Some new fourth-sound investigations of finite-size effects in He II using a porous-glass matrix*

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The superfluid fraction ρ_s/ρ of He II in Vycor porous glass has been studied in the range (approximately) 1.6–2.0 K using fourth sound. There is a qualitative difference in the temperature dependence of ρ_s/ρ between this work and that obtained for He II in comparable packed powders.

Considerable experimental work has been published over the past few years on the measurements of ρ_s/ρ in "finite geometries." From such work, a number of conclusions appear fairly certain. These are (a) T_0, the superfluid onset temperature, is depressed increasingly as the size of the confining geometry is reduced, and (b) in "large" finite geometries (approximating the bulk limit), near T_λ, $\rho_s/\rho = \alpha(T_\lambda - T)^x$, where $\alpha \approx 1.44$ and $x \approx \frac{2}{3}$.[1,2]

The behavior (a) is in reasonably good quantitative agreement with semiphenomenological theories.[3,4] However, these theories assume the validity of a free-energy expansion in powers of an order parameter, and agreement with experiment which has been observed far outside the region of validity of the expansion must, at present, be regarded as fortuitous.

As for (b), scaling theory requires that x be $\frac{2}{3}$. Also, Patashinskii and Pokrovskii[5] have predicted an excitation spectrum $E = Ap^{3/2}$ for liquid helium near T_λ and, although the existence of this spectrum remains questionable, its incorporation in semiphenomenological theories is consistent with x being $\frac{2}{3}$.

We are reporting some investigations of finite-size effects in He II by fourth-sound techniques in which Vycor 7930 glass constitutes the porous matrix. Some preliminary results for Vycor were obtained by Brewer *et al.*[6] and further data of Fraser and Rudnick[7] disagreed with these. Although Fraser and Rudnick felt that Vycor was not as suitable as packed powder for fourth-sound measurements, it seems to us that Vycor has a great advantage over packed-powder systems with regard to regularity of structure. Powder systems inevitably have a wide pore-size distribution and large range of possible pore geometries, whereas Vycor 7930 is seen to be an array of randomly dense-packed silica balls which are roughly spherical and about 300 Å in diameter.[8] Thus, Brunauer-Emmett-Teller (B.E.T.) absorption measurements quoted by Watson,[8] in which cylindrical pores are assumed, give mean pore radii of 20.3 Å and 96% of the cylinder volume is within 3 Å of this mean. Now, while we feel that the cylindrical pore approximation is most inappropriate for Vycor, the adsorption measurements (and mercury intrusion measurements of other workers) do give a clear indication of the regularity of the system. Hence, we may expect that Vycor measurements should not "smear" size-dependent effects as drastically as packed-powder systems.

Space does not permit description of our apparatus in the present communication, but the following points should be noted.

(1) A cylindrical resonator configuration was employed and the Nylon resonator body and Vycor rod were well finished, in order that thermal shrinkage would "seal" the rod, eliminating "bulk paths" along the walls and the resulting spurious resonances.

(2) The Vycor rod was cleaned in 30% H_2O_2 and outgassed in a vacuum oven. After the oven was cooled, dry helium gas was admitted at 1.5 atm, and the rod was loaded into the resonator via a break-seal bulb and a glove box. The Vycor was then maintained under high vacuum until performance of a run.

We feel that water vapor, which is very readily adsorbed by Vycor, and perhaps some air, can, unless excluded, form a coating on the pore walls. This coating, by changing the pore size (and the nature of the substrate), can give rise to "spurious" results.

Figure 1 shows data obtained in three cases. The first case is that in which runs (points undifferentiated here) were performed on specimens which had been transferred to the resonator without filling them with helium or using the glove box. The second case is that in which the full procedure was followed (two runs—points undifferentiated again). In the third case, only the use of the glove box was omitted (one run).

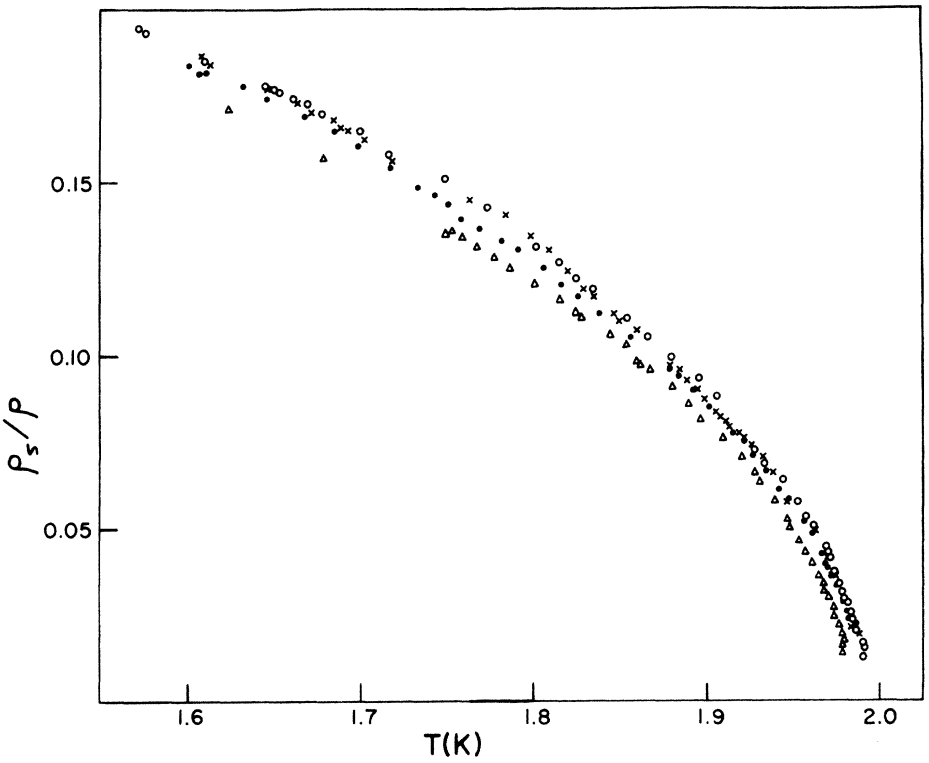

FIG. 1. ρ_s/ρ vs T data of present work. Open triangles, case 1, no helium-gas fill or glove-box stage; open circles and × crosses, case 2, full cleaning and transfer procedure; closed circles, case 3, helium-gas fill, but no glove-box stage.

It is interesting that data for the first case is consistent from run to run, suggesting that, despite different adsorption of water under different humidities, the layer remaining after pumping may be the same. The data for the third case might indicate that the "coating" of pores was considerably inhibited by filling the Vycor with helium gas before removing it from the oven.

The superfluid fraction was determined using the relation $\rho_s/\rho = C_4^2/C_1^2$, where C_1, C_4 are the first- and fourth-sound velocities, respectively. This relation is believed to be accurate to better than 1% in the temperature range we investigated, although recent work by Racz[9] suggests that this simple relation might not hold in systems with larger pores. In calculating C_4, a multiple-scattering correction factor $[\frac{1}{2}(3-P)]^{1/2}$ (where P is the porosity) was used. This relation, cited in Ref. 10, seems most appropriate for the Vycor structure (randomly dense-packed spheres) and takes a value of 1.16 for Vycor.

In Fig. 2 representative points from the data of various workers who have used Vycor 7930 in their investigations are plotted in such a way as to scale to our data at $T = 1.60$ K. Pobell *et al.*[11] measured the persistent angular momentum of He II in a Vycor ring. This should be proportional to ρ_s. Their data cover the temperature range 0.65–1.60 K and appears to complement our data very well in giving a continuous curve from 0.65 K to near T_0. The replotted data of Fraser and Rudnick[7] appears to give a lower value of T_0 (about 1.95 K) than ours. In their paper, they quote T_0 as being between 2.05 and 2.10 K on the basis of comparison with their packed-powder work. Packed powders seem[12] to exhibit an inflection in the ρ_s/ρ curve near T_0. Although there is some theoretical basis for this behavior from considera-

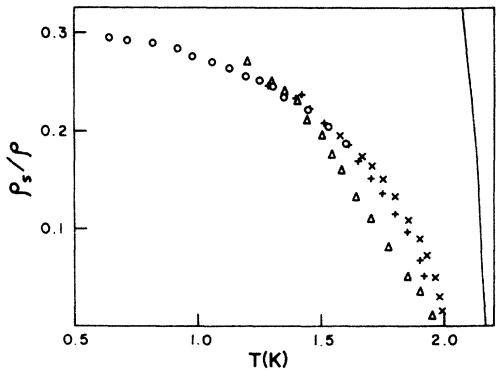

FIG. 2. Comparison of some pertinent ρ_s/ρ vs T data. (For clarity, a set of representative points is presented.) × crosses, present experiment "clean" Vycor 7930; open circles, Pobell *et al.* (Ref. 11) Vycor 7930; upright crosses, Fraser and Rudnick (Ref. 7) Vycor 7930; open triangles, Kriss and Rudnick (Ref. 1), a packed powder for which $T_0 = 2.040$ K. Solid line is curve for bulk HeII (Data of Pobell *et al.* and Fraser and Rudnick rescaled.)

tion of finite Bose-Einstein systems, it is more likely that much of the inflection observed is a result of the wide pore-size distribution of packed powders. We find no evidence in the present work of an inflection with Vycor, which has a narrow pore-size distribution, and think it inappropriate to incorporate an inflection in our extrapolation to $\rho_s/\rho = 0$.

At $T \to 0$, ρ_s/ρ in Vycor appears to tend to a value of about 0.31. Kriss and Rudnick[1] give ρ_s/ρ vs T data for a packed-powder–copper-wool system with T_0 and zero-temperature superfluid fraction similar to that in Vycor. Despite inaccuracies inherent in transcribing this data from a figure in a journal, one may observe in Fig. 2 that the powder curve lies well below and is more linear than the Vycor curve. Examination of curves for powders with the smallest pores yet used, in Refs. 1 and 12, suggests to us that linearity near T_0 may be a property of curves for such systems.

Although the transition in a very small system probably has no well-defined onset temperature, we can take a temperature "characterizing" the transition by linearly extrapolating the ρ_s/ρ curve to zero. The "T_0" value so obtained is 1.998 ±0.005 K. If we calculate a "pore diameter" from either the Ginzburg-Pitaevskii[3] or Mamaladze[4] theories by substituting the 0.174 K onset shift into the relevant expressions, we obtain values of 47 and 43 Å, respectively. Now these values are both close to the estimated "mean neck diameter" for Vycor pores of 40 Å as determined by mercury intrusion porosimetry, but, as previously mentioned, close experimental agreement with the above theories would seem to have little real significance.

Maki and Guyon[13] developed a free-energy expansion approach to finite-size effects in He II in which the order-parameter fluctuations, which are so important near a phase transition, are included. With this approach, our onset shift corresponds to a pore diameter of 28 Å after modification to cylindrical geometries. However, the most useful feature of the Maki-Guyon theory is that one may hope to obtain a theoretical expression for ρ_s/ρ as a function of temperature. [The expression given for $\rho_s(T_\lambda - T)$ in the paper would appear to diverge for "thin-film" geometries with boundary separation of greater than roughly 20 Å. However, this problem does not seem insuperable.]

Work is now in progress to measure ρ_s/ρ in Vycor of various mean pore sizes and, in the absence of adequate microscopic theories, the results of this investigation could, at any rate, facilitate a realistic experimental test of the best available free-energy expansion theories.

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