Dynamics of Liquid ^{4}He in Vycor

H. R. Glyde
Department of Physics and Astronomy, University of Delaware, Newark, Delaware 19716

O. Plantevin and B. Fåk
Département de Recherche Fondamentale sur la Matière Condensée, SPSMS/MDN, CEA Grenoble, 38054 Grenoble, France

G. Coddens
Laboratoire Léon Brillouin, CEA/CNRS, 91191 Gif-sur-Yvette Cedex, France

P. S. Danielson
Corning Incorporated, Sullivan Park FR-5, Corning, New York 14831

H. Schober
Institut Laue Langevin, BP 156, 38042 Grenoble, France
(Received 13 September 1999)

Using inelastic neutron scattering, we have observed well-defined phonon-roton (p-r) excitations in superfluid ^{4}He in Vycor over a wide wave-vector range, $0.3 \le Q \le 2.15$ Å^{-1}. The p-r energies and lifetimes at all temperatures 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 above $T_c = 1.952$ K, where $\rho_s(T) = 0$ in Vycor. 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.

PACS numbers: 67.40.–w

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 [1,2] to reveal the impact of disorder and finite length scales on excitations and phase transitions [3]. Understanding gained in helium can be transferred to other examples of bosons in disorder such as flux lines in superconductors [4], granular metal films [5], Cooper pairs in Josephson junction arrays [4], and (possibly) Cooper pairs in high-T_c materials if pairing occurs above the Bose-Einstein condensation (BEC) temperature that leads to superconductivity [6]. Direct measurement [7–12] and simulation [13–15] 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, $0.3 \le Q \le 2.15$ Å^{-1}, and temperature range, $0.5 \le T \le 2.31$ 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 [16]. 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 ± 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 μeV.

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

 0031-9007/00/84(12)/2646(4)$15.00

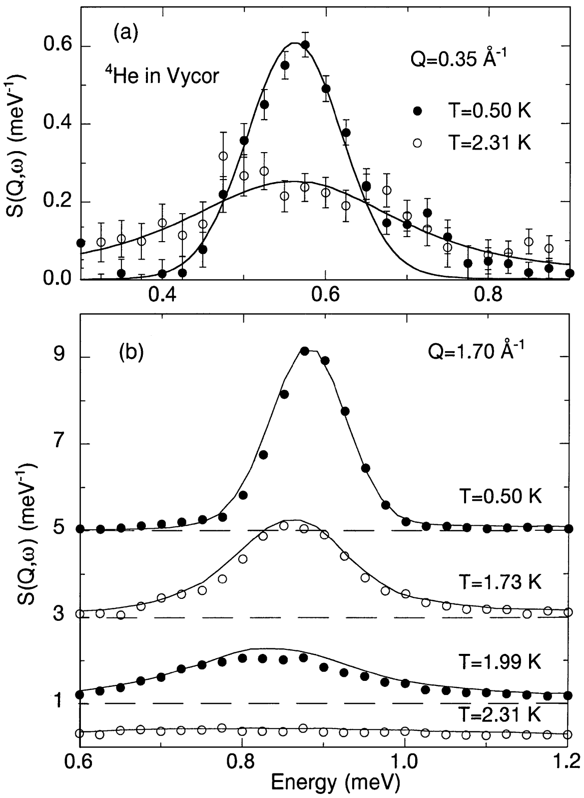

FIG. 1. Dynamic structure factor of liquid ^{4}He in Vycor at temperatures as shown. (a) $Q = 0.35$ Å^{-1} (phonon region). The lines are guides to the eye. (b) $Q = 1.7$ Å^{-1} (between the maxon and roton regions). Symbols are ^{4}He in Vycor, with errors smaller than the symbol size. The lines show bulk data for comparison.

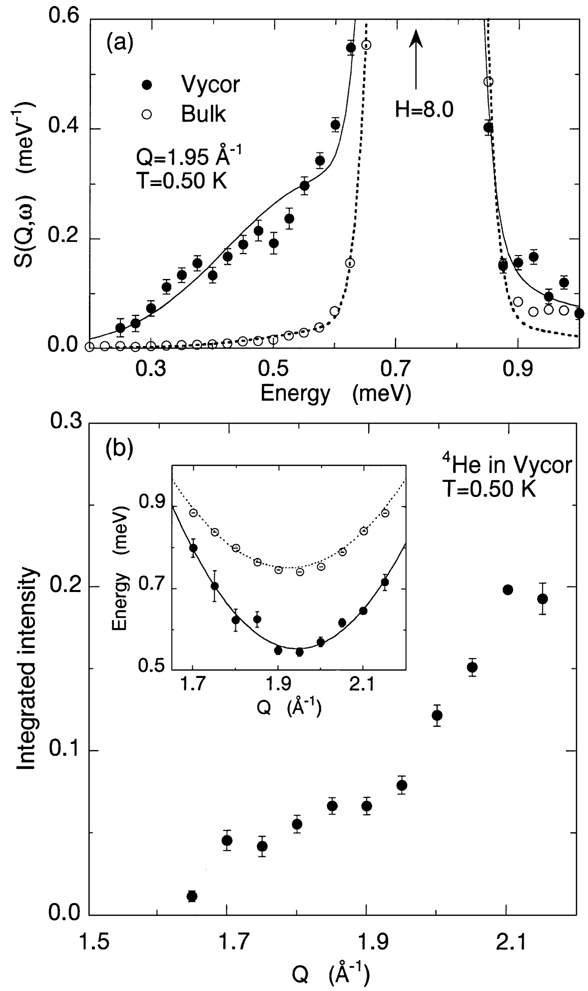

FIG. 2. (a) $S(Q,\omega)$ of liquid ^{4}He at $T = 0.5$ K in Vycor at the roton wave vector (solid circles and line). The dashed line is the component of $S(Q,\omega)$ arising from exciting the 3D roton in the liquid (peak height $H = 8.0$ meV^{-1}). The intensity at energies below the roton peak is 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).

($T = 0.5$ K) and normal ($T = 2.31$ 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 [17] (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 [19,20] 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 ω_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 (± 5 μeV).

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 [19,20]. 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

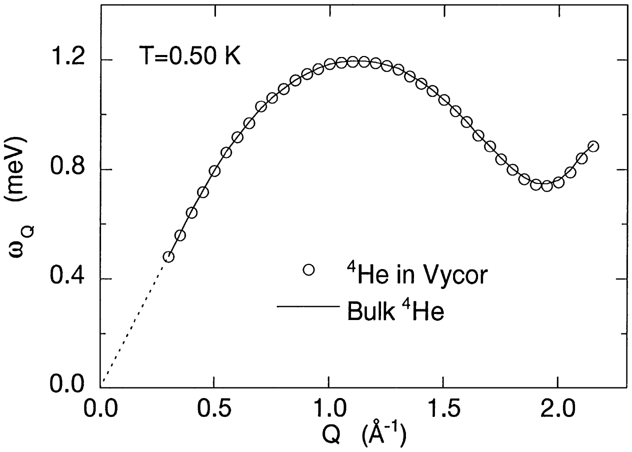

FIG. 3. Phonon-roton energy dispersion curve of liquid ^{4}He in Vycor (open circles) and in bulk ^{4}He (line). The error bars are much smaller than the symbol size.

Fig. 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 inset. 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 [21]. 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" Δ_{2D} is consistent with the gap energy 0.54 ± 0.03 meV of 2D rotons on graphon surfaces observed by Thomlinson *et al.* [22] and with that of 0.6 meV observed by Lauter *et al.* [23] on graphite surfaces. The difference between the 3D roton energy $\Delta = 0.742$ meV and Δ_{2D} is consistent with the differences predicted originally by Padmore [24] for 2D rotons and calculated more recently by Clements *et al.* [25]. 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.* [26] for the layer mode contribution to the specific heat in Vycor. Kiewiet *et al.* [27] found that the superfluid density $\rho_S(T)$ in Vycor for $T \leq 1.4$ K is well described if the normal density arises from exciting "one-dimensional" phonons and a rotonlike 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 $T \leq 1.4$ 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.* [27] 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.* [9] at an energy of 0.3–0.5 meV at the roton wave vector, although their intensity is 2 times greater (20% of the 3D roton). However, they could not determine the mode energy with precision.

As mentioned above, Woods and Svensson [18] noticed that the weight of the sharp p-r peak in the total $S(Q,\omega)$ in bulk liquid ^{4}He for $Q \gtrsim 1.1$ Å^{-1} scaled with temperature approximately as the superfluid density, $\rho_S(T)$. This approximate scaling was confirmed in subsequent measurements [19,20,28,29]. In particular, for $Q \gtrsim 1.1$ Å^{-1} there was no well-defined p-r excitation in normal ^{4}He where $\rho_S(T) = 0$. Glyde and Griffin [30] proposed that this could be understood if the sharp p-r excitation was associated with excitation of quasiparticles out of the condensate and the scaling followed $n_0(T)$ approximately rather than $\rho_S(T)$. In Fig. 1(b), which shows $S(Q,\omega)$ at $Q = 1.7$ Å^{-1}, we see that there is a broadened peak in $S(Q,\omega)$ in Vycor at $T = 1.99$ K, i.e., above $T_c = 1.952$ K, where $\rho_S(T) = 0$. The peak has disappeared by $T = 2.31$ K.

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:

$$\chi''(Q,\omega) = f_S(T)\chi''_S(Q,\omega) + f_N(T)\chi''_N(Q,\omega), \quad (1)$$

where $S(Q,\omega) = [1 - \exp(-\hbar\omega/k_BT)]^{-1}\chi''(Q,\omega)$. At $T = 0.5$ K, $\chi''_S(Q,\omega)$ 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 $\chi''_S(Q,\omega)$ 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 ρ_S/ρ 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

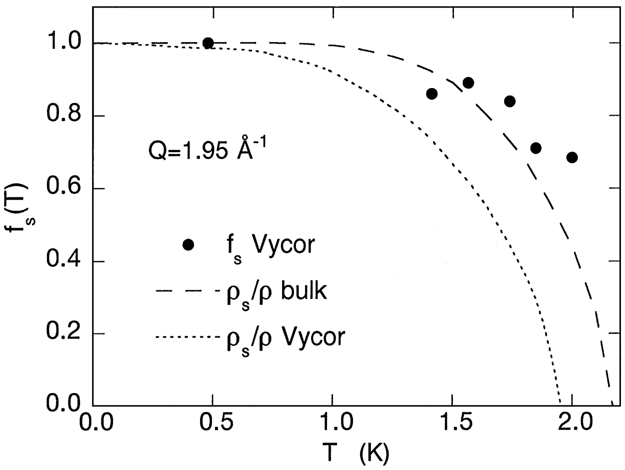

FIG. 4. Fraction $f_S(T)$ of the total $S(Q,\omega)$ that is taken up by the low-temperature component $S_S(Q,\omega)$ (chiefly the single-excitation component) as a function of temperature for Vycor [see Eq. (1)]. The $f_S(T)$ in Vycor does not scale with the superfluid fraction $\rho_S(T)/\rho$ of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

$\chi_N''(Q,\omega)$ is expected to be largely independent of T for $T \gtrsim T_\lambda$ (as it is in bulk liquid ^{4}He), the fitted $f_S(T)$ should not be sensitive to the temperature at which $\chi_N''(Q,\omega)$ 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) [30] proposed that the sharp excitation in $S(Q,\omega)$ at $Q \gtrsim 1$ Å^{-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_λ. If the latter is true, then we are observing the separation of the BEC temperature from the superfluid transition temperature (T_c) by disorder or confinement [4,31]. This intriguing possibility needs to be clarified by further measurements of $S(Q,\omega)$ between T_c and T_λ and direct measurements of n_0.

We thank the staff at the Institut Laue Langevin for technical assistance. This work was partially supported by the National Science Foundation, Grant No. DMR 96-23961.

[1] J. D. Reppy, J. Low Temp. Phys. **87**, 205 (1992).
[2] D. F. Brewer, in *The Physics of Liquid and Solid Helium*, Part II, edited by K. H. Benneman and J. B. Ketterson (Wiley, New York, 1978), p. 573.
[3] D. J. Thouless, Phys. Rep. **13C**, 93 (1974); M. P. A. Fisher *et al.*, Phys. Rev. B **40**, 546 (1989).
[4] G. Blatter *et al.*, Rev. Mod. Phys. **66**, 1125 (1994).
[5] M. C. Cha *et al.*, Phys. Rev. B **44**, 6883 (1991).
[6] M. Randeria, in *Bose Einstein Condensation*, edited by A. Griffin, D. Snoke, and S. Stringari (Cambridge University Press, Cambridge, England, 1995).
[7] J. De Kinder, G. Coddens, and R. Millet, Z. Phys. B **95**, 511 (1994); G. Coddens *et al.*, J. Non-Cryst. Solids **188**, 41 (1995).
[8] M. R. Gibbs *et al.*, Physica (Amsterdam) **213B–214B**, 462 (1995); P. E. Sokol *et al.*, Nature (London) **379**, 616 (1996); M. R. Gibbs *et al.*, J. Low Temp. Phys. **107**, 33 (1997); R. M. Dimeo *et al.*, Phys. Rev. Lett. **79**, 5274 (1997).
[9] R. M. Dimeo *et al.*, Phys. Rev. Lett. **81**, 5860 (1998).
[10] O. Plantevin *et al.*, Phys. Rev. B **57**, 10 775 (1998).
[11] C. R. Anderson *et al.*, Phys. Rev. B **59**, 13 588 (1999).
[12] R. T. Azuah *et al.*, J. Low Temp. Phys. **117**, 113 (1999).
[13] M. Makivić *et al.*, Phys. Rev. Lett. **71**, 2307 (1993).
[14] W. Krauth *et al.*, Phys. Rev. Lett. **67**, 2307 (1991); T. Giamarchi and P. Le Doussal, Phys. Rev. Lett. **76**, 3408 (1996).
[15] M. Boninsegni and H. R. Glyde, J. Low Temp. Phys. **112**, 251 (1998).
[16] M. J. Benham *et al.*, Phys. Rev. B **39**, 633 (1989).
[17] M. H. W. Chan *et al.*, Phys. Rev. Lett. **61**, 1950 (1988). [All Vycor is grown by Corning Inc. Measurements of T_c in Vycor grown with natural boron lie in the range $1.94 \leq T_c \leq 2.00$ K: cf. N. Mulders and J. R. Beamish, Phys. Rev. Lett. **62**, 438 (1989); G. M. Zassenhaus and J. D. Reppy, Phys. Rev. Lett. **83**, 4800 (1999). We assume that T_c is the same in the present sample grown with ^{11}B and having the same $S(Q)$.]
[18] A. D. B. Woods and E. C. Svensson, Phys. Rev. Lett. **41**, 974 (1978).
[19] H. R. Glyde, *Excitations in Liquid and Solid Helium* (Oxford University Press, Oxford, 1994); J. Low Temp. Phys. **93**, 349 (1993).
[20] A. Griffin, *Excitations in a Bose-Condensed Liquid* (Cambridge University Press, Cambridge, England, 1993).
[21] Unambiguous identification of the additional intensity as a layer mode will require measurements as a function of filling of the Vycor.
[22] W. Thomlinson *et al.*, Phys. Rev. Lett. **44**, 266 (1980).
[23] H. J. Lauter *et al.*, Phys. Rev. Lett. **68**, 2484 (1992); J. Low Temp. Phys. **87**, 425 (1992).
[24] T. C. Padmore, Phys. Rev. Lett. **32**, 826 (1974).
[25] B. E. Clements *et al.*, Phys. Rev. B **53**, 12 242 (1996).
[26] D. F. Brewer *et al.*, Phys. Rev. Lett. **15**, 182 (1965); D. F. Brewer, J. Low Temp. Phys. **3**, 205 (1970).
[27] C. W. Kiewiet, H. E. Hall, and J. D. Reppy, Phys. Rev. Lett. **35**, 1286 (1975).
[28] K. H. Andersen *et al.*, J. Phys. Condens. Matter **6**, 821 (1994); K. H. Andersen and W. G. Stirling, *ibid.* **6**, 5805 (1994).
[29] W. G. Stirling and H. R. Glyde, Phys. Rev. B **41**, 4224 (1990).
[30] H. R. Glyde and A. Griffin, Phys. Rev. Lett. **65**, 1454 (1990).
[31] K. Huang, in *Bose Einstein Condensation* (Ref. [6]), p. 31.