Vanishing Sticking Probabilities and Enhanced Capillary Flow of Spin-Polarized Hydrogen

J. J. Berkhout, E. J. Wolters, R. van Roijen, and J. T. M. Walraven
Natuurkundig Laboratorium, Universiteit van Amsterdam, NL-1018XE Amsterdam, The Netherlands

(Received 11 August 1986)

We report on a series of capillary-flow experiments with spin-polarized hydrogen in which we observe a strong enhancement (> 10×) of the flow rate in comparison to ordinary Knudsen flow. This is related to a vanishing sticking probability s for $T \rightarrow 0$ K as predicted by theory. With ^{4}He surfaces we find that $s = 0.33T$ for 145 mK $< T <$ 526 mK. Within experimental error s did not change upon the addition of up to 50% ^{3}He at 150 mK. Results for ^{3}He/^{4}He surfaces were obtained for 73 mK $< T <$ 174 mK and are also described by $s = 0.33T$.

PACS numbers: 67.65.+z, 47.45.Dt

The physics of scattering of hydrogen atoms from the surface of liquid helium has grown into an important subject in studies of spin-polarized hydrogen ($H\downarrow$) at low temperature.[1] Inelastic scattering provides the mechanism for the exchange of heat between the bulk $H\downarrow$-gas phase and the helium-covered walls of a sample cell and leads to a thermal-boundary resistance. In particular, at high $H\downarrow$ densities or low temperatures, where recombination to the (solid) molecular state H_2 is most effective, substantial temperature differences may develop between gas and surface. This is recognized as a major barrier in the achievement of Bose-Einstein condensation in $H\downarrow$ by compression methods.[2] In recent proposals to aim for Bose-Einstein condensation by pure field confinement in a magnetic trap, walls are of vital importance to provide a cooling mechanism during filling. There one has to strike a subtle balance between cooling efficiency and flux losses due to surface recombination or spin relaxation.[3]

Some experimental information is available for the sticking coefficient s, the probability per collision to be adsorbed, and for the accommodation coefficient α, characterizing the transfer of translational energy transfer per collision. The sticking coefficient was first determined by Jochemsen *et al.*[4] By means of magnetic resonance they found that $s = 0.046$ at $T \approx 200$ mK for ^{4}He surfaces and $s = 0.016(5)$ for ^{3}He surfaces at $T \approx 100$ mK. Salonen *et al.*[5] measured ballistic heat transport with a time-resolved method and found that $\alpha = 0.2(1)$ for H on ^{4}He. In a subsequent steady-state heat flux experiment Salonen *et al.*[6] (SJKTT) obtained $\alpha = 0.19(5)$ for $T = 440$ mK. Bell *et al.* found α to decrease from $\alpha = 0.8(4)$ at 0.6 K to $\alpha = 0.4(2)$ at 0.275 K.[7] Very recently Helffrich *et al.*[8] also found a decreasing α for decreasing temperature with an SJKTT-type experiment. We noted slightly different definitions of α by the various authors.

In this Letter we report on a series of capillary-flow experiments which are particularly suited to the estimation of s for H on liquid-helium surfaces. In comparison with ordinary Knudsen flow a dramatically reduced (> 10×) flow impedance is observed which we attribute to a small value of s. The impedance is decreasing with decreasing temperature. Our results show that the earlier observed differences between sticking and accommodation coefficients are mainly due to differences in temperature of observation. We also estimate the coefficient of self-diffusion for $H\downarrow$ in the gas phase.

Classically one expects s to approach unity in the zero-temperature limit. Quantum-mechanical theory predicts s to vanish as $T^{1/2}$ for $T \rightarrow 0$ K and shows excitation or deexcitation of ripplons to be the dominant mechanism of energy exchange between gas and liquid.[9-12] Also, the Kapitza conductance is calculated to vanish in the $T \rightarrow 0$ limit but the numerical values, in particular below 100 mK, vary by orders of magnitude depending on the adsorption potential and the scattering channels included in the calculation.[11-13]

The experimental cell is shown schematically in Fig. 1. The atoms are introduced from the bottom into a buffer volume ($V_b \approx 10$ cm^3) where the pressure may be monitored with a capacitive pressure transducer having a sensitivity of 1.74 pF/Torr and a noise equivalent pressure of 5×10^{-7} Torr (1 standard deviation at a bandwidth of 1.25 Hz). The minimum detectable density is 1×10^{13} cm^{-3} at 500 mK. The buffer volume is connected to a pump volume (V_p) via a Pyrex capillary of length $l = 21.97(3)$ mm and inner radius of $a = 0.248(2)$ mm. All surfaces in the cell are covered with a film of liquid helium. To pump the $H\downarrow$ the helium film is boiled off from a copper plate of area $A_p = 7.2$ cm^2 suspended by three 16-μm-diam tungsten wires, exposing the $H\downarrow$ to a substrate offering a strong ($\gg 1$ K) binding energy and hence causing massive recombination in the pump volume. A heating power of 15 μW is needed to heat the pump plate to its operational temperature $T = 2.1(1)$ K. The whole assembly acts as a miniature atomic-beam apparatus. The beam detectors are not discussed in this article. The cell is placed in a 6-T magnetic field homogeneous to within 1:10^4 over the capillary region. The flow through the capillary may be throttled by local reduction of the field to less than 5 T with a small coil.

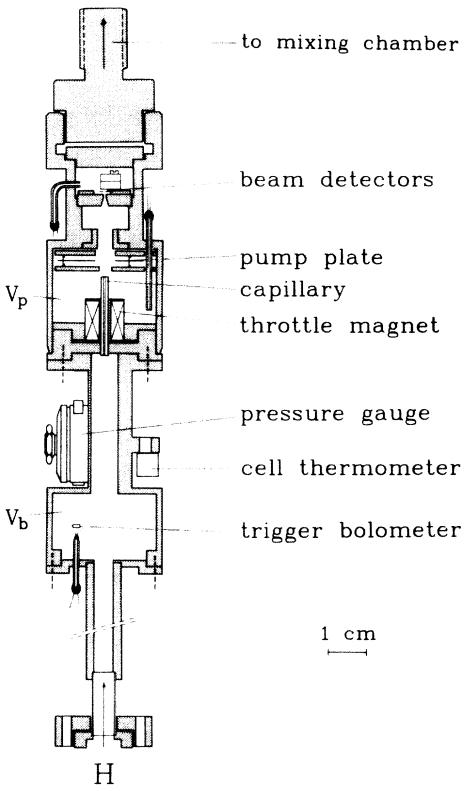

FIG. 1. Schematic drawing of the apparatus. For details see text.

Great care was exercised to avoid magnetic surface impurities in the buffer volume in order to minimize recombination losses.[14]

To determine s for a given temperature we measure the flow impedance of the capillary, either by using the pressure gauge or by measuring recombination heat dissipated into the pump plate. By measurement with the pressure gauge the whole cell is first filled with H$\downarrow$ up to a density $n \approx 2\times 10^{15}$ cm^{-3}. Then the pump plate is heated to its operational temperature and one monitors, after a short (<1 s) transient due to the recombination of the gas in V_p, the exhaust of the buffer volume through the capillary. Since $A_p \gg A_0 \equiv \pi a^2$ the probability of the H$\downarrow$ atoms in returning to V_b is negligibly small ($\ll 10^{-3}$). By measurement with the pump plate one monitors the power required to stabilize the plate temperature to a fixed value of 2.1 K. The sensitivity of this method is at least 3 orders of magnitude better than that of the pressure measurements.

The flux Φ through the capillary is described by

$$-dN/dt = \Phi = \tfrac{1}{4} K N \bar{v} A_0 / V_b \equiv K\Phi_0, \qquad (1)$$

where N is the number of atoms in the buffer volume V_b, $\bar{v} = (8kT/\pi m)^{1/2}$ is the average atomic speed, Φ_0 is the rate at which atoms enter the capillary, and K is the Clausing factor,[15] expressing the probability for an atom to pass through the tube without returning to V_b. In the Knudsen regime for ordinary capillary flow $K = K_{\rm K} \approx 8a/3l$, purely determined by geometrical factors and thus temperature independent. For our geometry $K_{\rm K} = 0.0284$.[16] We determine K experimentally by measuring the characteristic time $\tau \equiv \tau_0 / K \equiv (\frac{1}{4} K \bar{v} A_0 / V_b)^{-1}$, typically 5–10 s, in which the buffer volume is discharged through the capillary. Here τ_0 is the characteristic time for effusion through the entrance orifice A_0. Once τ is known we extract the corresponding value for s using a computer simulation of the flow.

The nature of surface scattering only enters into Eq. (1) through the Clausing factor K. If one assumes the scattering to be purely diffusive (according to a cosine distribution), as was done by Clausing, one expects that $K = K_{\rm K}$. On the other hand, if no momentum is transferred parallel to the surface, as in the case of specular reflection, Φ is entirely determined by the impedance of the entrance orifice and one should have $K = 1$. We simulate the capillary flow with a one-speed-test-particle Monte Carlo method.[17] At each wall collision the atom is given a probability (s_u) to scatter diffusively, otherwise the atom scatters specularly. Our results for s_u vs K were calculated to within 1% using 10^5 particles and are given within 3% by the empirical form

$$s_u = \frac{1-K}{1-K_{\rm K}} \left(\frac{K_{\rm K}}{K} \right)^{1/2} e^{-3.915\Delta K} \{1 + 4.2\Delta K^2\}, \qquad (2)$$

where $\Delta K = K - K_{\rm K}$. The model clearly represents a simplified picture of the physics involved. Our measurement is sensitive for any process that leads to momentum transfer in the direction along the axis of the capillary tube. This is most effectively done by atoms that stick some time to the surface, but more generally results from any inelastic-scattering process. Hence our model delivers an upper estimate for s. However, in view of the theoretical assertion[10–12] that the sticking channel is most effective in exchanging energy and momentum between gas and surface, it seems reasonable to assume that $s_u \approx s$. Surface roughness may also lead to nonspecular reflection as is well known for ultracold neutrons[18] and light.[19] Using a scanning electron microscope with a maximum resolution of ~ 100 Å we found the capillary inner surface to be extremely flat except for some isolated grains of loose dirt. We assume any irregularities of size < 100 Å to be smoothed out by the helium film. The influence of roughness due to thermally excited ripplons appears to be small.[20]

Our simulation also allows us to study the effect of collisions between atoms in the gas phase and to extract the diffusion coefficient from the measurements. After a randomly generated collision the atoms are restarted in random directions, sampling an angular distribution chosen to be isotropic in the laboratory system. Relating the mean free path λ to n according to[21] $\lambda = (16\sqrt{2} n \sigma /$

$9\pi)^{-1}$, where σ is the hard-sphere scattering cross section, we may describe our simulation results for the density dependence of K by

$$K \approx K_0(1-\sigma\lambda_0 K_0 n). \quad (3)$$

Here K_0 is the Clausing factor in the zero-density limit and $\lambda_0 \equiv 6.1$ mm is a reference length provided by the simulation. Equation (3) only holds in the extremely rarefied limit. At higher densities K should show a minimum before entering the viscous regime.[22] In our measurements $\lambda > 2$ cm. Once σ is known the coefficient of self-diffusion follows with $D = \frac{1}{3}\bar{v}\lambda$.[21]

With the aid of Eqs. (1) and (3) and allowing for recombination we may express the rate of loss of atoms from the buffer volume by a second-order rate equation

$$dn/dt = C_1 n + C_2 n^2, \quad (4)$$

where $C_1 \equiv -K_0/\tau_0$ and $C_2 \equiv \sigma\lambda_0 K_0^2/\tau_0 - K_{\rm rec}$. $K_{\rm rec}$ is a rate constant accounting for recombination. Equation (4) is integrated and fitted to the pressure decay data to extract the first- and second-order decay constants. Data were only collected in a temperature regime where the helium vapor pressure did not significantly impede the flow and recombination losses could be neglected, as was verified with the aid of the throttle magnet.

We note that the fraction of the atoms entering the tube over the surface is small, 4.7% for ^{4}He surfaces at $T = 145$ mK. At this temperature the adsorption time $\tau_s = (h/skT)\exp(\varepsilon_a/kT) \approx 7\ \mu$s ($h$ is the Planck constant, $s = 0.05$, and $\varepsilon_a = 1$ K is the adsorption energy[1]), much longer than the momentum relaxation time 1.1 ns of adsorbed atoms.[23] Hence the motion of the adsorbed atoms is diffusive. The mean square displacement from the adsorption site is very small ($\sim 5\ \mu$m) and as such negligible.

The results for pure ^{4}He surfaces are shown in Fig. 2(a), where we plot K_0 vs T. The open circles are obtained from pressure-gauge measurements; the filled circles are our pump-plate results. Note the excellent agreement between both methods and the enormous enhancement in conductance in comparison to the value $K_{\rm K}$ to be expected for ordinary Knudsen flow. Each data point represents the average of at least four measurements, the error bar being the standard deviation of the results, augmented by a small systematic error. By applying Eq. (2) we obtain our results for s_u, which are shown in Fig. 2(b). The data fit well to a straight line $s_u = 0.33T$ for 145 mK $< T <$ 526 mK. Our value for 200 mK is approximately 30% larger than the Jochemsen *et al.* result [encircled 4 in Fig. 2(b)]. Within experimental error we find agreement with the SJKTT result at 440 mK (filled square). We also indicate in Fig. 2(b) the result (the large rectangle) obtained by Salonen *et al.* in Amsterdam and the result of Bell *et al.*[7] (inverted filled triangle). Note that we used $s = \frac{2}{3}\alpha$ to plot the accommodation results.[12] The dashed curve in Fig. 2(b) corresponds to the $V_{\rm II}$ curve of Goldman.[12] The straight line corresponds to $s_u = 0.33T$. In proof we note very good agreement between the recent results of Hellfrich *et al.*[8] and our ^{4}He data.

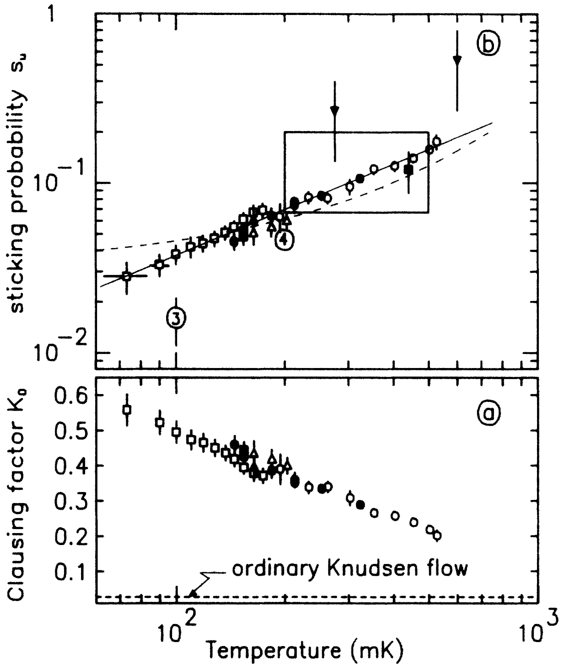

FIG. 2. (a) Measured values for the Clausing factor K_0 vs temperature. (b) Fraction s_u of the atoms scattered diffusively from the surface, $s_u \approx s$. Our results are the following: open circles, ^{4}He; open triangles, 0.2% ^{3}He; open triangles, 5% ^{3}He (pressure gauge); filled circles, ^{4}He; open squares, 3%–50% ^{3}He (pump plate). Other data: open circles, Ref. 4; filled squares, Ref. 6; big rectangle, Ref. 5; inverted filled triangles, Ref. 7; dashed curve, theory of Ref. 12. The solid line corresponds to $s = 0.33T$.

In adding a small amount of ^{3}He (0.2%) to the ^{4}He in our cell we noticed a clear reduction of the recombination rate, indicating a reduction[24] of the binding energy and implying a definite change in adsorption potential and in ripplon dispersion [the surface tension falls from $\gamma = 0.378(2)$ ergs cm^{-2} to $\gamma = 0.153(4)$ ergs cm^{-2} when changing from pure ^{4}He to pure ^{3}He surfaces].[25] However, within experimental error no effect was observed on K or s_u (open triangles in Fig. 2). After the increase of the ^{3}He fraction to 5%, a small ($\approx 10\%$) increase in s_u was observed at 164 mK with the pressure gauge (filled triangle). Under these conditions we estimate the local ^{3}He concentration in the cell to be at least 30% and hence expect the H↓ to scatter from a ^{3}He-rich phase. Most of our 5% ^{3}He results, shown as open squares in Fig. 2(b), were obtained with the pump plate and are in fair agreement with the 164-mK pressure-gauge result. Although the reliability of our thermometry rapidly decreases below 100 mK, the trend of decreasing sticking probabilities with decreasing temperatures clearly continues down to our lowest temperature. In comparing our 100-mK value with the ^{3}He result of Jochemsen *et al.* (encircled 3) we note that our value is a factor of 2–3 larger. With the addition of 50% ^{3}He no further pressure measurements could be done as bulk liquid started to collect in the pressure gauge. With the pump plate no change was observed on s.

Values for C_2 were obtained with the pressure gauge, starting the fits at a fixed initial density $n_0 = 5\times10^{14}$ cm^{-3}. We found that $C_2\times10^{17} = 6.2(15) - 8(14)[T - 0.27]$ cm^3 s^{-1} which leads to a diffusion constant which is temperature independent within experimental error. Averaging over all data we find that $Dn = 1.3(5)\times10^{18}$ cm^{-1} s^{-1}, in good agreement with the theoretical value of Lhuillier,[26] $Dn = 1.5\times10^{18}$ cm^{-1} s^{-1}. In view of the large experimental error in C_2 no effort was made to refine the treatment of diffusion in our simulation.

Our results show that earlier observed differences between sticking and accommodation coefficients are largely due to differences in temperature of observation [see Fig. 2(b)]. Differences between our results and those of Bell *et al.*[7] may not be significant in view of their large experimental error. The surface densities in their experiments are not sufficiently high to cause enhanced sticking through the mechanism of Collaudin, Hébral, and Papoular.[27] The discrepancy with the ^{3}He result of Jochemsen *et al.* remains unexplained. Remarkable is the small, < 10%, change in s when one adds ^{3}He into the cell.

The authors wish to thank O. H. Höpfner for technical support, J. Koelman for pointing out an inconsistency in our computer simulation, and I. D. Setija, I. Lunding, and J. Mosk for assistance with the measurements and data analysis. We profited from discussions with H. Beijerinck, A. Lagendijk, M. Papoular, and R. Sprik. The financial support of the Stichting FOM is gratefully acknowledged.

---

[1] I. F. Silvera and J. T. M. Walraven, in *Progress in Low Temperature Physics,* edited by D. Brewer (North-Holland, Amsterdam, 1986), Vol. 10, Chap. D.

[2] R. Sprik, J. T. M. Walraven, and I. F. Silvera, Phys. Rev. Lett. **51**, 479, 942(E) (1983); H. F. Hess, D. A. Bell, G. P. Kochanski, R. W. Cline, D. Kleppner, and T. J. Greytak, Phys. Rev. Lett. **51**, 483 (1983); T. J. Tommila, S. Jaakkola, M. Krusius, I. Krylov, and E. Tjukanov, Phys. Rev. Lett. **56**, 941 (1986).

[3] H. F. Hess, Bull. Am. Phys. Soc. **30**, 854 (1985); R. V. E. Lovelace, C. Mehanian, T. J. Tommila, and D. M. Lee, Nature (London) **318**, 30 (1985); H. F. Hess, to be published; T. J. Tommila, to be published.

[4] R. Jochemsen, M. Morrow, A. J. Berlinsky, and W. N. Hardy, Phys. Rev. Lett. **47**, 852 (1981); we use the result of the reanalysis by M. Morrow and W. N. Hardy, Can. J. Phys. **61**, 956 (1983).

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

[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 U. Eckern, A. Schmid, W. Weber, and H. Wühl (North-Holland, Amsterdam, 1984), p. 543.

[7] D. A. Bell, H. F. Hess, G. P. Kochanski, S. Buchman, L. Pollack, Y. M. Xiao, D. Kleppner, and T. J. Greytak, to be published.

[8] J. Helffrich, M. Maley, M. Krusius, and J. C. Weatley, Phys. Rev. B (to be published).

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

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

[11] Yu. Kagan, G. V. Shlyapnikov, and N. A. Glukhov, Pis'ma Zh. Eksp. Teor. Fiz. **40**, 287 (1984) [JETP Lett. **40**, 1072 (1984)].

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

[13] B. Castaing and M. Papoular, J. Phys. (Paris), Lett. **44**, L537 (1983).

[14] R. Sprik, J. T. M. Walraven, G. H. van Yperen, and I. F. Silvera, Phys. Rev. B (to be published).

[15] P. Clausing, Ann. Phys. (Leipzig) **12**, 961 (1932).

[16] R. J. Cole, Prog. Astronaut. Aeronaut. **51**, 261 (1977).

[17] G. A. Bird, *Molecular Gas Dynamics* (Clarendon, Oxford, 1976). For another method see K. Yamamoto, J. Hara, and K. Hirose, J. Phys. Soc. Jpn. **51**, 3729 (1982).

[18] M. Brown, R. Golub, and J. M. Pendlebury, Vacuum **25**, 61 (1975).

[19] J. M. Elson and J. M. Bennet, J. Opt. Soc. Am. **69**, 31 (1979).

[20] D. O. Edwards and P. P. Fatouros, Phys. Rev. B **17**, 2147 (1978).

[21] J. O. Hirschfelder, C. F. Curtiss, and R. B. Bird, *Molecular Theory of Gases and Liquids* (Wiley, New York, 1954).

[22] M. Knudsen, Ann. Phys. (Leipzig) **28**, 75 (1908).

[23] D. S. Zimmerman and A. J. Berlinsky, Can. J. Phys. **62**, 590 (1984).

[24] A. P. M. Matthey, J. T. M. Walraven, and I. F. Silvera, Phys. Rev. Lett. **46**, 668 (1981).

[25] D. O. Edwards and W. F. Saam, in *Progress in Low Temperature Physics,* edited by D. F. Brewer (North-Holland, Amsterdam, 1978), Vol. 7A, p. 283.

[26] C. Lhuillier, J. Phys. (Paris) **44**, 1 (1983).

[27] B. Collaudin, B. Hébral, and M. Papoular, J. Phys. (Paris) **47**, 1503 (1986).

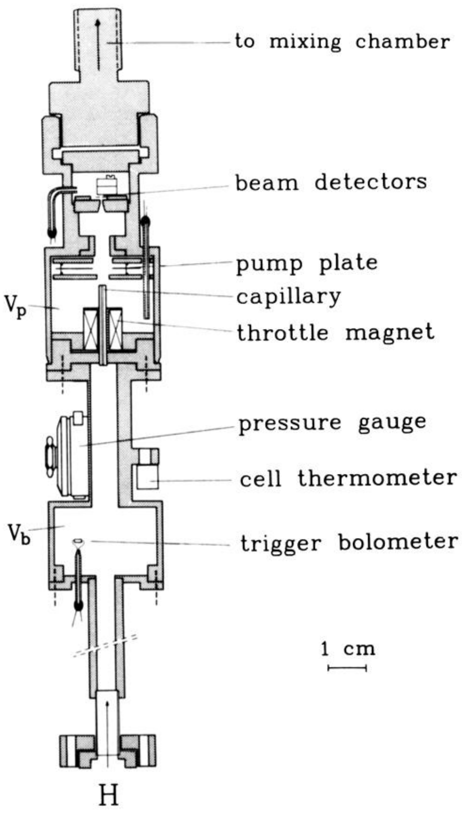

FIG. 1. Schematic drawing of the apparatus. For details see text.