warmed to  $\approx 270$  °K in order to initiate a crosschannel jump. After cooling to below 50 °K, the atom appearing in the adjacent channel is pulse desorbed and its charge-to-mass ratio is measured. It is apparent from the data in Fig. 3 that after a cross-channel event, an iridium atom rather than the tungsten atom initially deposited on the surface is present in the adjacent channel.

Our measurements thus lead us to conclude that for tungsten atoms on Ir(110), cross-channel motion occurs by exchange of the tungsten adatom with an atom from the substrate, presumably from the protruding rows forming the channels. If this is indeed what happens, then we should also expect to find a tungsten atom incorporated in the substrate on analyzing the first lattice layer. These measurements have been done. After cross-channel motion of a tungsten adatom and subsequent field evaporation of the adatom remaining on the surface we do indeed find tungsten incorporated in the lattice. In a comparable number of blank runs on the substrate, tungsten was never detected.

Several other observations, which will be reported fully elsewhere, also support the conclusion that cross-channel motion of tungsten atoms on the (110) plane of iridium occurs by exchange with lattice atoms. We presume that a similar mechanism accounts for channel crossings in self-diffusion on Ir(110), and also on Pt(110). It is of interest that for both iridium<sup>8</sup> and platinum,<sup>9, 10</sup> LEED measurements on (110) yield a  $1 \times 2$  pattern suggesting a channel spacing twice the normal value. In contrast, for rhodium (110), no rearrangement is indicated by LEED data,<sup>11, 12</sup> and neither are there any indications of crosschannel diffusion.<sup>13</sup> This suggests that the forces responsible for the restructuring of the (110) plane may also favor cross-channel motion by an exchange mechanism.

We are indebted to R. S. Chambers for expert assistance with the atom probe. The work reported here was supported by the National Science Foundation under Grant No. DMR 77-23723; the Coordinated Science Laboratory is operated under Contract No. N00014-79-C-0424 from the Joint Services Electronics Program.

<sup>1</sup>For reviews of early measurements, see G. Ehrlich, Surf. Sci. <u>63</u>, 422 (1977) or D. W. Bassett, Surf. Sci. 53, 74 (1975).

53, 74 (1975). <sup>2</sup>D. W. Bassett and P. R. Webber, Surf. Sci. <u>70</u>, 520 (1978).

<sup>3</sup>J. W. Wrigley and G. Ehrlich, to be published.

<sup>4</sup>T. Halicioglu, Surf. Sci. <u>79</u>, L346 (1979).

<sup>5</sup>E. W. Müller and T. T. Tsong, *Field Ion Microscopy* (American Elsevier, New York, 1969).

<sup>6</sup>For a general description of our atom probe, see R. S. Chambers and G. Ehrlich, J. Vac. Sci. Technol. <u>13</u>, 273 (1976). More details are given by R. S. Chambers, University of Illinois at Urbana-Champaign, Coordinated Science Laboratory Report No. R-702, 1975 (unpublished).

<sup>7</sup>C. -M. Chan, K. L. Luke, M. A. Van Hove, W. H. Weinberg, and E. D. Williams, J. Vac. Sci. Technol. <u>16</u>, 642 (1979).

<sup>8</sup>K. Christmann and G. Ertl, Z. Naturforsch. A <u>28</u>, 1144 (1973).

<sup>9</sup>H. P. Bonzel and R. Ku, J. Vac. Sci. Technol. <u>9</u>, 663 (1972).

<sup>10</sup>C. M. Comrie and R. M. Lambert, J. Chem. Soc. Faraday Trans. 1. <u>72</u>, 1659 (1976).

<sup>11</sup>C. W. Tucker, J. Appl. Phys. 37, 4147 (1966).

<sup>12</sup>R. A. Marbrow and R. M. Lambert, Surf. Sci. <u>67</u>, 489 (1977).

<sup>13</sup>G. Ayrault and G. Ehrlich, J. Chem. Phys. <u>60</u>, 281 (1974).

## Ripplons, <sup>3</sup>He, and Heat Conduction on the Surface of Superfluid <sup>4</sup>He

I. B. Mantz, D. O. Edwards, and V. U. Nayak Physics Department, Ohio State University, Columbus, Ohio 43210 (Received 11 December 1979)

A gradient in the concentration of adsorbed  ${}^{3}$ He, produced by a current of ripplons, has been observed in heat-conduction measurements on the surface of superfluid  ${}^{4}$ He. The  ${}^{3}$ He spreading pressure and  ${}^{3}$ He-ripplon collision rate have been determined from the data.

According to the phenomenological theory of Andreev and Kompaneets<sup>1</sup> the free surface of superfluid <sup>4</sup>He is governed by a two-dimensional (2D) version of the classic Landau theory of superfluidity. "Ripplons" (quantized capillary waves) are the elementary excitations of the sur-

© 1980 The American Physical Society

face, in analogy to phonons in the bulk liquid. <sup>3</sup>He may be added to form quasiparticles which are bound to the surface, and which, at low <sup>3</sup>He concentration, behave like a 2D Fermi gas. The 2D hydrodynamic equations of the surface are similar to the 3D, two-fluid equations in bulk helium, but with a reduced number of kinetic coefficients. So far none of the 2D kinetic coefficients have been measured.<sup>2</sup>

In this Letter we describe experiments on the surface thermal conductivity and, in particular, the effect of very small concentrations of adsorbed <sup>3</sup>He. The heat is carried by "two-fluid convection" in which a hydrodynamic flow of ripplons produces a heat current

$$\vec{q} = S_R T \vec{v}_R , \qquad (1)$$

with a corresponding opposing flow of superfluid. In (1),  $\bar{\mathbf{v}}_R$  is the drift velocity of the ripplon gas, assumed to be in internal equilibrium via the three-ripplon process.<sup>3</sup> The ripplon entropy per unit area  $S_R$  is  $-d\alpha_R/dT$ , where  $\alpha_R = -6.5 \times 10^{-3} \times T^{7/3}$  erg cm<sup>-2</sup> is the ripplon contribution to the surface tension.<sup>2</sup>

With small amounts of <sup>3</sup>He on the surface we observe an effect whose 3D analog is well known, the "heat flush", in which the heat flow produces a <sup>3</sup>He concentration gradient. By determining when the heat flush is complete, i.e., when all <sup>3</sup>He has been swept off the surface, we can measure the spreading pressure of the adsorbed <sup>3</sup>He with extremely high sensitivity. Analysis of the conductivity data gives the <sup>3</sup>He-ripplon scattering time  $t_{3R}$  and its temperature dependence.

To make evaporation, and conduction by phonons, negligible, the measurements<sup>4</sup> were made at low temperatures and on a saturated helium film. The substrate is a vertical, free-standing, Mylar sheet, 13  $\mu$ m thick, 4.1 cm wide, and 10 cm high. There is a heater at the top, and two thermometers, Acheson DAG 580 colloidal graphite films, at heights of 4.2 and 7.2 cm. The bottom of the sheet is in thermal contact with the copper cell. With enough liquid to give a saturated film, the free surface of all the film in the cell has an area  $A = 1.28 \text{ m}^2$  for the adsorption of <sup>3</sup>He. This was determined in earlier experiments<sup>2</sup> using a capacitor to measure the reduction in the surface tension by large concentrations of <sup>3</sup>He. Before cooling, the cell was flushed with neon so that the substrate of the film is actually neonplated Mylar. The thickness of the saturated <sup>4</sup>He film was measured by flash evaporation from a heater.<sup>4</sup> It is approximately 100 Å at the height

of the thermometers, in agreement with the van der Waals field of neon.<sup>5</sup> The thickness is large enough for corrections to the bulk values of  $\alpha_R$  and  $S_R$  to be very small.

Figure 1 shows the measured thermal conductance between the thermometers, as a function of their mean temperature T. The data are for the substrate alone, the substrate with a saturated film of specially purified <sup>4</sup>He, then the <sup>4</sup>He film and substrate with two different amounts of adsorbed <sup>3</sup>He. When divided by A the latter correspond to mean <sup>3</sup>He surface densities  $\overline{N}_3^{s}$  of 2.27  $\times 10^{12}$  and  $8.82 \times 10^{12}$  cm<sup>-2</sup>. The data were taken with the cell and the bottom of the Mylar at various constant temperatures  $T_0$ , low enough for all the <sup>3</sup>He to remain on the surface. The 2D thermal conductivity of the He film is found by subtracting the conductance of the Mylar, then multiplying by the ratio of the thermometer separation to twice the width of the Mylar. For the Mylar and for the pure <sup>4</sup>He film the conductivity is accurately independent of the heat current (verified over one order of magnitude) but with <sup>3</sup>He, because of the heat flush, it depends on  $\overline{q}$  and therefore on  $T_0$ .

Let us first consider the conductivity of the pure <sup>4</sup>He surface,  $K_4(T)$ . In bulk <sup>4</sup>He the conduction of heat by two-fluid convection is limited by



FIG. 1. Thermal conductance of neon-plated Mylar (hexagons); of Mylar covered with saturated pure <sup>4</sup>He film (solid squares); then <sup>4</sup>He with two mean surface concentrations of <sup>3</sup>He,  $\overline{N}_3^{s}$ , and various cell temperatures  $T_0$ :  $\overline{N}_3^{s} = 2.27 \times 10^{12}$  cm<sup>-2</sup>,  $T_0 = 22$  mK (open circles);  $T_0 = 100$  mK (open squares) and  $T_0 = 125$  mK (open triangles);  $\overline{N}_3^{s} = 8.82 \times 10^{12}$  cm<sup>-2</sup>,  $T_0 = 22$  mK (solid diamonds) and  $T_0 = 101$  mK (solid triangles).

the viscosity of the normal fluid  $\eta_n$  and the boundary condition  $\vec{v}_n = 0$  at the walls of the channel. The free surface, on the other hand, has no boundaries and the momentum loss per unit area,  $\nabla \alpha = -S_R \nabla T = (S_R/K_4)\vec{q}$  for the pure <sup>4</sup>He surface, must be transferred to the Mylar substrate. We note from (1) that  $\nabla \alpha$  and  $\vec{v}_R$  are linearly related:

$$\nabla \alpha = (S_R/K_4)\vec{\mathbf{q}} = (S_R^2 T/K_4)\vec{\mathbf{v}}_R.$$
 (2)

The mechanism for momentum transfer to the substrate is not yet clear, but one idea is as follows. Ripplons of energy near  $k_B T$  are confined within about 20 Å of the surface, since this is their average inverse wave number  $k^{-1}$ . However, they continually emit and absorb ripplons of much smaller k by the three-ripplon process.<sup>3</sup> The velocity field of the low-k ripplons penetrates the full thickness of the film and so they can create or absorb phonons in the substrate. The momentum transfer and the conductivity are expected to vary with the film thickness, and from one Mylar sample to another. This is observed experimentally,<sup>4</sup> although there is no quantitative theory to compare with the data.

In analyzing the conductivity with <sup>3</sup>He on the surface we assume that, for a given  $\overline{q}$ , the momentum transfer to the substrate is the same as for the pure <sup>4</sup>He film. This is reasonable provided that all the heat is transported convectively by the ripplons, so that (1) remains valid. (Note that in the steady state the <sup>3</sup>He drift velocity is zero.) We thereby obtain a relation between  $\nabla \alpha_3$ , the gradient in the <sup>3</sup>He contribution to the surface tension  $(-\alpha_3$  is the <sup>3</sup>He spreading pressure), and the heat current. Using  $\nabla \alpha = \nabla \alpha_3 + \nabla \alpha_R$  and (2), we find

$$\nabla \alpha = \nabla \alpha_3 - S_R \nabla T = (S_R / K_4) \mathbf{\bar{q}}.$$
 (3)

So

$$\nabla \alpha_3 = S_R (1 - K/K_A) \nabla T, \qquad (4)$$

where  $K(T, T_0)$  is the conductivity with <sup>3</sup>He. In Fig. 1 we see that K becomes equal to  $K_4$  when the thermometers are sufficiently hot  $(T > T_s \text{ say})$ compared to the cell temperature  $T_0$ ; this happens when the heat current is large enough for all the <sup>3</sup>He to be swept clear from the space between the thermometers. Integrating (4) between  $T_0$  and  $T_s$ , using the empirical  $K(T, T_0)$  and  $K_4(T)$ , gives the <sup>3</sup>He spreading pressure at the cell temperature,  $\alpha_3(T_0)$ . The results are in Fig. 2, which compares  $\alpha_3(T_0)$  with values calculated from the known surface concentrations  $\overline{N_3}^s$  and the <sup>3</sup>He surface effective mass and interaction.<sup>2</sup>



FIG. 2. Spreading pressure of <sup>3</sup>He from conductivity,  $-\alpha_3(\exp)$ , compared with theory calculated from the cell temperature  $T_0$  and surface concentration  $\overline{N_3}^s$ . The symbols are the same as in Fig. 1.

(The concentrations are so small that  $\alpha_3$  is close to the ideal-gas value  $-\overline{N}_3 {}^sk_B T_0$ .) Except for the lowest point, the agreement is very good, since there are no adjustable parameters. The lowest point may be in error as a result of an extraneous constant heat leak; this would cause additional depletion of the <sup>3</sup>He on the surface.

The same analysis can be used to "correct" the measured conductivity K for the effect of the substrate to obtain an idealized surface conductivity. We call this  $K_{eff}$  in analogy with the corresponding 3D quantity in bulk helium.<sup>5</sup> It clearly depends on the local values of the <sup>3</sup>He surface number density  $N_3^{s}$ , and T. To obtain  $K_{eff}(T, N_3^{s})$ we subtract from the measured thermal resistance the resistance due to momentum transfer to the substrate:

$$1/K_{eff}(T, N_3^{s}) = 1/K(T, T_0) - 1/K_4(T).$$
(5)

To interpret  $K_{eff}$  we need to obtain  $N_3^s$  from  $T_0$ and T. This is done by integrating (4) to find  $\alpha_3(T) - \alpha_3(T_0)$  and using the theoretical formula for  $\alpha_3(T)$  to give  $N_3^s$ .

The results for  $K_{\rm eff}$  are in Fig. 3 which shows  $K_{\rm eff}$  vs  $N_3^s$  on a log-log graph. The temperaturedependence of  $K_{\rm eff}$  is very slight, and we see that  $K_{\rm eff} \propto 1/N_3^s$ . This is exactly as expected for scattering of the ripplons by the <sup>3</sup>He, since the <sup>3</sup>He Fermi temperature is very small and each <sup>3</sup>He atom acts independently.

We now define  $t_{3R}$ , the mean scattering time



FIG. 3. The 2D thermal conductivity  $K_{\rm eff}$ , after correcting for momentum transfer to the substrate, as a function of <sup>3</sup>He surface density. The straight line represents  $K_{\rm eff} N_3^{\ s} = 4.5 \times 10^{10} \, {\rm erg \ K^{-1} \ cm^{-2} \ sec^{-1}}$ . The symbols are the same as in Fig. 1.

for a <sup>3</sup>He atom in the ripplon gas, by the equation

$$K_{\rm eff} N_3^{\ s} = S_R^{\ 2} T t_{3R} / m_3, \tag{6}$$

where  $m_3$  is the bare <sup>3</sup>He mass. In bulk He the analogous 3D formula has been derived from the phonon-<sup>3</sup>He Boltzmann equations.<sup>6</sup> For the surface we use the following argument. Suppose that, in each collision, the momentum acquired by a <sup>3</sup>He atom is  $m_3 \vec{v}_R$ . Then the momentum transfer per unit area is  $\nabla \alpha_3 = -N_3 \, {}^sm_3 \vec{v}_R/t_{3R}$ , and (6) follows with use of (1), (3), and (5).

If we neglect the slight temperature dependence of  $K_{eff}$ , the straight line through the data in Fig. 3 corresponds to  $t_{3R} = 4.5[(0.1 \text{ K})/T]^{3.67} \mu \text{sec.}$  Recently Saam<sup>7</sup> has calculated the rate at which a moving <sup>3</sup>He of momentum p emits and absorbs ripplons. With use of the mean thermal value of p his theory gives a collision time of  $1.8[(0.1 \text{ K})/T]^{3.5}$  µsec. This is only an approximation for  $t_{3R}$ , since a precise result requires solution of the <sup>3</sup>He-ripplon Boltzmann equations; however, the T dependence, which agrees with the experiment, should be correct.

Finally we consider the possibility that the <sup>3</sup>He quasiparticle gas has a measurable 2D conductivity  $K_3$ , requiring an additional term  $-K_3 \nabla T$  in (1). For a constant effective interaction  $V^s(k) = V^s(0)$ , effective mass M, and averaging over spin states, the 2D He<sup>3</sup>-He<sup>3</sup> cross section is  $\sigma_v = M|V^s(0)|^2/(4\hbar^3 v)$  for relative velocity v. Substituting in the approximate formula  $K_3 \sim k_B v/(4\sigma_v)$  with  $v \sim v_{\rm rms}$  gives  $K_3 \sim 7 \times 10^{-5}T$  erg sec<sup>-1</sup> K<sup>-2</sup>. This is too small to be measurable in the present experiment.

This work was supported by National Science Foundation Grants No. DMR 75-19546 and No. 79-01073.

<sup>1</sup>A. F. Andreev and D. A. Kompaneets, Zh. Eksp. Teor. Fiz. <u>61</u>, 2459 (1971) [Sov. Phys. JETP <u>34</u>, 1316 (1972)].

<sup>2</sup>For a review of the He surface, see D. O. Edwards and W. F. Saam, *Progress in Low Temperature Physics*, edited by D. F. Brewer (North-Holland, Amsterdam, 1978), Vol. 7A, p. 283.

<sup>3</sup>W. F. Saam, Phys. Rev. A <u>8</u>, 1918 (1973), gives the lifetime at T = 0 for a ripplon to decay into two others. For a ripplon energy of ~ 0.1 K the result is 20  $\mu$  sec. However, at T > 0, the lifetime should be much smaller because of stimulated emission and collisions with low-energy ripplons.

<sup>4</sup>Preliminary measurements for <sup>4</sup>He on a different Mylar substrate are reported in D. O. Edwards, I. B. Mantz, and V. U. Nayak, J. Phys. (Paris), Colloq. <u>39</u>, C6-300 (1978).

<sup>5</sup>E. S. Sabisky and C. H. Anderson, Phys. Rev. A <u>7</u>, 790 (1973).

<sup>6</sup>I. M. Khalatnikov, Introduction to the Theory of Superfluidity (Benjamin, New York, 1965), p. 170.

<sup>7</sup>W. F. Saam, private communication.