

Sticking Probability of ^4He on Solid Surfaces at Low Temperature

M. Sinvani, M. W. Cole,^(a) and D. L. Goodstein

California Institute of Technology, Pasadena, California 91125

(Received 23 May 1983)

The first low-temperature (< 4 K) determination of the sticking probability S of a ^4He atom on a solid surface is presented. It is found that $\frac{2}{3} \lesssim S < 1$ for a thermal distribution of incident atoms with beam temperature 10 to 20 K. This result is not affected by the presence of a preadsorbed, $\lesssim 1$ -monolayer He film.

PACS numbers: 67.70.+n, 68.40.+e, 79.20.Rf, 82.65.My

The sticking coefficient S is an important and controversial parameter in the study of desorption and evaporation as well as atomic beam scattering. For ^4He incident on a solid surface, classical theory predicts $S \rightarrow 1$ as $T \rightarrow 0$, but the quantum theory prediction is less clear.¹⁻⁴ The only related measurements for ^4He give an energy accommodation coefficient $\alpha \approx 0.02$ for incidence on a tungsten surface,⁵ and $S \approx 0.98$ for incidence on the surface of liquid ^4He at low temperature.⁶

In this Letter we present data showing that S is more than about $\frac{2}{3}$, but distinctly less than 1 for ^4He incident with energies ~ 10 – 20 K on a metallic or dielectric surface at low temperature (< 4 K). We also find that S does not depend on whether the metallic surface is very nearly bare, or is covered instead with close to a monolayer of preadsorbed ^4He . These surprising results emerge from atom scattering experiments based on flash-desorption techniques.^{7,8}

The experimental arrangement is shown schematically in the inset in Fig. 1. An Ohmic heater h_1 (~ 600 -Å Nichrome film) and a superconducting-transition bolometer b (~ 2000 -Å Sn) are mounted on an optically polished sapphire substrate. h_1 and b are separated by a groove to suppress signals due to phonons. Parallel to this surface at a distance $l = 0.3$ mm away is a second sapphire surface, on which a heater h_2 , identical to h_1 , has been placed such that if atoms desorbed from h_1 reflect specularly to b , they reflect from the surface of h_2 . The heaters and bolometer have dimensions $\sim 0.3 \times 0.3$ mm² and thermal time constants ~ 10 nsec. There is ^4He vapor in the cell at chemical potential $\mu = 70$ K (pressure $\sim 10^{-5}$ Torr), so that all surfaces are initially covered with a film of ~ 0.7 layer at ambient temperature 3.6 K.^{7,8} The surfaces of the heaters, bolometers, and crystal have received no special treatment beyond normal laboratory care in handling and cleaning.

Figure 1 shows the result of a particular sequence of pulses applied to the heaters. A pulse

lasting 40 μsec , from t_2 to t_2' , is applied to h_2 , raising its temperature to $T_{h_2} \approx 19$ K in ~ 10 nsec,^{7,8} and giving rise to the first large bolometer signal due to desorption from h_2 . 20 μsec after t_2 , at t_1 , while h_2 is still hot and essentially bare,⁹ a 30-nsec pulse is applied to h_1 , raising its temperature to $T_{h_1} \approx 19$ K. This gives rise to the second, smaller signal which is due to atoms reflected from either h_2 or the surrounding sapphire surface at ambient temperature.

The dependence of the reflection signal on T_{h_2} is examined in Fig. 2, where the uppermost curve corresponds to the second, smaller signal in Fig. 1. The lowest curve is the signal with $T_{h_2} = 3.6$ K, the ambient temperature. The arrival time of this peak is what would be expected for specular reflection from the upper surface. As T_{h_2} is increased, the signal becomes larger and the peak at first moves to later times. However, when $T_{h_2} = T_{h_1} = 19$ K (the uppermost curve) the arrival time is once again equal to that of the lowest

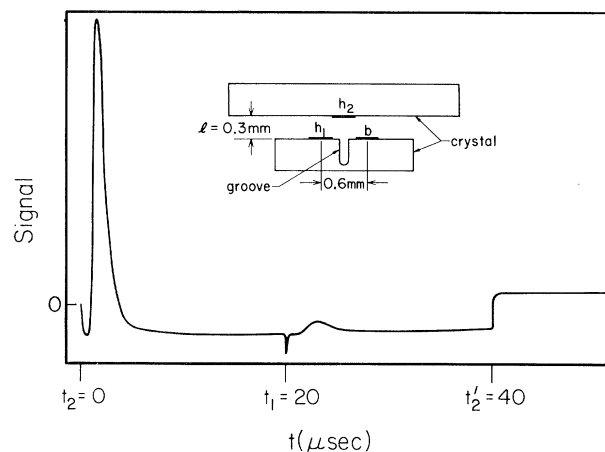


FIG. 1. Signal at bolometer b due to desorption from heaters h_2 and h_1 (see inset for schematic). The large peak arises from an h_2 pulse extending from $t_2 = 0$ to $t_2' = 40$ μsec . The small peak is due to atoms desorbed by a short pulse (30 nsec width at time t_1) applied to h_1 which have been reflected from the upper surface.

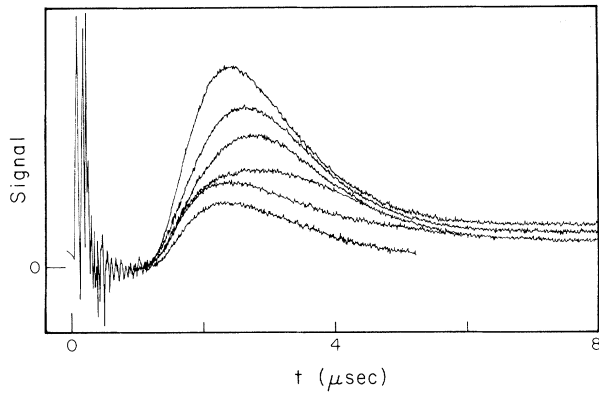


FIG. 2. Bolometer signals as a function of time following pulse on lower heater h_1 . Signal is shown for various temperatures T_{h_2} of the upper heater: $T_{h_2} = 3.6, 7.6, 8.5, 9.5, 11,$ and 19 K. In all cases, the pulse width is 30 nsec, the temperature $T_{h_1} = 19$ K, and ambient $T = 3.6$ K.

curve.

These observations may be interpreted as follows: Those atoms that stick to a surface at 3.6 K linger for a time that is very long compared to all other times in the experiment, without re-desorbing. Indeed, it is this fact which is responsible for the bolometer signal itself. However, the desorption time is an exponential function of the surface temperature.⁷ Thus as T_{h_2} increases, re-desorption occurs at successively shorter times, giving rise to the increased signal at b . At $T_{h_2} = 19$ K, the re-desorption is effectively instantaneous. Thus the uppermost curve in Fig. 2 is the signal at b when all of the atoms incident at h_2 are quickly ejected. Since the uppermost signal has about 3 times the area of the lowest, we are able to conclude that at least $\frac{2}{3}$ of the atoms incident on He-coated h_2 at $T_{h_2} = 3.6$ K stuck to the surface¹⁰:

$$S \gtrsim \frac{2}{3}.$$

The argument for this inequality derives from considering extreme opposite alternatives for the reflection process, specular or diffuse. If the reflection is specular, the re-desorbed beam will be less efficient than the reflected beam in directing atoms toward b , so that comparison of the signals underestimates the contribution of the desorbed beam. Conversely, if the reflection is diffuse, the lowest curve is due to reflection from the entire surface of the upper sapphire crystal, and we have overestimated the contribution due to reflection from the cold surface of h_2 . Thus in either case, the ratio of the lowest to the

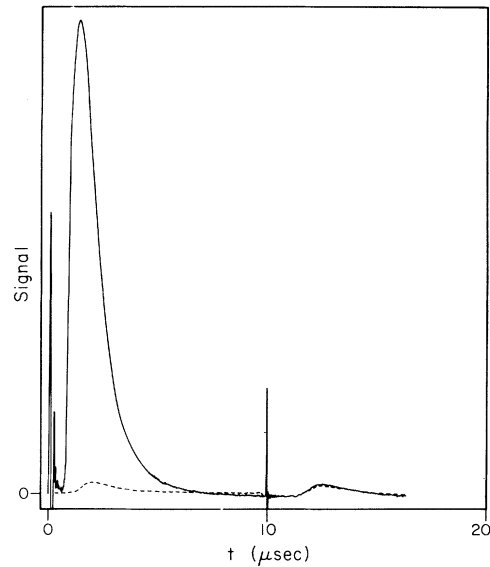


FIG. 3. Signal at b due to desorption from h_2 followed by signal (the small one) due to desorption from h_1 and reflection from h_2 . Dashed curve: small initial pulse in h_2 leaving it covered with the He film. Solid curve: large initial pulse in h_2 leaving it bare.

uppermost signal in Fig. 2 means that $S \gtrsim \frac{2}{3}$ for a surface at $T = 3.6$ K covered with about 0.7 layers of ^4He . Obviously the fact that there is a reflected signal when $T_{h_2} = 3.6$ K means that S is measurably less than 1.

It has been shown previously⁸ that under the conditions of these experiments, pulsing either heater to $T_h \approx 19$ K for a time longer than the desorption time will desorb ΔN atoms/cm², where $\Delta N/S \approx 10^{15}$, or approximately a monolayer. Thus, our finding that $S \gtrsim \frac{2}{3}$ permits us to conclude that such a pulse strips the surface essentially bare. This conclusion is lent support by equilibrium thermodynamic data¹¹ for ^4He adsorbed on graphite: Warming the surface at constant gas pressure from 4 to 15 K reduces the equilibrium film from more than one layer to less than 1% of a layer. We expect the surfaces in the present experiment to exhibit similar behavior.

To find S for a surface which is both bare and cold, we make use of the fact that once the surface has been pulsed bare, it cools back down in ~ 10 nsec, but a very long time is required (> 0.01 sec) before the gas is able to replenish the film.⁸ Thus it remains bare and cold long enough to do a reflection experiment. In Fig. 3, h_2 is pulsed for 30 nsec at $t=0$, giving rise to the first, or desorption, signal, and h_1 is pulsed for 30 nsec at $t=10$ μsec , giving rise to the

smaller reflection signal. Since the pulse in h_2 is narrow compared to 10 μsec reflection takes place from a cold, bare surface. The dashed curve is for a 6-K pulse in h_2 , leaving the surface covered, but the solid curve is for a 19-K pulse in h_2 , leaving it bare, as may be seen from the large desorbing signal. The reflected signal is essentially the same in both cases. Since this signal is equivalent to the lowest curve in Fig. 2, we conclude that S lies between $\frac{2}{3}$ and 1 also for reflection from a bare, cold surface. We also find the same signal when h_2 is absent, so that reflection takes place from the helium-covered sapphire surface.

Figure 4 shows a sequence which tends to verify the picture we have presented of the earlier experiments. Here, h_2 is pulsed every 10 μsec with a pulse 30 nsec wide. The first few pulses (not shown) denude the surface, and one sees no desorption signal from subsequent pulses at times t_2 because there is not enough time for the film to readorb. At time t_1 , however, h_1 is pulsed, giving rise to a reflection signal. Then on the next pulse of h_2 , a desorption signal is now seen; these are the atoms from h_1 which stuck to h_2

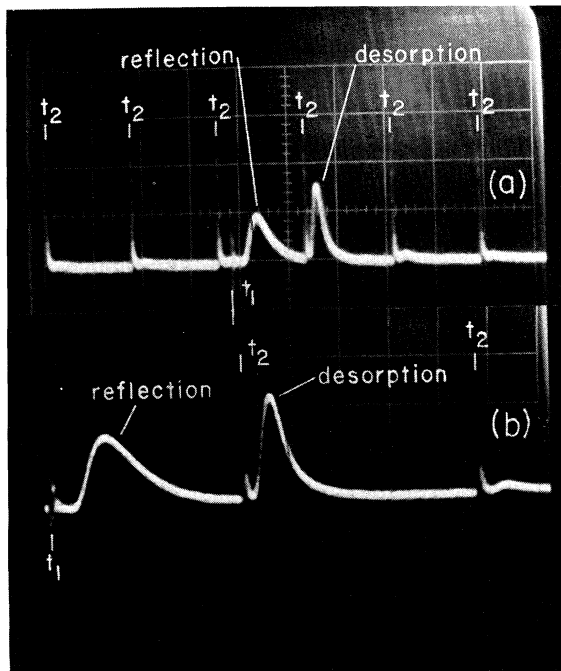


FIG. 4. (a) Signal when h_2 is pulsed at 10- μsec intervals (times t_2) keeping it bare. h_1 is pulsed at t_1 resulting in reflection signal, followed after next t_2 by desorption of those atoms from h_1 that stuck on h_2 . (b) Same sequence with an expanded time scale.

rather than reflecting. (Because the pulses on h_2 are very narrow, a very small signal may also be seen after the next pulse.)

This experiment shows directly both sticking and reflection at h_2 when it is cold and bare. It also shows that h_2 remains bare for long times unless atoms from h_1 are supplied to it.

It is at first puzzling that the desorption peaks in Fig. 4 are so small. On the basis of Fig. 2, one might have expected the total area under the desorption peaks to be about twice that under the reflection peak, i.e., the difference between the uppermost and lowest curves in Fig. 2. Clearly this is not the case. Our interpretation is as follows: The redesorption in the upper curve of Fig. 2 takes place so rapidly that thermal equilibrium with the substrate is never established. Thus the redesorbed atoms come off the surface with some memory of where they came from. They are preferentially redesorbed about the direction of specular reflection. This supposition is plausible because emitting or absorbing a small number of substrate phonons can have little effect on the component of the atom's momentum parallel to the surface. On the other hand, for the experiment in Fig. 4, the adsorbed atoms have ample time to come to equilibrium. They redesorb preferentially normal to the surface (for the same kinematical reason),¹² and thus tend not to arrive as often at the bolometer.

In summary, we have made the first direct measurement of sticking probability for ^4He on a solid surface at low temperature. We find that $1 > S \gtrsim \frac{2}{3}$ on a bare metallic surface, and on a metallic or dielectric surface covered with just under a layer of helium. A more detailed analysis of these data leading to a more precise value of S will be presented elsewhere. We find that $S \approx 0.8$ in all of these cases.

Our data at higher surface temperature (19 K) could be interpreted to give $S \approx 0$. They are thus consistent with earlier accommodation coefficients.⁵ It is clear from our data, however, that this result arises from a disappearance of a distinction between direct reflection and redesorption, phenomena that may be separated in time only at lower temperature.

This work was supported in part by U. S. Office of Naval Research Contract No. N0014-80-C-0447.

(a)Permanent address: Physics Department, Penn-

sylvania State University, University Park, Pa. 16802.

¹F. O. Goodman, in *Progress in Surface Science*, edited by S. Davison (Pergamon, New York, 1975), Vol. 5, p. 261; F. O. Goodman, *J. Phys. Chem.* **84**, 1431 (1980).

²M. W. Cole and F. Toigo, in *Interfacial Aspects of Phase Transformations*, edited by B. Mutaftschiev (Reidel, Dordrecht, Holland, 1982), p. 223.

³T. R. Knowles and H. Suhl, *Phys. Rev. Lett.* **39**, 1417 (1977).

⁴F. O. Goodman and N. Garcia, *Phys. Rev. B* **20**, 813 (1979).

⁵L. B. Thomas, in *Rarefied Gas Dynamics*, edited by C. L. Brundin (Academic, New York, 1967), Vol. 1, p. 155.

⁶D. O. Edwards and P. P. Fatouros, *Phys. Rev. B* **17**, 2147 (1978). For a related measurement involving

hydrogen, see T. R. Govers, L. Mattera, and G. Scoles, *J. Chem. Phys.* **72**, 5446 (1980).

⁷M. Sinvani, P. Taborek, and D. L. Goodstein, *Phys. Rev. Lett.* **48**, 1259 (1982); M. Sinvani, D. L. Goodstein, M. W. Cole, and P. Taborek, to be published.

⁸M. Sinvani and D. L. Goodstein, *Surf. Sci.* **125**, 291 (1983).

⁹To show that the surface is "essentially bare" we must first show that S is of order unity. This we do below.

¹⁰Analogous behavior has been seen for Xe on Pt by J. E. Hurst, C. A. Becker, J. P. Cowin, K. C. Janda, L. Wharton, and D. J. Auerbach, *Phys. Rev. Lett.* **43**, 1175 (1979).

¹¹R. Elgin, thesis, California Institute of Technology, 1973 (unpublished).

¹²P. Taborek, *Phys. Rev. Lett.* **48**, 1737 (1982).

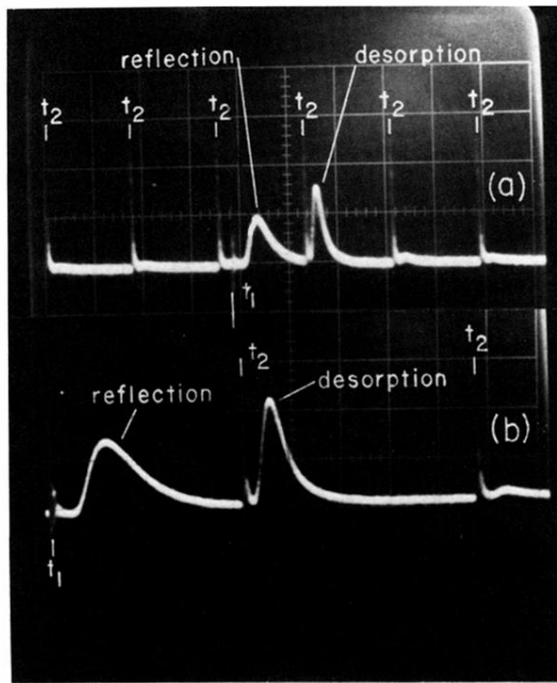


FIG. 4. (a) Signal when h_2 is pulsed at 10- μ sec intervals (times t_2) keeping it bare. h_1 is pulsed at t_1 resulting in reflection signal, followed after next t_2 by desorption of those atoms from h_1 that stuck on h_2 . (b) Same sequence with an expanded time scale.