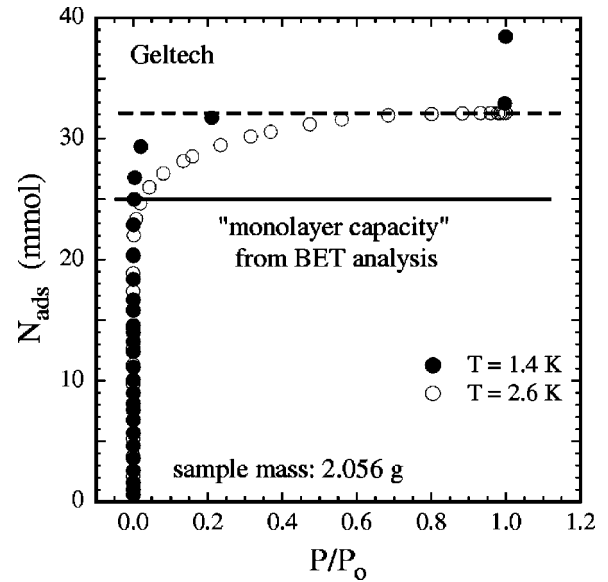


FIG. 1.  $^4\text{He}$  pressure isotherms in  $25 \text{ \AA}$  pore diameter Geltech silica. The dashed line gives the total capacity ( $32.1 \text{ mmol}$ ) and the solid line gives the “monolayer capacity” ( $25 \text{ mmol}$ ) of the sample.

$\text{mm}$  height each. The rods were baked and flushed under a mixture of deuterated water and methanol to replace as much as possible of the hydrogen contained in the  $\text{OH}^-$  groups attached to the walls of the sample by deuterium. This cleaning and substitution was performed in order to reduce the elastic scattering from the Geltech sample. We also took special care thereafter never to expose the Geltech sample to air.

Helium pressure isotherm measurements were made on one of the rods at  $1.4 \text{ K}$  and  $2.6 \text{ K}$  as shown in Fig. 1. The weight and volume of the rod was  $2.056(1) \text{ g}$  and  $1.56(6) \text{ cm}^3$ , respectively. Using the bulk value for the density of silica,  $2.2 \text{ g/cm}^3$ , these values give a porosity of 40%. From Fig. 1 a total absorption capacity of  $32.1 \text{ mmol}$  of  $^4\text{He}$  for the sample ( $15.6 \text{ mmol/g}$  and  $20.6 \text{ mmol/cm}^3$ ) is obtained. Miyamoto and Takano<sup>26</sup> quote a total absorption capacity of  $22.7 \text{ mmol/cm}^3$  for their sample. A Brunauer, Elmet, and Teller (BET) analysis yields a “monolayer” absorption capacity of  $25.0 \text{ mmol}$  or  $12.2 \text{ mmol/g}$ . The corresponding BET surface area of the sample is approximately  $1500 \text{ m}^2$  or  $950 \text{ m}^2/\text{cm}^3$ . Assuming, as a simple model, that the pores consist entirely of cylinders of  $25 \text{ \AA}$  diameter, the area per volume is  $4/25 \text{ \AA}^{-1}$  or  $640 \text{ m}^2/\text{cm}^3$ . Thus Geltech must contain significant microporosity or the “monolayer” capacity should be interpreted as all the  $^4\text{He}$  that is tightly bound to the surface. The simple model also predicts that 52% of the total capacity is in the first layer (83% in the first two layers). The “monolayer” capacity shown in Fig. 1 is 78% of the total. As will be seen below, excitations in the liquid begin to propagate at 70% filling. We therefore interpret the “monolayer” capacity shown in Fig. 1 as an estimate of the “deadlayer” capacity.

An important matter is the distribution of the size of the pores. Figure 1 shows that at 90% filling  $p/p_0 \approx 0.3$  and at 95% filling,  $p/p_0 \approx 0.5$ . Thus even for the last 5% of  $^4\text{He}$  entering the Geltech, the vapor pressure is still much less than the bulk value,  $p_0$ . The last few percent of  $^4\text{He}$  entering

Geltech is still tightly bound to the Geltech surface in small pores. Figure 1 is consistent with the  $N_2$  isotherms carried out by the Geltech corporation. Comparing Fig. 1 with isotherms for Vycor, which has a significantly different shape, suggests any larger pores in Geltech have diameters less than those in Vycor (70 Å).

Thus the isotherm measurements show that the Geltech sample contains uniform diameter pores, nominally 25 Å, with at most a few percent of the volume consisting of larger pores of diameter less than 70 Å. At full filling, the two rod sample holds 1.78 cm<sup>3</sup> of liquid  $^4\text{He}$  at  $T=1.4$  K and SVP. This sample was fitted in a cylindrical aluminum sample cell. The total free volume between the Geltech sample and the cell walls, arising from variations in sample diameter, was measured to be 0.22 cm<sup>3</sup>. The sample cell was mounted in a dilution refrigerator, which was installed on the neutron spectrometer.

The measurements were performed on the IN6 time-of-flight spectrometer at the high-flux reactor of the Institut Laue–Langevin, using an incident neutron energy of 3.83 meV and an energy resolution (FWHM) of about 110  $\mu\text{eV}$ . We performed standard data treatment in order to obtain the dynamical structure factor  $S(Q, \omega)$  of liquid  $^4\text{He}$  in the Geltech sample. The integrated intensity of the sharp excitation peak observed in the data (see below Fig. 4) is normalized to bulk measurements.<sup>28</sup> As the data contains other features, this normalization is not absolute, but allows a direct comparison with spectra obtained from bulk  $^4\text{He}$ . We obtained  $S(Q, \omega)$  over a wide wave-vector range,  $0.4 \leq Q \leq 2.1$  Å<sup>-1</sup> at low temperature (between 35 mK and 0.3 K, depending on the filling studied) and for several fillings between 56% and 105% of the full sample (105% corresponds to a fully filled Geltech sample plus approximately 0.1 cm<sup>3</sup> of liquid  $^4\text{He}$  in the gap). For a filling fraction of 83%, we measured the dynamical structure factor at three low temperatures ( $T=35$  mK, 0.1 K, and 0.75 K). For the slightly over-full sample, we measured the temperature dependence of the dynamical structure factor between  $T=35$  mK and  $T=2.5$  K. The scattering from the empty Geltech sample was also measured at  $T=50$  K. In a separate measurement on the same spectrometer, we measured the scattering from the sample cell completely full (Geltech sample + free volume of 0.22 cm<sup>3</sup> between the sample and the cell wall) at low temperature,  $T=0.5$  K.

### III. RESULTS

Figure 2 shows the observed intensity of inelastic neutron scattering from superfluid  $^4\text{He}$  in Geltech silica as a function of energy transfer for different fillings and for the empty Geltech sample, measured at  $T=50$  K. Since the empty cell measurement was taken at a higher temperature, the observed intensity next to the elastic peak can be higher than the filled sample case, due to low-energy excitations in Geltech itself. The 56% full and empty cell intensities are very similar showing that there is little or no intensity arising from the  $^4\text{He}$  up to 56% filling and we have therefore taken the 56% filling data as the reference, background data. At 83% filling the data show a well-defined excitation at all

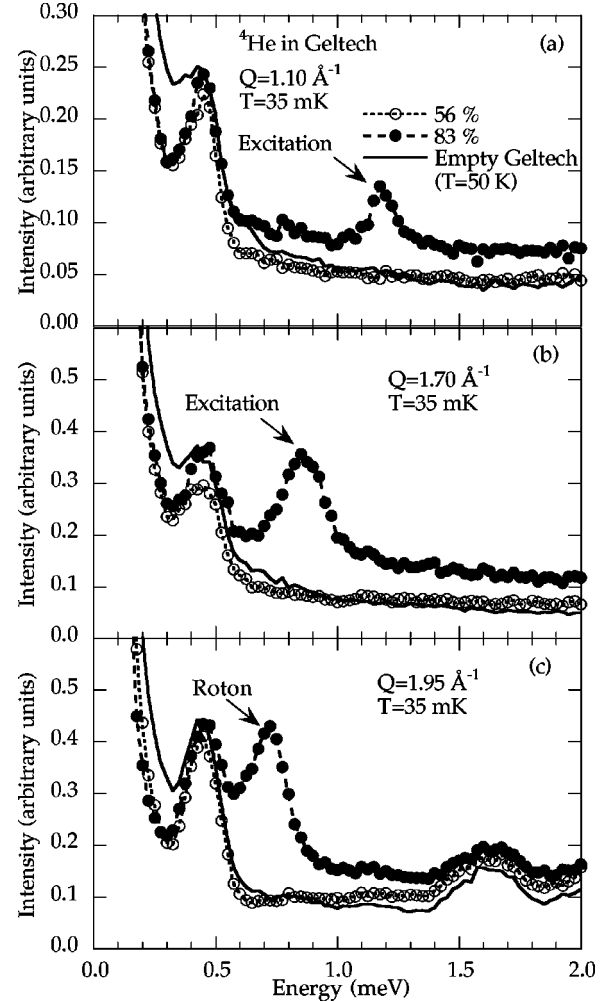


FIG. 2. Spectra obtained at low temperature before any background subtraction at the maxon (a), between the maxon and the roton (b), and at the roton wave vector (c) for partial fillings of 56% (open circles) and 83% (solid circles) at low temperatures (35 mK) as well as for the empty Geltech sample (solid line) at  $T=50$  K. The peaks centered about 0.45 meV and 1.65 meV seen in the empty Geltech sample data are parasite peaks corresponding to scattering by the cryostat walls.

wave vectors investigated  $0.4 \leq Q \leq 2.1$  Å<sup>-1</sup>, plus a nearly constant inelastic intensity in the energy range  $0.5 \leq \omega \leq 2.0$  meV. Energies outside this range were not investigated. As discussed in the text, the origin of this nearly energy independent and temperature independent (up to  $T=2.5$  K) intensity is not known.

Figure 3 shows the dynamical structure factor,  $S(Q, \omega)$ , of superfluid  $^4\text{He}$  in Geltech at low temperature ( $T \leq 0.3$  K) for fillings between 60% and 105%. At 60%, filling  $S(Q, \omega)$  is very small or negligible. At 68% filling,  $S(Q, \omega)$  is largely constant (independent of  $\omega$ ) over the energy range shown. The excitation is seen first at 76% filling. At 105% filling the excitation is sharp and comparable to that observed in bulk superfluid  $^4\text{He}$ . Assuming Geltech consists of 25 Å diameter, cylindrical pores and that the densities in the first and second  $^4\text{He}$  layers on the Geltech are the same as observed on graphite, the first layer on Geltech takes

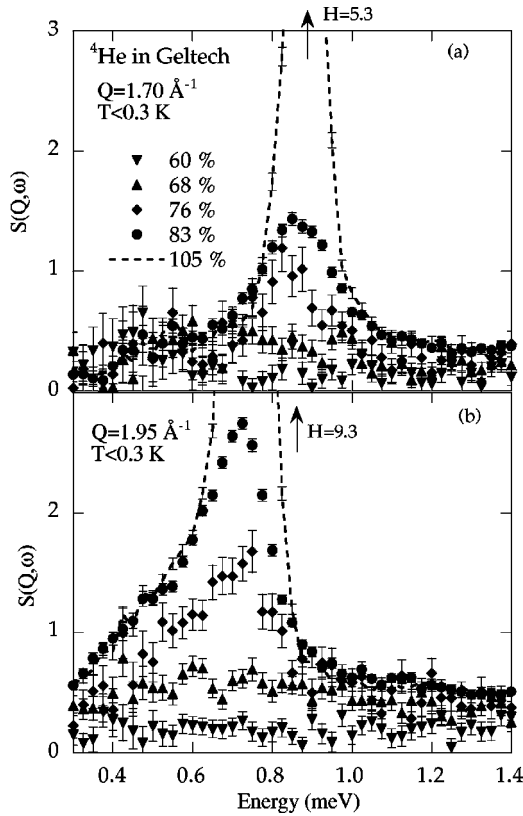


FIG. 3. Low temperature dynamical structure factor of liquid  ${}^4\text{He}$  in Geltech at  $Q=1.70 \text{ \AA}^{-1}$  (a) and at the roton wave vector (b) for several fillings as indicated.

up 52% of the total  ${}^4\text{He}$  volume required to fill the pores, the two first layers 83%. This calculation provides only a most approximate indication. Also, the pores will fill irregularly with some crevices filling by capillary action after the first layer is complete.

In Fig. 4 we show fits to  $S(Q, \omega)$  that we performed in order to interpret the data. There are clearly three components. The main peak is interpreted as the 3D roton excitation as observed in bulk superfluid  ${}^4\text{He}$ . The broad peak centered below the 3D excitation energy is the 2D layer mode, as observed in Vycor and in aerogel of 87% porosity. Its width is approximately three times the energy resolution in the present experiment. This 2D layer mode propagates in the liquid layers next to the media walls. The 2D layer mode is not observed at fillings of 68% or less. It is first observed at 76% filling. The intensity in the mode increases between 68% and 83% filling and saturates at higher fillings. The third component is nearly energy independent with a very broad maximum somewhat above the 3D excitation energy. The intensity in this very broad component does not increase significantly with filling beyond 76%.

Figure 5 shows the integrated intensity in the 2D layer mode and the 3D mode at  $Q=2.00 \text{ \AA}^{-1}$  as a function of filling. The relative intensity in the 2D and 3D components of  $S(Q, \omega)$  depends somewhat on the way the data at low filling (e.g., at 76%) is fitted. At 68% filling we do not observe any 2D or 3D mode. Intensity in the modes is first seen at 76% filling. The intensity in the 2D mode increases with

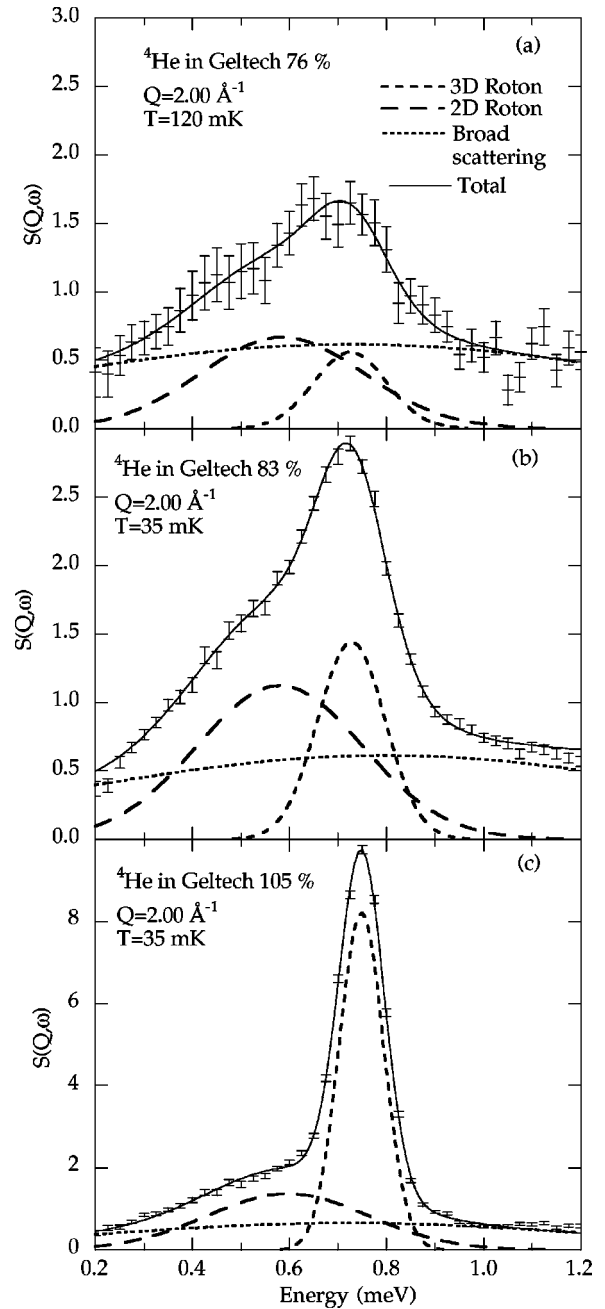


FIG. 4.  $S(Q, \omega)$  at low temperature of liquid  ${}^4\text{He}$  in Geltech near the roton wave vector for a filling of 76% (a), 83% (b), and 105% (c). The dotted line represents the broad, constant, contribution. The dashed and long-dashed lines represent, respectively, 3D-roton and 2D-roton excitations. The solid line is the sum of the three components.

filling less rapidly than that in the 3D mode. In aerogel,<sup>12,13</sup> the intensity in the 2D mode saturates with filling. After seven layers were deposited in aerogel, the intensity did not increase further. In Geltech, the second layer fills between 52% and 83% filling and full filling corresponds to one/three liquid layers. The data in Fig. 5 is consistent with saturation of the 2D mode intensity with filling if sufficient layers could be deposited. In Fig. 5 the 2D mode intensity is fitted to the empirical expression introduced by Thomlinson *et al.*<sup>29</sup> for



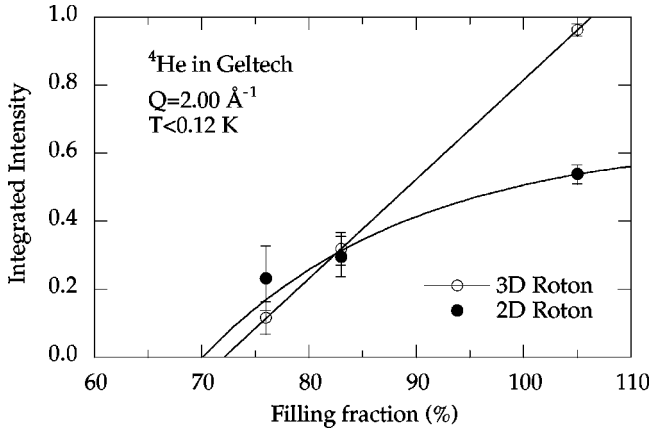


FIG. 5. Integrated intensity of the 3D bulklike roton (open circles) and the 2D layer mode (closed circles) in superfluid  $^4\text{He}$  in Geltech silica at  $T \leq 0.12$  K as a function of filling. The solid lines show a linear fit to the 3D mode and a fit of the empirical expression introduced by Thomlinson *et al.* (Ref. 29) given by Eq. (1) to the 2D mode.

helium on graphite,

$$I(x) = I_\infty \{1 - \exp[-(x - x_0)/\zeta]\}, \quad (1)$$

where  $x$  is the filling fraction,  $x_0$  the value of  $x$  where the 2D intensity starts to grow, and  $\zeta$  is a characteristic filling thickness. This expression describes the filling dependence well for films on graphite<sup>29</sup> and in aerogel<sup>12,13</sup> and is consistent with the present data. The parameter values obtained from the fit shown in Fig. 5 are  $I_\infty = 0.64$ ,  $x_0 = 70\%$ , and  $\zeta = 19\%$ .

The intensity in the 3D mode increases linearly with filling. At 105% filling there is some bulk liquid  $^4\text{He}$  around the sample. The Geltech sample holds a volume of  $1.78 \text{ cm}^3$  of liquid  $^4\text{He}$  at SVP. There is a gap of  $0.22 \text{ cm}^3$  around the sample—between the sample and the cell wall. Measurements of the 3D roton intensity when the Geltech and the gap are completely full, show that at 105% filling the gap is approximately half full. At 105% filling, approximately 2/3 of the 3D roton intensity shown in Fig. 5 comes from  $^4\text{He}$  in the Geltech, and 1/3 from bulk  $^4\text{He}$  in the gap. All of the 2D mode intensity must come from liquid  $^4\text{He}$  in the Geltech.

From fits of the type shown in Fig. 4, we obtain the 3D phonon–roton excitation energy,  $\omega_Q$ , shown in Fig. 6. Within the present precision (the size of the circles), the 3D phonon–roton energy of superfluid  $^4\text{He}$  observed in Geltech silica at full filling (105%) is the same as in the bulk. In the roton region, the mode energy in Geltech silica at 83% filling may be somewhat less than that in the bulk. As in Vycor and aerogel,<sup>13</sup> a 2D layer mode can be identified for wave vectors in the roton region,  $1.7 \leq Q \leq 2.1 \text{ \AA}^{-1}$ . At lower  $Q$ , ( $Q \leq 1.7 \text{ \AA}^{-1}$ ), a 2D mode component could not be distinguished from the 3D mode and the very broad component of  $S(Q, \omega)$ . The energy of the 2D layer mode at the roton wave vector in Geltech silica,  $\Delta_{2D} = 0.59(1) \text{ meV}$ , lies between the value observed in Vycor [ $\Delta_{2D} = 0.55(1) \text{ meV}$ ] and that observed in 87% porous aerogel [ $\Delta_{2D} = 0.63(1) \text{ meV}$ ].<sup>12,13,15</sup>

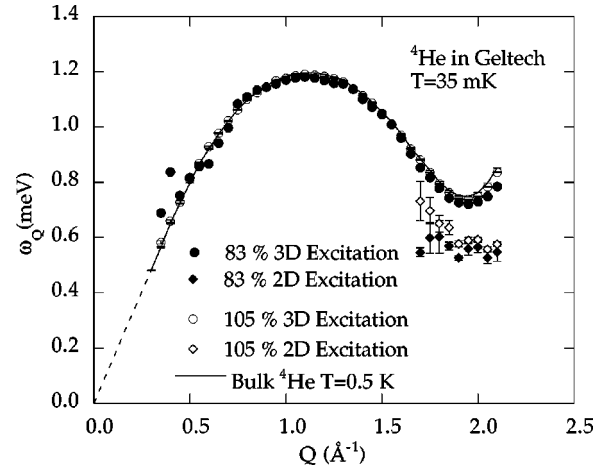


FIG. 6. Low temperature 3D phonon–roton and 2D excitation energy dispersion curves of liquid  $^4\text{He}$  in Geltech for a filling of 83% (closed symbols), and 105% (open symbols).

Similarly, from fits of the type shown in Fig. 4, we obtain the full width at half maximum and the weight in the 3D phonon–roton excitation shown in Figs. 7(a) and 7(b), respectively. At 83% filling, the width is definitely larger than that in the bulk (see Refs. 15 and 28) for wave vectors  $Q \gtrsim 1.5 \text{ \AA}^{-1}$ . At 105% filling, the width may be marginally greater than in the bulk. However, a separate fit to the data shows that the width of the net intensity added between 83%

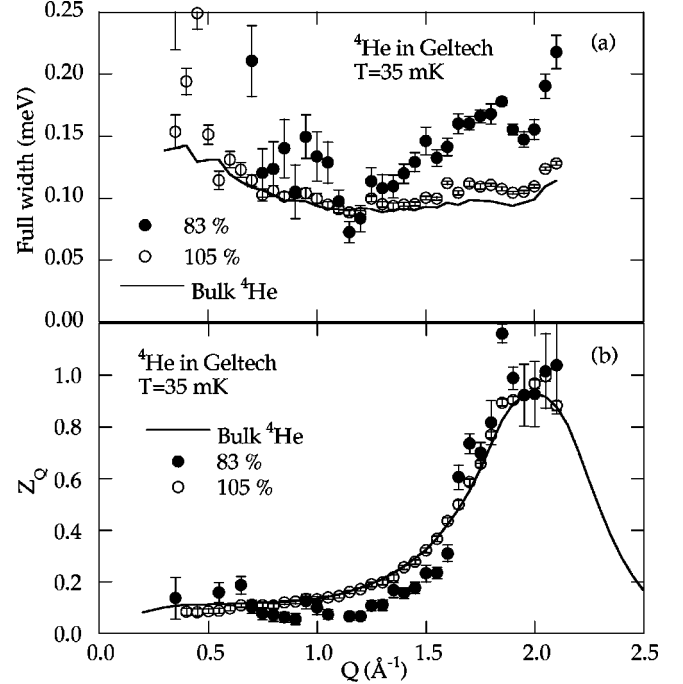


FIG. 7. phonon–roton full width at half maximum (a) and relative weight (b) of liquid  $^4\text{He}$  in Geltech for a filling of 83% (closed symbols), and 105% (open symbols). The weights in (b) are normalized to the bulk value at  $Q = 1.95 \text{ \AA}^{-1}$ . The solid lines correspond to bulk liquid  $^4\text{He}$ ; (a) measured in a previous experiment on the same spectrometer (Ref. 15) and (b) values from the literature (Ref. 30).

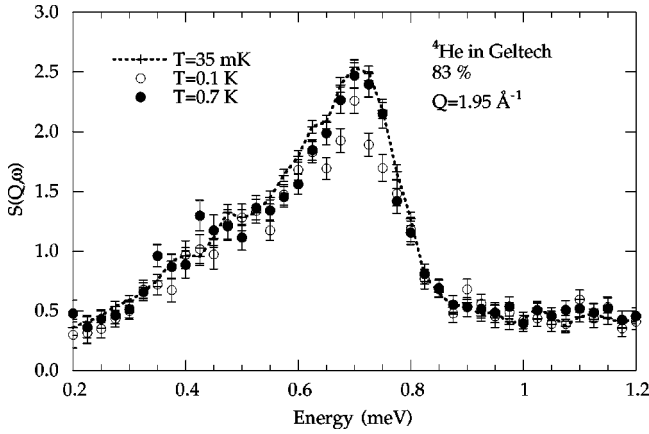


FIG. 8. Dynamic structure factor of liquid  ${}^4\text{He}$  in Geltech at the roton wave vector for a filling of 83% at temperatures as shown.

and 105% is the same as in the bulk [see Fig. 7(a)]. Thus, the apparently larger width at 105% arises entirely from superfluid  ${}^4\text{He}$  in those regions that are filled before the Geltech is 83% full. The intensity in the phonon–roton excitation at 83% filling also differs somewhat from that in the bulk.<sup>30</sup>

In Fig. 8 we show  $S(Q, \omega)$  at the roton wave vector in Geltech at 83% filling at two low temperatures ( $T = 35$  mK and  $T = 0.1$  K) and at  $T = 0.75$  K. In this temperature range,  $S(Q, \omega)$  does not change significantly with temperature except for a possible lowering of the peak height at the intermediate temperature  $T = 0.1$  K. In bulk  ${}^4\text{He}$  the weight of the single phonon–roton in  $S(Q, \omega)$  for  $Q \approx 0.7 \text{ \AA}^{-1}$  scales approximately with the superfluid density  $\rho_S(T)$  [or condensate fraction  $n_0(T)$ ]. At temperatures above  $T_\lambda = 2.17$  K where  $\rho_S(T) = 0$ , the intensity that was in the sharp phonon–roton peak at low temperature is spread over a wide energy range with no clear excitation remaining. If this scaling carried over to liquid  ${}^4\text{He}$  in Geltech, where  $T_c = 0.725$  K as observed in a torsional oscillator experiment<sup>26</sup> and  $\rho_S(T) = 0$  for  $T > T_c$ , we would expect to see a significant temperature dependence of  $S(Q, \omega)$  between  $T = 0.1$  K and  $T = 0.75$  K ( $T > T_c$ ) and only a broad  $S(Q, \omega)$  would remain above  $T_c$ . This is clearly not the case. The 3D roton and 2D layer mode components of  $S(Q, \omega)$  remain well defined up to  $T_c$  and display no significant temperature dependence (see Fig. 8).

Figure 9 shows the temperature dependence of  $S(Q, \omega)$  in Geltech at 105% filling and in bulk liquid  ${}^4\text{He}$  from low temperature to  $T > T_\lambda$ . At low temperatures,  $S(Q, \omega)$  in Geltech displays a well-defined 3D roton peak and a 2D layer mode at lower  $\omega$  as shown in more detail in Fig. 4(c). In the bulk, only the 3D roton is observed. As the temperature is increased, the height of the 3D roton peak and the weight in the 2D mode decreases. By  $T = 1.5$  K, a 2D layer mode is not needed to get a good fit to  $S(Q, \omega)$  in Geltech. At the temperature  $T = 1.5$  K and above, there is no evidence for a 2D layer mode in Geltech. The weight in the 3D roton peak scales with  $T$  similarly in Geltech and in the bulk. If the weight in the well-defined roton peak scaled with  $\rho_S(T)$  in Geltech, we would expect to see a rapid temperature dependence of  $S(Q, \omega)$  in Geltech since<sup>26</sup>  $\rho_S(T) \rightarrow 0$  at

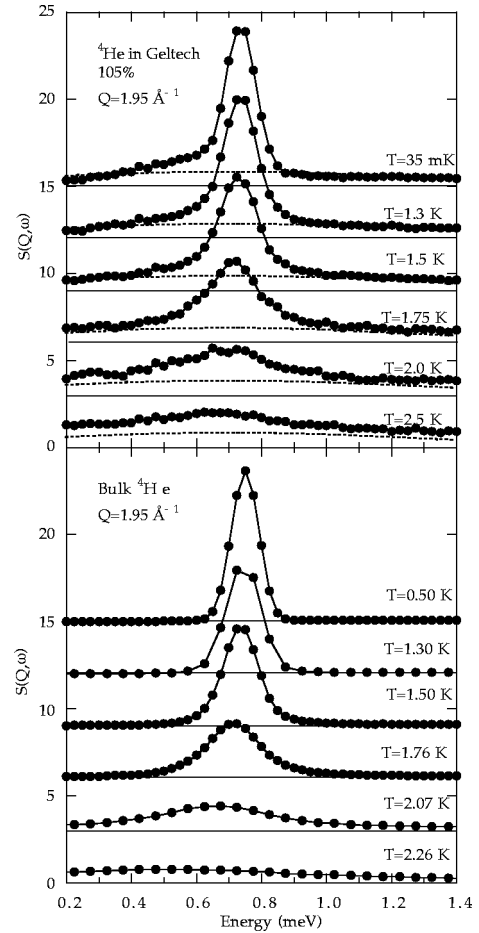


FIG. 9. Dynamic structure factor of liquid  ${}^4\text{He}$  in Geltech for a filling of 105% (upper panel) and of bulk liquid  ${}^4\text{He}$  (lower panel) at the roton wave vector at temperatures as shown. The dotted line represents the broad temperature-independent contribution.

$T_c = 0.725$  K. Specifically, we observe a well-defined roton peak at temperature  $T > T_c$  in Geltech where  $\rho_S(T) = 0$  as observed in a torsional oscillator experiment. Since the existence of well-defined excitations at higher wave vectors above the phonon (sound) region depends on the existence of a Bose–Einstein condensate (BEC), the observation of a well-defined roton peak above  $T_c$  suggests that there is Bose condensation above  $T_c$  in Geltech. This BEC is probably localized in favorable pores providing phase coherence over pore size distances only. Superfluidity would be observed in torsional oscillator experiments only at lower temperatures when this phase coherence extends across the whole sample.

#### IV. DISCUSSION

$S(Q, \omega)$  of liquid  ${}^4\text{He}$  in Geltech silica observed here has three components: a broad nearly energy independent component that extends over the whole energy range investigated, a layer mode at wave vectors in the roton region and a 3D phonon–roton excitation at all wave vectors investigated.

### A. Filling dependence

Each component has a different filling dependence. The broad component is first observed at 60% filling and the intensity in it does not increase further beyond 68% filling. The first layer of  $^4\text{He}$  on the Geltech walls surface is complete at approximately 52% filling, a value obtained if we assume cylindrical pores of 25 Å diameter and  $^4\text{He}$  layer densities as observed on graphite.<sup>29</sup> This first layer is expected to be tightly bound to the substrate. After the first layer is complete, the filling is expected to be liquid and irregular by capillary action, with some regions filling much more completely than others. If Geltech was filled by layers the second liquid layer would be complete at approximately 83% filling. Thus the broad component arises from some dynamic process involving the atoms added just after the first layer is complete. The intensity of scattering from the process is also unaffected by the addition of further  $^4\text{He}$  layers (beyond 68% filling). A broad component has not been reported in Vycor and aerogel. However the fraction of  $^4\text{He}$  close to a surface is much higher in the present Geltech sample than in Vycor or aerogel. For example, the surface area in Geltech is 650–900 m<sup>2</sup>/cm<sup>3</sup> compared to approximately 170 m<sup>2</sup>/cm<sup>3</sup> in aerogel and Vycor. In a second experiment on the present Geltech sample, we observed a somewhat more intense broad component than shown here, suggesting that the intensity in the broad component is sensitive to the surface condition.

The 2D layer mode is observed first at 76% filling, before the second layer (first liquid layer) is complete. In aerogel, the 2D layer mode propagates in the third to sixth liquid layer adjacent to the media walls. In Geltech, the layer mode begins at lower coverage (corresponding to less than one complete liquid layer) although as noted, the Geltech probably fills very irregularly with some regions having significantly thicker coverage than others. The intensity in the layer mode saturates at higher fillings as in aerogel.<sup>12</sup>

The third component, the 3D phonon–roton excitation, is observed at fillings of 76% and above. At partial fillings, such as 83%, the phonon–roton energy is slightly lower than the bulk value. This is opposite to aerogel, where the energy at partial fillings is higher than in bulk  $^4\text{He}$ . The  $p$ – $r$  mode arising from the  $^4\text{He}$  added beyond 83% filling has the same energy as in the bulk.

### B. Temperature dependence

The temperature dependence of each component is also interesting. The broad component observed in Geltech appears to be independent of temperature. For example, the broad component identified in Fig. 4 at low  $T$  is shown as the dashed line in the upper frame of Fig. 9. At  $T=35$  mK in Fig. 9, this broad component appears as additional intensity observed in Geltech that is not observed in bulk  $^4\text{He}$  (lower frame of Fig. 9). There appears to be additional intensity observed in Geltech that is not seen in the bulk at all temperatures in Fig. 9 suggesting a broad component in Geltech that is approximately independent of  $T$ .

The temperature dependence of the 2D mode, which contributes intensity at energies below the main 3D excitation

peak and which leads to the asymmetric roton peaks shown in the upper frame of Fig. 9 at  $T=35$  mK, is difficult to determine unambiguously. At higher temperature the peaks are less asymmetric suggesting that the 2D intensity is smaller at higher temperature. Also, a good fit to these broadened asymmetric peaks can be obtained without including a 2D mode at higher  $T$ . However, because the 3D mode dominates  $S(Q, \omega)$  and itself broadens with increasing temperature it is difficult to determine the temperature dependence of the 2D mode with any certainty.

The  $T$  dependence of the 3D mode is most interesting. At 83% filling, as shown in Fig. 8, the roton has no  $T$  dependence between low  $T$  and  $T=0.75$  K, as in bulk  $^4\text{He}$ . The 3D roton shows no loss of intensity or broadening over this temperature range. At 83% filling the  $^4\text{He}$  vapor pressure is much less than the bulk value so that all the  $^4\text{He}$  is in the Geltech. Miyamoto and Takeno<sup>26</sup> find that at a filling of 88% the macroscopic superfluid density in Geltech goes from  $\rho_S(T)=1$  to  $\rho_S(T)=0$  over this  $T$  range. Thus the 3D roton intensity does not scale with  $\rho_S(T)$  in Geltech as it does in the bulk. Particularly there is a well-defined 3D excitation in Geltech for  $T>T_c$  where  $\rho_S(T)=0$ . Since well-defined  $p$ – $r$  excitations are usually associated with BEC, this suggests that there is BEC in Geltech at temperatures above  $T_c$ . Similarly, at 105% filling where approximately 2/3 of the intensity comes from liquid  $^4\text{He}$  in Geltech (the rest comes from the surrounding bulk liquid), the temperature dependence of the 3D roton mode shown in Fig. 9 is very similar to that in the bulk. In the bulk, the intensity in the phonon–roton mode for wave vectors beyond the phonon region ( $Q>0.7$  Å<sup>-1</sup>) is observed to scale approximately as  $\rho_S(T)$ . Glyde and Griffin<sup>31–33</sup> have proposed that this scaling really follows the condensate fraction,  $n_0(T)$  rather than  $\rho_S(T)$ , since well-defined excitations in a Bose fluid are associated with a condensate. Since the temperature dependence of the intensity in the 3D mode in Geltech is similar to that in bulk superfluid  $^4\text{He}$  this suggests that there is a localized Bose condensate fraction  $n_0(T)$  in Geltech above  $T_c$  that has a  $T$  dependence similar to that in bulk  $^4\text{He}$ .

### C. Localized Bose–Einstein condensate

To suggest a possible picture of  $^4\text{He}$  in porous media consistent with this temperature dependence, we note that the normal to superfluid transition temperature,  $T_c$  in confinement as observed in torsional oscillator (TO) measurements lies below that in the bulk,  $T_\lambda=2.172$  K. The greater the degree of confinement, the lower is  $T_c$ . In fully filled aerogel (pore diameters  $d>100$  Å), Vycor ( $d\sim 70$  Å) and Geltech silica ( $d\sim 25$  Å),  $T_c=2.167$  K,  $T_c=1.95$  K, and  $T_c=0.725$  K, respectively.<sup>4,26</sup>

At low  $T$ , the thermal de Broglie wavelength of  $^4\text{He}$  atoms is long (e.g., at  $T=20$  mK,  $\lambda=60$  Å). At low  $T$  and low coverage (dilute),  $^4\text{He}$  in Vycor appears to behave like a Bose gas.<sup>34</sup> That is, at low coverages the  $^4\text{He}$  atoms (above a completed inert layer of  $^4\text{He}$  on the Vycor walls) translate freely along the interconnected Vycor pores and  $\lambda$  is comparable to the interatomic spacing (40 Å) and to the Vycor pore diameter. The observed  $T_c$  is quite well reproduced by

the expression for Bose–Einstein condensation in a bulk ideal Bose gas  $n\lambda^3 = 2.612$ . Particularly, the observed change of  $T_c$  with  $^4\text{He}$  density in Vycor arising from the interaction between the  $^4\text{He}$  atoms appears to agree well<sup>34</sup> with the predictions for an ideal Bose gas.<sup>35</sup> Thus at low  $T$  and low coverages where  $\lambda$  is comparable to  $d$ , confinement does not appear to cutoff or shorten  $\lambda$  below ideal gas values.

As  $T$  is increased  $\lambda$  shortens and becomes less than typical pore volume diameters (e.g., at  $T = 1$  K,  $\lambda \sim 10$  Å). At liquid  $^4\text{He}$  densities  $\lambda$  is greater than the interatomic spacing (3.7 Å). At these densities and temperatures the dilute gas picture does not hold and confinement/disorder appears to be able to shorten or cutoff the wave function or otherwise reduce coherence (reduce  $T_c$ ) below bulk values. As noted the reduction of  $T_c$  depends on the confining media (the degree of confinement). Within a given medium, this could also be the case so that we have BEC in large pore volumes but not in necks between pore volumes or in small pore volumes. In a specific range of  $T$ , there could be BEC (phase coherence) localized in the larger pores separated by regions of normal fluid in necks and small volumes, much as in Josephson junction arrays. Macroscopic superfluidity across the whole sample would be observed at a lower  $T$  when the phase coherent regions overlap so there is phase coherence across the whole sample. This lower temperature defines  $T_c$ . At a higher temperature  $T > T_c$  there could be a BEC localized in the larger pores only, up to temperatures close to  $T_\lambda$  as in aerogel. In this picture, we observe well-defined excitations in neutron scattering measurements above  $T_c$  because there is BEC in the larger pores above  $T_c$ . Where there is BEC we expect to see well-defined phonon–roton excitations. Neutron scattering is a local probe and can observe localized BEC whereas a TO measurement requires phase coherence across the whole sample.

Localized BEC is discussed in models of disorder. Huang and Meng<sup>36</sup> consider a dilute Bose gas containing static ran-

dom disorder described by a white noise spectrum. The disorder depletes both the condensate and the superfluid density. At  $T = 0$  K the depletion of the superfluid density is greater than that of the condensate by a factor of 4/3 so that it is possible to have zero superfluid density and a finite condensate fraction. This effect is attributed to localization of the condensate or dragging of the superfluid by the quenched disorder.

Sheshardri *et al.*<sup>37</sup> consider a Hubbard model with on-site disorder. Within a mean field model, they find localized regions of phase coherence. The transitions to superfluidity is associated with the joining (percolation) of these localized regions so that there is phase coherence across the whole sample (see also Huang.<sup>38</sup>) Krauth *et al.*<sup>39</sup> have simulated bosons on a 2D lattice having site disorder and on-site inter-boson repulsion. For a given on-site repulsion, there is a critical value of the disorder at which the superfluid density goes to zero. The transition from finite to zero  $\rho_S(T)$  is associated with a transition to a disorder-localized phase (Bose glass or Mott localized phase).

Effects of localized superfluidity above  $T_c$  have been observed in aerogel.<sup>40</sup> For example, a fountain pressure above  $T_c$  has been observed which suggests a pressure arising from superfluidity above  $T_c$ , perhaps localized in larger pore regions but not extending across the whole sample.

In future work, we plan direct observation of the condensate in porous media by inelastic neutron scattering at high momentum transfers.

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