ment, to Mr. Wolf Bank and Mr. Willy Grothe for invaluable technical assistance, and to Professor George Uhlenbeck for encouragement throughout the experiment.

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*Work supported in part by the National Science Foundation under Grant No. GK-4708 and in part by the University of Colorado Computing Center.

[1] J. D. Foch, Jr., G. E. Uhlenbeck, and M. Fuentes Losa, to be published.

[2] M. Greenspan, J. Acoust. Soc. Amer. 28, 644 (1956).

[3] E. Meyer and G. Sessler, Z. Phys. 149, 15 (1957).

[4] G. J. Prangsma, R. M. Jonkman, and J. J. M. Beenakker, Physica (Utrecht) 48, 323 (1970).

Quartz Microbalance Studies of an Adsorbed Helium Film*

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(Received 7 June 1972)

The resonant frequency of a quartz crystal oscillating in the thickness-shear mode changes by virtue of the mass loading due to any type of film laid down on its surfaces. For a helium film only the normal fraction remains rigidly coupled to the substrate motion. Adsorption isotherms of He^4 measured with this technique show clear departures from conventional nonsuperfluid behavior. We attribute this effect to the onset and presence of superfluidity in the film.

There is considerable interest in the question of the disappearance of superfluidity in very thin films of liquid helium.[1-4] In particular it has been well established[5] that in sufficiently thin films superflow disappears even though there remains a finite superfluid fraction in the film. The point at which the superfluid fraction itself disappears has been open to some conjecture. It is to this question of the direct measurement of ρ_s/ρ in thin films that this experiment pertains.

The resonant frequency of a quartz crystal oscillating in the thickness-shear mode is lowered by virtue of the mass loading due to any film laid down on its surfaces.[6-8] In particular, under the conditions which obtain in our experiments, this change in the frequency of the crystal, Δf, is given by the formula

$$-\Delta f = 2(2f^2/c\rho_q)\sigma. \qquad (1)$$

Here σ represents that part of the adsorbed mass per unit area which remains rigidly coupled to the substrate motion. f is the resonant frequency,[9] c is the shear-wave velocity in the crystal, and ρ_q is the density of the quartz. The extra factor of 2 allows for loading on both of the two faces of the quartz crystal. The validity of applying this formula to a liquid-helium film is based on the fact that for the regimes of interest in our experiments we find (employing the normal fluid viscosity η and density ρ_n) that the viscous wave length $\lambda = (\eta/\pi\rho_n f)^{1/2}$ always far exceeds the thickness of the films with which we work, except perhaps at relative saturations well in excess of 99%.

By virtue of this relationship we have measured the mass adsorbed as a function of the helium gas pressure P in our experimental chamber up to the saturation vapor pressure $P_0(T)$ for a series of temperatures. The resulting adsorption isotherms $[\sigma$ versus $P/P_0(T)]$ for liquid He^4 show a characteristic point of departure from that expected from a nonsuperfluid. Figure 1 exhibits several representative isotherms obtained by employing this technique on liquid-He^4 films. By comparison, Fig. 2 represents an isotherm of liquid He^3 obtained at $T = 1.356$°K with exactly the

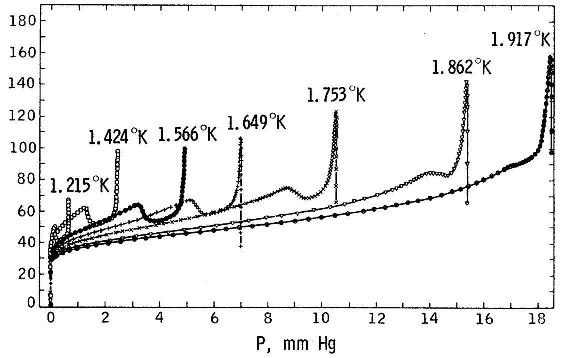

FIG. 1. Representative experimental data on the frequency changes observed for different helium gas pressures over the range of temperatures explored.

same apparatus. We attribute the evident difference in behavior between these two systems to the superfluid fraction in the He^4 film. This fraction remains uncoupled to the substrate and hence does not load the quartz crystal as does a normal film.

If we take as the onset of superfluidity those points in Fig. 1 at which the adsorption isotherm curve begins to turn away from a monotonic increase, our onset points fall as shown in Fig. 3. The dashed curves in Fig. 3 represent the limits between which past reported results[5] lie and the full curve represents some mean of these results. The fact that our points generally fall slightly lower than this curve suggests that we are seeing earlier into the commencement of superfluidity than heretofore reported.

We employed very thin quartz crystals, approximately 0.07 mm thick and 6.3 mm in diameter, of the Y-cut family, $\varphi = 39°49'$, with a resonant frequency of about 24 MHz. For this cut the resonant frequency is essentially stationary with respect to temperature[10] below 4.2°K. The stability of 0.1 Hz/h of the system was achieved via this fact and by directly immersing the oscillator circuit in the cryogenic bath and connecting it to the crystal in the adsorption cell via a hermetically sealed feedthrough over the short distance of 2 cm. The crystal surfaces were optically polished and over these surfaces were vacuum deposited either gold or aluminum electrode disks 3.0 mm in diameter and between 1000 to 1500 Å in thickness. The crystals were mounted with minimum constraint to allow for thermal contractions, and they were in good thermal contact with the bottom plate of the adsorption cell. This cell was made of oxygen-free high conductivity (OFHC) copper and totally immersed in the cryogenic bath.

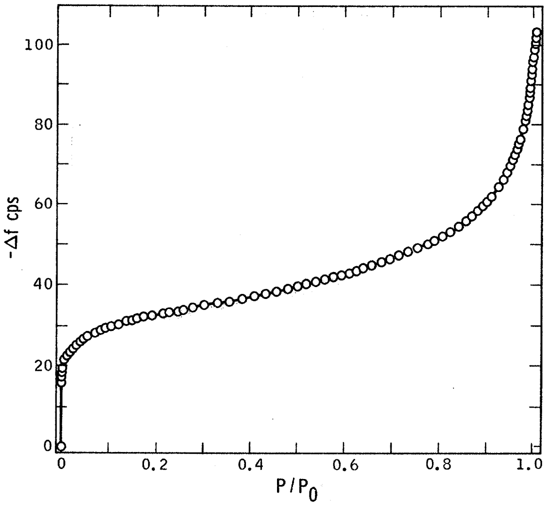

FIG. 2. Experimental adsorption isotherm of He^3 at $T = 1.356°K$, employing exactly the same apparatus as that used for the He^4 observations of Fig. 1.

The total thermal conductance from the crystal to the helium bath is estimated approximately to be 5×10^{-3} W/°K at liquid-helium temperatures. Hence for the crystal power dissipation of less than 2 μW allowed in this experiment the local film temperature cannot have risen more than 400 μdeg above the ambient. Therefore the thermal conduction increase associated with the commencement of superfluid behavior could not have made a significant contribution to temperature changes in the film. The maximum velocity of the quartz surfaces corresponding to a 2 μW drive level is estimated[7] as being approximately

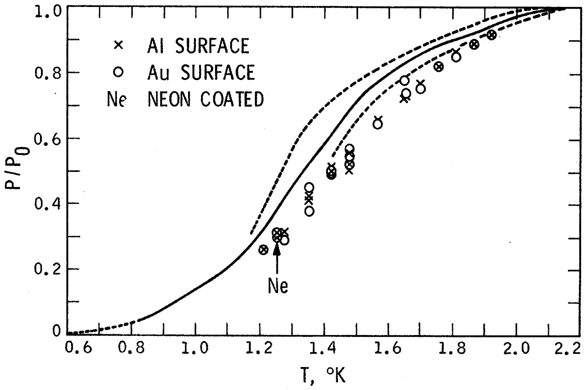

FIG. 3. Onset of superfluidity as a function of temperature. The points shown represent our results for three different substrates. (The point marked "Ne" was was obtained with neon substrates over the Au and Al electrodes.) The dashed curve represents the limits between which past reported results lie. (Cf. Fig. 18 of Ref. 5.)

0.08 cm/sec, which is well below the critical velocity in a helium film. Hence both mechanisms are eliminated as possible explanations for the relatively abrupt onset of superfluidity observed in these experiments.

Two independent quartz-crystal channels were monitored simultaneously, the aluminum electrode one and the gold electrode one. No significant difference in data was observable. Measurements both in adsorption and desorption were found to be coincident over essentially the entire range explored. Only at pressures in the range of 10^{-5} Torr or less do we find a difference between adsorption and desorption. At this low pressure, however, only the adsorption is physically meaningful by virtue of experimental practicalities.[11] One cannot desorb all of the residual helium gas condensed at liquid-helium temperatures. However, in adsorption one starts with a vacuum prepared at higher temperatures without any residual helium submonolayer. Another unusual aspect exhibited in Fig. 1 is that for temperatures greater than about 1.64°K the adsorption signal appears to turn around and actually decrease after the pressure approaches within about a percent of the saturation pressure $P_0(T)$. One expects the signal, $-\Delta f$, to increase indefinitely at $P_0(T)$. For the moment this behavior remains somewhat of a mystery. Other researchers[12,13] investigating this regime have also reported anomolous results there. Hence we simply offer our data for public scrutiny without further comment on these effects until we gain further insight into them. These anomalous effects occur only within a few percent of saturation. Our interest centers on the superfluidity onset. The latter occurs at much lower saturation fractions except in the vicinity of the λ point.

Figure 4 is a plot of an observed adsorption signal, $-\Delta f$, against $[\log P_0(T)/P]^{-1/3}$ at a temperature 1.649°K. On the basis of the Frenkel-Halsey-Hill theory[14] this should reflect the behavior of the nonsuperfluid mass adsorbed as a function of the thickness D of the film in the following manner:

$$D/D_0 = \{\theta/[T\ln P_0(T)/P]\}^{1/3}. \tag{2}$$

Here D/D_0 represents the "number of film layers," with D_0 currently[15] taken as 3.6 Å. The characteristic temperature θ measures the strength of the Van der Waals force between the wall and the helium atoms.

For relative saturations $P/P_0(T)$ between 0.1 and 0.72, before the onset of superfluidity, a

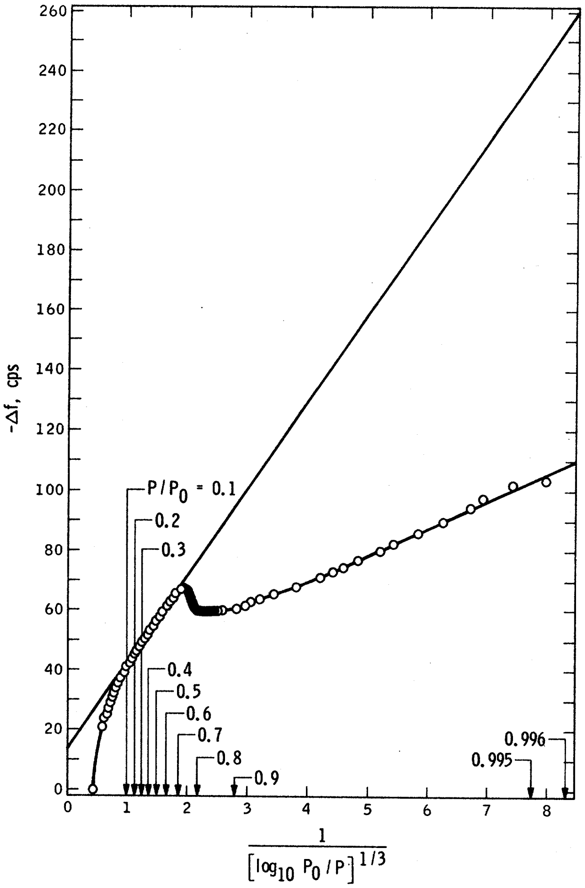

FIG. 4. Experimental adsorption signal at $T = 1.649$°K, plotted in Frenkel-Halsey-Hill fashion as a function of $(\log P_0/P)^{-1/3}$ to illustrate the method of obtaining the Van der Waals strength θ and the normal-fluid fraction as a function of film thickness. The absisca is proportional to the film thickness.

linear regime is observed as expected. The slope of the extrapolated line drawn through this regime should reflect the bulk total helium density ρ because at these levels of saturation one expects

$$d\sigma/dD = \rho_{\mathrm{bulk}} = 0.146\ \mathrm{g/cm^3}, \quad 0.1 < P/P_0 < 0.7, \quad T = 1.649\text{°K}. \tag{3}$$

Employing (2), the sensitivity of our instrument as obtained via Eq. (1) $[d\sigma/d(-\Delta f) = 3.9\times10^{-10}$ g/cm^2 Hz$]$, and the notion embodied in (3), one finds $\theta = 39 \pm 6$°K, a value quite consistent with other estimates for this number.[15]

Our experimental data points in Fig. 4 fall below the extrapolated total fluid density line discussed above for relative saturations in excess of 72%. As mentioned, we attribute this to the elimination of the superfluid fraction from the film-mass loading effect. Hence we expect our data points to extrapolate to a straight line of lower slope $d\sigma_n/dD = \rho_n$(bulk). As one can see from the figure, this is, in fact, the case. Using this notion one calculates a residual superfluid fraction of about $(75 \pm 3)\%$ whereas the tabulated vapor pressure bulk value is 80%. Considering the fact that the whole of the abscissa of Fig. 4 only spans about 30 atomic layers, we consider our results in remarkably good agreement with expectations and accept them as further evidence that we are indeed making direct measurements of the superfluid fraction in very thin films.

We wish to thank Dr. C. D. Stockbridge, Dr. William H. King, Jr., and Mr. D. R. Montgomery for some helpful discussions on the quartz microbalance. Credit goes to Mr. S. C. Choudhery for the original construction of the cryogenic adsorption probe and we wish to acknowledge useful discussions with Professor I. Rudnick.

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*Work supported by a grant from the California Institute of Technology President's Fund.

[1] R. S. Kagiwada, J. C. Fraser, I. Rudnick, and D. Bergman, Phys. Rev. Lett. 22, 338 (1969).

[2] R. P. Henkel, G. Kukich, and J. D. Reppy, in *Proceedings of the Eleventh International Conference on Low Temperature Physics, St. Andrews, Scotland, 1968,* edited by J. F. Allen, D. M. Finlayson, and D. M. McCall (St. Andrews University Press, St. Andrews, Scotland, 1969).

[3] R. P. Henkel, E. N. Smith, and J. D. Reppy, Phys. Rev. Lett. 23, 1276 (1969).

[4] D. F. Brewer and K. Mendelssohn, Proc. Roy. Soc., Ser. A 260, 1 (1961).

[5] K. R. Atkins and I. Rudnick, in *Progress in Low Temperature Physics,* edited by C. J. Gorter (North-Holland, Amsterdam, 1970), Vol. VI, Chap. 2.

[6] J. B. Brown and M. G. Tong, in *Proceedings of the Twelfth International Conference on Low Temperature Physics, Kyoto, Japan, 1970,* edited by E. Kanda, (Keigaku Publishing Co., Tokyo, 1971), p. 113.

[7] C. D. Stockbridge, in *Vacuum Microbalance Techniques,* edited by K. H. Behrndt (Plenum, New York, 1966), Vol. 5, pp. 147–205.

[8] G. Sauerbrey, Z. Phys. 155, 206 (1959).

[9] Since we employ a Colpitt-Clapp type of oscillator circuit, the crystal operates in a parallel resonance mode shunted by approximately 20 pF and the resonant frequency of the ensemble is essentially the natural one of the quartz crystal itself.

[10] P. A. Simpson and A. H. Morgan, National Bureau of Standards Report No. 6080, 1959 (unpublished).

[11] J. P. Hobson, Can. J. Phys. 37, 300 (1959).

[12] R. Bowers, Phys. Rev. 91, 1016 (1953).

[13] D. F. Brewer and K. Mendelssohn, Proc. Roy. Soc., Ser. A 260, 1 (1961).

[14] D. M. Young and A. D. Crowell, *Physical Adsorption of Gases* (Butterworths, London, 1962).

[15] The value $\theta \simeq 87°$K has been employed in the work of Rudnick *et al.* [I. Rudnick, R. S. Kagiwada, J. C. Fraser, and E. Guyon, Phys. Rev. Lett. 20, 430 (1968)], but measurements of C. H. Anderson and E. S. Sabisky [Phys. Rev. Lett. 24, 1049 (1970)] yielded $\theta = 16°$K. Cf. also R. Bowers, Phil. Mag. 44, 485 (1953).

Positive Impurity Ions in He II†

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(Received 24 April 1972)

We have developed a technique for introducing a variety of impurity atomic ions into liquid helium and measuring their mobility. We find that, in contrast with predictions of the "snowball" model of Atkins, the ion mobility depends on the atomic number of the core ion in a way that cannot be explained simply in terms of the mass of the ion. The relative mobility of ^{40}Ca and ^{48}Ca ions has been measured and is consistent with existing theories.

For many years ions have served as useful tools in investigations involving liquid helium. The drag force encountered by an ion in motion through the superfluid exhibited the energy spectrum of the elementary excitations.[1] Ions have been used to generate quantized vortex rings,[2] and studies of the interactions between ions and vortex lines provided useful information about quantized vorticity.[3]

The nature of the ions themselves has been the subject of considerable research. The observation of the energy well seen by the electron in its bubble,[4] and the explanation of the low-temperature mobility in terms of resonant oscillations of the bubble surface,[5] have provided strong evidence for the correctness of the bubble model for