RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 34, NUMBER 9 1 NOVEMBER 1986

Measurement of thermal accommodation of spin-polarized hydrogen on a saturated ^{4}He film at 0.18–0.4 K

J. Helffrich, M. P. Maley, M. Krusius, and J. C. Wheatley*
Los Alamos National Laboratory, University of California, P.O. Box 1663, Los Alamos, New Mexico 87545

(Received 1 August 1986)

The thermal accommodation of H-gas atoms on a saturated superfluid-^{4}He-film-covered surface has been measured in the temperature range 0.18–0.4 K. A straightforward stationary-state measuring technique is employed whereby we determine the heat flux conducted by H-gas atoms from a superfluid-^{4}He-film-covered bolometer, biased above ambient temperature. The results agree with recent calculations of the thermal-accommodation properties dominated by sticking-evaporation collisions mediated by ripplon creation.

Measurements on the thermal accommodation of gas-phase H atoms on a liquid-He surface have so far not established a consistent picture of the thermalization mechanisms. On the other hand, recent theoretical work[1-3] seems to have arrived at a consensus: Treating the problem as that of one H atom interacting with an ideal bulk ^{4}He surface, which has a sharp density cutoff, the energy exchange is found to be dominated by sticking and subsequent evaporation with a kinetic energy corresponding to the surface temperature. In a sticking collision the incident H-gas atom is adsorbed to the ^{4}He surface with a binding energy of ~ 1 K and the excess energy is transferred to a single ripplon, or quantized surface capillary wave. With decreasing temperature the sticking mechanism is found to approach zero with an asymptotic temperature dependence of $\sqrt{T}$, reflecting the increasing mismatch of the thermal de Broglie wavelength $\Lambda_H \sim 17$ Å/$\sqrt{T}$ of the incident H atom with the width of the ^{4}He surface potential profile. However, the limiting temperature dependence is not expected until extreme millikelvin temperatures, while above this region the thermalization characteristics are assumed to be a sensitive function of the H-atom–He-surface potential. A measurement of either the thermal accommodation or the sticking would thus provide an efficient probe of the interaction potential.

Aside from the intrinsic interest in the H-atom–He-surface interaction, knowledge of the thermal accommodation properties is of importance for cooling gas-phase H atoms to below 0.1 K. It now appears that the critical density for the expected Bose-Einstein phase transformation can only be reached in the extreme-low-temperature regime at low densities where three-body recombination processes are negligible. However, a decreasing efficiency of thermal accommodation via surface collisions would inhibit cooling of the gas to these temperatures.

Here we report on a measurement of the thermal accommodation coefficient $\alpha(T)$ on a saturated superfluid ^{4}He film in the temperature interval 0.18–0.40 K. The results are found to be consistent with recent calculations of the one-ripplon sticking-evaporation mechanism and remove the ambiguity and inconsistency in earlier data.

The thermal equilibration of gas atoms proceeds via gas-surface collisions, with the temperature of the gas being equal to the surface temperature in equilibrium. We measure the rate at which heat can be transported from an isolated hot surface, the bolometer, whose temperature can be measured, to the gas surrounding it. The maximum amount of heat transfer is obtained if all atoms which strike the surface rebound with the temperature of the surface (perfect accommodation). We define the accommodation coefficient as the ratio of the actual heat transferred by H atoms ΔQ_H to this maximum:

$$\alpha = \frac{\Delta Q_H}{Q_H(T_s) - Q_H(T_g)} . \tag{1}$$

Here $Q_H(T)$ is the translational energy flux of H-gas atoms in the molecular flow regime, where the mean free path λ is greater than the characteristic cell dimensions:

$$Q(T) = n_g \bar{v}(T) k_B T A_{bol}/2 , \tag{2}$$

with n_g the gas-phase density and $\bar{v}(T)$ the average thermal velocity. The calculation of α by Eq. (1) involves the experimentally unobservable gas temperature T_g. Simple arguments based on the much larger probability of gas collisions with the cell walls compared to the hot bolometer surface ($A_{cell}/A_{bol} \sim 150$) indicate that setting $T_g = T_{cell}$ is a good approximation.

The measurement of ΔQ_H is performed by comparing the bolometer bias heating Q_{el} in the absence and in the presence of a H$\downarrow$ charge at a constant bolometer temperature T_b. In the empty cell the electrical heating Q_{el} of the germanium bolometer[4] is balanced by the heat loss Q_L conducted via the two 50-μm gold leads of 3 mm length and by the net evaporation Q_{ev} of ^{4}He atoms from the ^{4}He coverage of the bolometer. When the cell is filled with H$\downarrow$ two additional heat-transfer mechanisms appear: the heat loss ΔQ_H and the heating Q_R absorbed at the bolometer surface from H$\downarrow$ recombination. Subtracting the heat balance equations in the reference and measuring situations allows one to write

$$\Delta Q_H = \Delta Q_{el} + Q_R - \Delta Q_{ev} . \tag{3}$$

Here ΔQ_{el} is the difference in the bias heating in the two situations. ΔQ_{ev} is a correction term originating from a subtle difference in the evaporative cooling caused by the presence of the Kapitza resistance R_K between the bolometer and the ^{4}He film. It is written as $\Delta Q_{ev} = -\Delta Q_{el} R_K$

$\times(\partial Q_{ev}/\partial T_s)$ and thus vanishes for small Q_{ev} below 0.35 K.

The measurements have been performed in a dilution refrigerator using a superfluid-He-temperature pulsed rf discharge source[5] for the dissociation of H_2 molecules. The dissociated atoms effuse from the dissociator along a superfluid-^{4}He-covered flow tube into the copper $H\downarrow$ sample cell. The cell is located in an 8-T polarizing field and is thermally anchored to the mixing chamber. The cell temperature is measured and controlled by a ^{3}He melting pressure gauge. The bolometer[4] is a rectangular germanium chip $0.5\times0.7\times4.2$ mm^3, suspended by two leads from the cell wall. As opposed to the Aquadag bolometers, which have been employed in earlier measurements,[6,7] and which consist of a colloidal film of carbon particles, a crystalline Ge chip with no porosity and a well-defined surface is expected to exhibit a uniform temperature distribution in the presence of the bias heating and the Kapitza resistance.

The variables recorded during the measurement comprise the following: (1) the dc bias heating of the bolometer, (2) its dc resistance which has been calibrated against the ^{3}He melting pressure gauge to read the bolometer temperature T_b, (3) the $H\downarrow$ pressure $P=n_g k_B T_g$ determined with a capacitive Kapton-foil manometer, and (4) the $H\downarrow$ cell temperature T_c. The ^{4}He film temperature T_s is derived from $T_s=T_b-(Q_{el}-Q_L)R_K$. The Kapitza resistance R_K is measured in a separate experiment from the temperature difference T_b-T_c between the heated bolometer and the cell wall when the cell is filled with bulk liquid ^{4}He. The heat conduction Q_L along the bolometer leads is determined from similar bolometer load curve measurements but now in the presence of only the ^{4}He film coating at temperatures below 0.35 K. Finally, the average gas temperature T_g is obtained self-consistently from

$$T_g=T_c+2(Q_{el}-Q_L+Q_{RC})/(\alpha n_g \bar{v} A_{cell} k_B)\ ,$$

where Q_{RC} is the total recombination heating from the $H\downarrow$ sample as determined from the decay of the cell pressure with time.

The stationary-state technique for measuring α, as outlined above, is straightforward and easy to carry out in practice. However, several observations should be made with regard to its applicability.

(i) The measurement has to be performed at low $H\downarrow$ densities to avoid interparticle collisions and a thermal gradient in the gas phase. By limiting the density to less than 10^{15} H atoms/cm^3, $\lambda > 0.5$ cm and the density correction to α will remain below 1% for the $H\downarrow$ cell with a radius $R\sim1$ cm. On the other hand, the density has to be maintained above 10^{14} H atoms/cm^3 in order to reach a similar precision in the pressure measurement.

(ii) A second difficulty is the rapidly increasing Kapitza resistance R_K at low temperatures. The apparent accommodation coefficient α' obtained by using Eq. (1) with T_s replaced by T_b, i.e., $\alpha'=\alpha(T_s-T_g)/(T_b-T_g)$, falls increasingly below the true α with decreasing temperature since $T_s=T_b-\Delta Q_{el}R_K$. For instance, at $T_b=0.2$ K, a measurement with a typical ΔT of $(T_b-T_c)/T_c\sim10\%$ gives $\alpha\sim1.3\alpha'$ in the presence of $\Delta Q_H\sim4.5$ nW and $Q_R\sim160$ pW. Experimentally, the Kapitza problem is revealed by two easily distinguishable signatures: First, the measured result is density dependent since the above correction leads to $\alpha'\simeq\alpha/(1+n_g\times\text{const})$. Second, the deviation increases with decreasing temperature like $\alpha'\simeq\alpha/(1+T^{-3/2}\times\text{const})$.

(iii) Towards high temperatures the measuring region is restricted by the exponentially increasing ^{4}He-gas density. At about $T_c=0.5$ K, the He density becomes comparable to the $H\downarrow$ density and starts to reduce the $H\downarrow$ mean free path. A more stringent limitation is the increase in the evaporative cooling of the bolometer: With $T_b=0.40$ K one finds $Q_{ev}/\Delta Q_H\sim5$ in typical measuring conditions with $(T_b-T_c)/T_c\sim5\%$. The first-order correction to the imperfect cancellation of the evaporative cooling in the reference and measuring situations is of the form

$$\frac{\alpha}{\alpha'}=\frac{T_b-T_g}{T_s-T_g}\left[1+R_K\left(\frac{\partial Q_{ev}}{\partial T_s}\right)\right]\ ,$$

and gives $\alpha\sim1.3\alpha'$, when $\Delta Q_H/\Delta Q_{ev}\sim13$. Consequently, the correction will rapidly swamp ΔQ_H at temperatures above 0.4 K.

(iv) In thermal accommodation measurements the geometry of the measuring arrangement is an important consideration: Ideally all atoms colliding at the bolometer surface should have been perfectly accommodated to the cell walls if Eq. (1) is to apply. However, as the accommodation properties deteriorate towards low temperatures this requirement becomes increasingly difficult to satisfy and calls for a large cell with a small probe. In the present measuring configuration the probe was not centrally located in the cell but we estimate α' to fall below α by less than 5% at 0.2 K.

(v) Finally, it should be pointed out that recombination heating is not a serious difficulty. First of all, the measurements are performed at low density $n_g<1\times10^{15}$ H atoms/cm^3 which limits even surface recombination at our lowest temperatures to low values. Second, a measurement of α is performed at fixed T_b and T_c by monitoring n_g and Q_{el} over a time span of 2–4 h with the same $H\downarrow$ sample before it is recombined and the reference values are recorded. This procedure ensures that the $H\downarrow$ sample is approaching the nuclear relaxation limited doubly polarized state, whereby recombination is minimized. Third, the recombination heating Q_R absorbed at the bolometer is monitored during the measurement by determining the temperature rise of the bolometer above the cell temperature at low excitation voltages for which the bolometer is known not to self-heat. For instance, with $T_c\sim0.20$ K and $n_g\sim5\times10^{14}$ $H\downarrow$ atoms/cm^3 the gas temperature is calculated to be $T_g\sim T_c+0.5$ mK while the bolometer reads a temperature $T_b\sim T_c+1.1$ mK. The higher bolometer temperature is interpreted to result from the heat flux Q_R carried by excited particles which have been part of a recombination event and which in ballistic flight conditions are expected to dump their excess energy uniformly on all surfaces in multiple wall collisions.[5] Compared to the $H\downarrow$ heat flux ΔQ_H the recombination contribution Q_R is found to be a factor of 10–30 smaller.

The results from the correcting procedure are illustrated in Fig. 1, where both α and α' have been plotted as a function of the measured $\Delta T=T_b-T_c$. After applying the

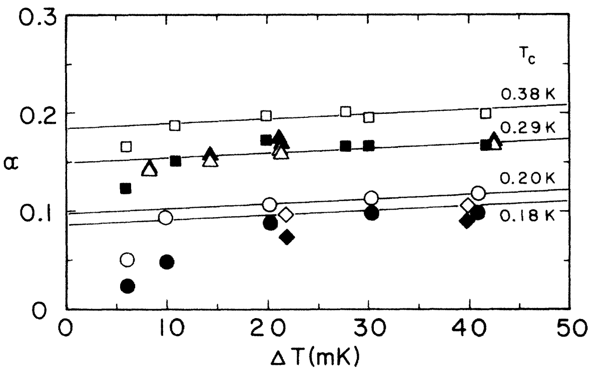

FIG. 1. The thermal accommodation coefficient α vs $\Delta T = T_b - T_c$ at different cell temperatures $T_c \approx T_g$. The open symbols represent α values corrected for the Kapitza resistance, the recombination heating, and the evaporative cooling. The filled symbols illustrate the uncorrected α' extrapolated to $n_g \rightarrow 0$. The different cell temperatures have been plotted using the following symbols: 0.380 K, □; 0.290 K, △; 0.200 K, ○; and 0.180 K, ◇.

corrections for the Kapitza resistance, the evaporative cooling and the recombination heating the α values, shown as the open symbols, become independent of the H↓-gas density n_g but still display a weak residual dependence on ΔT. The lines in the figure have been fitted to the corrected data at large ΔT values and have a common slope of $(\partial\alpha/\partial T_s)_{T_g} = 5.0 \times 10^{-4}$ mK^{-1}. The final data in Fig. 2 have been reduced to $\Delta T \rightarrow 0$ using this slope; this correction is interpreted to remove any systematic problems of the measurement arising from $T_s \neq T_g$ as well as the weak dependence[2] of α on T_s. The filled symbols in Fig. 1 represent the uncorrected $\alpha' = 2\Delta Q_{el}/(n_g \bar{v} A_{bol} k_B \Delta T)$ which depends strongly on the gas density n_g at the measuring conditions. However, the α' values plotted in Fig. 1 have been linearly extrapolated to $n_g \rightarrow 0$ from measurements over several hours while the sample is gradually decaying towards lower densities. Since most of our corrections are ultimately a function of n_g, the extrapolation to zero density provides a simple correction procedure. Indeed, at around $T_c \sim 0.3$ K, which is the most suitable temperature region for the present measuring technique, the open and the filled symbols are seen to agree closely in Fig. 1. Finally, it should be pointed out that at very low ΔT values the precision of the present measuring technique falls off and the data drop below the linear extrapolations from large ΔT in Fig. 1.

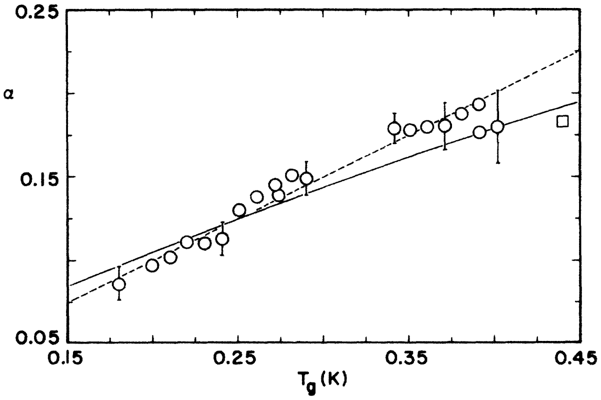

FIG. 2. The thermal accomodation coefficient α as a function of the gas temperature T_g. The solid curve represents the theoretical result from Ref. 2. The dashed line corresponds to a fit to the measurements of Ref. 9 and is given by $\alpha = 0.50T$. The data point marked with □ at 0.44 K is the experimental result from Ref. 6.

In Fig. 2 the measured thermal accommodation coefficient α is plotted as a function of temperature, along with conservative uncertainty limits as estimated from the systematic error sources in the corrections. In addition, an uncertainty of ±10% remains in the value of the surface area of the bolometer which affects all data points uniformly.

Previous measurements of $\alpha(T)$ are scattered and, if examined individually, inconclusive. Nevertheless, they mostly turn out to be consistent with the present data within the combined uncertainty limits. Employing similar measuring techniques Salonen *et al.*[6] obtain $\alpha = 0.18 \pm 0.05$ at 0.44 K in good agreement with our data if their effective α values are extrapolated to zero ΔT. A ballistic heat pulse measurement by Salonen, Silvera, Walraven, and van Yperen[7] gives $\alpha = 0.2 \pm 0.1$ in the temperature region 0.2–0.4 K. Finally, Jochemsen, Morrow, Berlinsky, and Hardy[8] determine from the zero field hyperfine resonance frequency shift caused by the ^{4}He-covered walls a sticking factor $s = 0.046$ in the temperature range 0.18–0.27 K. This value translates to an accommodation coefficient[3] $\alpha \sim 3s/2$ which brings it in reasonable agreement with the present measurement at 0.18 K. A measurement of the sticking coefficient, performed by Berkhout, Wolters, van Roijen, and Walraven[9] at the same time as the present α measurements by monitoring the flow of H atoms through a ^{4}He coated capillary at 0.15–0.53 K, concludes $s = 0.33T$, which provides an excellent fit also to our data as shown by the dashed line in Fig. 2.

In Fig. 2 the measurements are compared to a recent calculation of α by Statt.[2] The agreement is good in view of the experimental uncertainties and the approximations in the calculation. This fact suggests that the theoretical analysis of the thermalization characteristics rests on a satisfactory basis and, more specifically, that sticking evaporation by the one-ripplon mechanism is the dominant process on a ^{4}He surface. Statt used a H-atom–^{4}He-surface potential of Morse type fitted to the potential of Mantz and Edwards,[10] with the well depth adjusted to yield a bound-state energy of 1 K. Within the class of Morse potentials other choices for the parameters result in much poorer agreement with the measurements. Goldman[3] has shown that the choice of the potential, in particular, with regard to its long-range attractive tail and the limiting value of the repulsive core strength (i.e., the chemical potential of a H atom in liquid ^{4}He), produces large differences in the H-atom–^{4}He-surface interaction properties foremost at temperatures below 0.2 K. Even at 0.2 K his different *Ansätze* for the potential result in a spread of α values by a factor of 2, with the present measurements located roughly in the middle of this range.

In conclusion, the present measurements, though limited in their temperature range by the applicability of the stationary-state measuring technique, nevertheless, clearly corroborate the recent calculations on the interaction mechanisms of H atoms with a liquid-^{4}He surface. In order to obtain more conclusive information on the details of the interaction potential, measurements at lower temperatures are required.

This work has been supported by the Division of Materials Sciences of the Department of Energy Office of Basic Energy Sciences.

---

[*] Deceased March, 1986.

[1] D. S. Zimmerman and A. J. Berlinsky, Can. J. Phys. **61**, 508 (1983).

[2] B. W. Statt, Phys. Rev. B **32**, 7160 (1985).

[3] V. V. Goldman, Phys. Rev. Lett. **56**, 612 (1986).

[4] Lake Shore Cryotronics, Inc.: model GR-200A-50 germanium thermometer chip with gold leads and a surface area $A_B = 0.148$ cm^2, which includes a pair of unused loose Cu leads.

[5] J. Helffrich *et al.*, J. Low Temp. Phys. (to be published).

[6] K. Salonen, S. Jaakkola, M. Karhunen, E. Tjukanov, and T. Tommila, in *Proceedings of the Seventeenth International Conference on Low Temperature Physics*, edited by V. Ekern, A. Schmid, W. Weber, and H. Wühl (North-Holland, Amsterdam, 1984), p. 543.

[7] K. Salonen, I. F. Silvera, J. T. M. Walraven, and G. H. van Yperen, Phys. Rev. B **25**, 6002 (1982).

[8] R. Jochemsen, M. Morrow, A. J. Berlinsky, and W. N. Hardy, Phys. Rev. Lett. **47**, 852 (1981). See also M. Morrow and W. N. Hardy, Can. J. Phys. **61**, 956 (1983).

[9] J. J. Berkhout, E. J. Wolters, R. van Roijen, and J. T. M. Walraven, Phys. Rev. Lett. (to be published).

[10] I. B. Mantz and D. O. Edwards, Phys. Rev. B **20**, 4518 (1979).