

Evidence for Universal Quantum Reflection of Hydrogen from Liquid ^4He

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Measurements of the sticking probability $s(T)$ for H on bulk liquid ^4He reveal the onset of the universal $\sqrt{T}$ dependence expected at very low atom temperatures. Studies of $s(T)$ as a function of film thickness clearly demonstrate the influence of the van der Waals-Casimir force due to the substrate, in agreement with recent theories.

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The nature of a collision between an extremely low temperature atom and a surface has been of interest since the earliest days of quantum mechanics [1], but only recently has it become possible to study the problem experimentally. Although a classical analysis reveals that the sticking probability $s(T)$ approaches unity as the temperature T of the incoming atom goes to zero [2], the quantum picture is more complex. Fundamental questions remain unresolved. For instance, it is not known whether in the case of very strong interactions between the atom and the surface excitations $s(0)$ can take arbitrary values or is limited to the values 0 or 1 [3,4]. For interactions of modest strength and finite range, however, there is apparent agreement that $s(T)$ should approach zero as $\sqrt{T}$ [5-7]. This universal signature of quantum reflection, expected to occur when the atom's de Broglie wavelength is large compared to the range of the surface potential, has never been observed experimentally. Measurements by Berkhout *et al.* [8] for hydrogen on helium at temperatures from 145 to 500 mK, as well as results deduced from the related accommodation coefficient [9], revealed that $s(T)$ varied linearly with temperature in that range. Previous measurements in this laboratory of submillikelvin hydrogen on a helium film showed a gradual increase in the sticking probability as the temperature decreased [10]. We report here observations of collisions between hydrogen atoms and bulk helium that clearly reveal the signature of $\sqrt{T}$ behavior.

Our previous measurements prompted Hijmans, Walraven, and Shlyapnikov [11] to propose that atoms colliding with the liquid helium film might be affected by the long-range van der Waals-Casimir potential of the underlying substrate. Calculations of $s(T)$ by Hijmans, Walraven, and Shlyapnikov [11] and Carraro and Cole [12] revealed that the substrate interaction could indeed account for the observed behavior. We have carried out new measurements that allow us to measure $s(T)$ as the film thickness is varied from thin to the bulk liquid limit. In the bulk case we observe the onset of universal quantum reflection. The deviation of $s(T)$ from strict $\sqrt{T}$ behavior is in excellent agreement with theoretical predictions of the exact temperature dependence in this region [11,12]. As the film thickness is reduced, $s(T)$ increases

in a manner consistent with the theory, though these latter studies are essentially qualitative because of uncertainties in the film thickness.

The cold hydrogen atoms used in this experiment are magnetically trapped and evaporatively cooled using techniques described previously [10]. However, the trapping geometry was modified to restrict the surface collisions to a small region at the bottom of the cell. The gas cloud is in the form of a long thin cylinder 0.5 mm in radius and 18 cm long. It is oriented vertically with its lower end 7 cm above the bottom of the sample chamber (see Fig. 1). The gas density on axis is typically 10^{13} atoms/cm³. The temperature of the atoms is determined from the measured energy distribution [13], $n(E)$. Small deviations of $n(E)$ from a thermal equilibrium distribution, possibly due to nonergodic behavior of the atoms in the trap, give rise to an uncertainty in the determination of T that varies from 5% to 20%. The atom temperature in these experiments varies between 300 μK

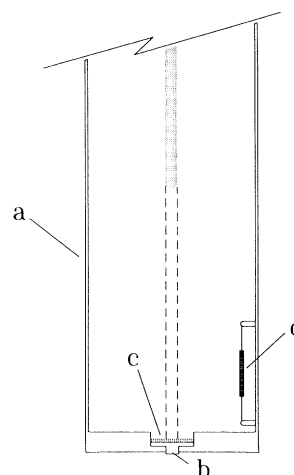


FIG. 1. Schematic diagram of the bottom of the atom trapping cell. (a) Cell wall; (b) copper disk; (c) sintered silver; and (d) bolometer. The shaded region represents the lower portion of the atom cloud in the retracted position. The dashed lines indicate the extension of the cloud when it is brought into contact with the helium surface.

and 4 mK, corresponding to thermal de Broglie wavelengths between 100 and 28 nm.

The bottom of the sample chamber contains a copper insert of 6 mm radius centered below the gas. The upper surface of the copper is covered by a 1 mm thick layer of sintered silver (70 nm nominal particle size). The sinter assures thermal equilibrium between the helium and the copper disk. Liquid helium covers the bottom of the cell and fills the pores of the sinter. The temperature of the copper is measured by a thick film RuO_2 thermometer [14]. The temperature of the copper, and thus the helium, was held at 85 mK. The sticking probability is expected to be independent of the surface temperature as long as the surface temperature is well below the energy of the elementary excitations created when an atom is absorbed. In our case, the absorption of an atom on the surface of the helium results in the creation of a ripplon of energy 0.6 K [6]. We searched for a dependence of $s(T)$ on surface temperature at several atom temperatures and film thicknesses and found none for surface temperatures between 85 and 200 mK.

The thickness of the helium film is estimated by using a bolometer [13] located 1.8 cm above the bottom of the cell and having only a weak thermal coupling to the wall. We measure the heat required to evaporate the helium film from the bolometer, a quantity that is proportional to the thickness of the film. As helium is added to the cell, three distinct regions of behavior are observed. While the sinter is filling, the thickness rises slowly. When the sinter becomes filled, the film is still subsaturated elsewhere; at this point the film thickness begins to rise rapidly. After saturation is reached, the film thickness at the bolometer varies little as helium is added and a puddle of liquid accumulates above the sinter. Based on the height of the bolometer above the puddle, we have taken the saturated film thickness at the bolometer to be 24 nm [15]. This value calibrates our thickness measurements at lower coverages. In presenting the results for subsaturated films we give the film thickness at the bolometer and assume that this is representative of the film thickness at the sinter. Because of this assumption, our values for the thickness of subsaturated films are only qualitative. Above saturation the thickness of the liquid layer is determined volumetrically.

When measuring the sticking probability atoms are brought into contact with the helium on the bottom of the cell by turning off the portion of the magnetic field which defines the lower end of the trapping region. The effective length of the gas cloud is then 25 cm. The time necessary to carry out this 39% increase in the length varies with the strength of the confining field. At the lowest temperature it is about 0.1 s, on the borderline between free (isothermal) and adiabatic (cooling) expansion; at 4 mK it is about 1 s, and the expansion is adiabatic. However, in our geometry an adiabatic expansion of this size would only lower the temperature by 10%.

We have adjusted all our atom temperatures according to our best estimate of the actual cooling; the uncertainty in this correction contributes less than 5% to the quoted temperature errors.

Repeated measurements during the course of the experiment determined that the number of atoms accumulated during a filling and cooling cycle is reproducible to better than 5%. For each film thickness and atom temperature, atoms are held in contact with the surface for varying times. The number of atoms remaining at the end of the contact interval is plotted versus the contact time. Since the sticking process preferentially removes atoms at higher energies, the energy distribution is changed from an equilibrium one by contact with the wall. To account for this, we extrapolate the slope of the decay curve, $N^{-1}dN/dt$, to zero contact time. A small correction is made to the decay rate (always less than 10%) for the two-body relaxation process in the gas. The resulting decay times are analyzed, as discussed below, to determine $s(T)$. The times themselves, however, are striking. For a thick film at a temperature of 300 μK , for example, the atoms' decay time while in contact with the bulk helium surface is 21 s.

The temperature dependent sticking probability is defined as the ratio of the loss rate due to sticking to the collision rate Γ :

$$s(T) = \frac{(-1/N)dN/dt}{(1/N)\Gamma}. \quad (1)$$

In an ideal geometry the wall collision rate is $\frac{1}{4}\bar{v}(A/V)N$ where $\bar{v} = (8kT/\pi m)^{1/2}$ is the mean atomic speed. The effective area to volume ratio A/V in a magnetic trap varies with temperature. We have determined the wall collision rate in our geometry by numerical simulation and converted the results to an effective A/V which is found to vary smoothly from 0.062 cm^{-1} at 300 μK to 0.049 cm^{-1} at 4 mK. The sticking probability data reported here were taken over a period of 8 weeks during which the helium film was completely removed and reformed several times. The reproducibility of the data throughout that period is consistent with the uncertainty in the determination of the decay rate from a sequential set of fills.

Figure 2 shows the sticking probabilities on bulk helium determined by using Eq. (1). The two most recent theoretical predictions [11,12] are also shown. These theories use different parametrizations of the short-range part of the potential, but the major difference in the numerical results may result from the inclusion by Carraro and Cole of higher order corrections to the distorted-wave Born approximation. Although both theoretical curves approach $\sqrt{T}$ behavior as T approaches zero, finite temperature effects cause some flattening of the slope at our temperatures. The slope of the data is in excellent agreement with both theories and, over this limited region of temperatures, can be fit by a temperature exponent of

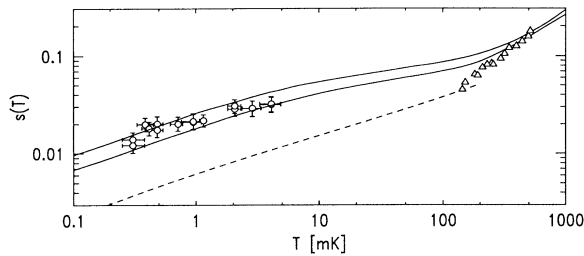


FIG. 2. Sticking probability of atomic hydrogen on thick helium films. $\circ$, our data; $\triangle$, data of Berkhout *et al.* [8]; solid curves, theory of Carraro and Cole [12] (upper curve, $E_B = 1.1$ K; lower curve, $E_B = 1.0$ K); dashed curve [17], theory of Hijmans, Walraven, and Shlyapnikov [11] ($E_B = 1.0$ K).

0.30 ± 0.03 . The magnitude of $s(T)$ is in close agreement with the values calculated by Carraro and Cole [12]. We show their prediction for two slightly different values of the binding energy E_B of a hydrogen atom to the liquid helium surface. (Seven recent measurements of E_B by a variety of techniques [16] give values ranging from 0.96 to 1.06 K with an average of 1.01 K.) It is interesting to note that the values calculated for $s(T)$ by Carraro and Cole would have been higher by about a factor of 2 if the effect of retardation had been omitted in the van der Waals-Casimir potential [18]. The temperature axis in Fig. 2 has been extended to 1 K in order to relate the results to the higher temperature data of Berkhout *et al.* [8]. The difference in slope between the high temperature data and the two theories suggests that at temperatures above 100 mK processes occur that are not adequately described by the current theories of quantum sticking.

Figure 3(a) shows our measurements of $s(T)$ for three representative subsaturated film thicknesses as well as the limiting behavior for thick films. The 10 nm film thickness corresponds to a situation in which the sinter (70 nm diameter) is fully covered, at 3 nm the pores in the sinter have just filled, and at 0.5 nm the pores should be open. The dotted line is representative of the lowest temperature data taken in our previous experiment. In that case the substrate was epoxy and the helium film thickness, determined from the height above a puddle, was approximately 6 nm. Considering the uncertainty in the thickness estimates and the difference in the substrates, the agreement is satisfactory. There is little doubt that the previous measurements were strongly influenced by the substrate. The solid curve is a calculation by Carraro and Cole [12] for a film thickness of 5 nm and a van der Waals coefficient C_3 (for the substrate) of 5.0 K nm^3 . The dashed curve represents the calculation of Hijmans, Walraven, and Shlyapnikov [11] at the same thickness and $C_3 = 4.6 \text{ K nm}^3$. We do not know C_3 for the sintered silver, but for comparison we note that for graphite $C_3 = 4.4 \text{ K nm}^3$ and for copper 5.5 K nm^3 .

Figure 3(b) shows $s(T)$ for 300 μK atoms (thermal de

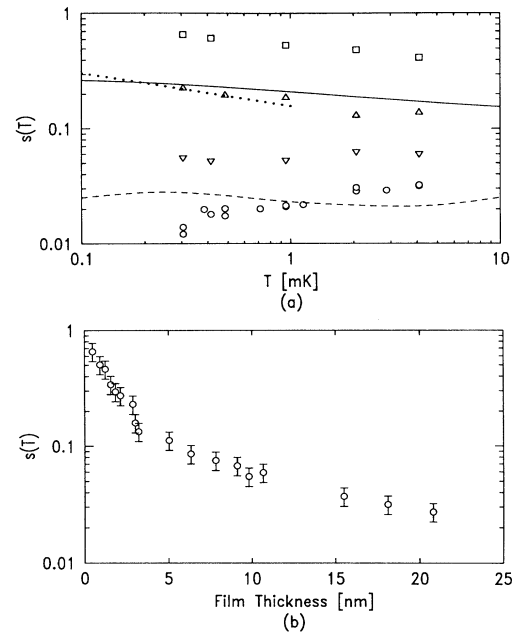


FIG. 3. Dependence of the sticking probability on film thickness. (a) $\square$, 0.5 nm thickness; $\triangle$, 3 nm; ∇ , 10 nm; $\circ$, bulk limit; dotted line, our results from a previous experiment [10]. Theory for a thickness of 5 nm: solid curve [17], Carraro and Cole [12]; dashed curve [17], Hijmans, Walraven, and Shlyapnikov [11]. (b) $s(T)$ as a function of film thickness at a temperature of 300 μK .

Broglie wavelength of 100 nm) as a function of film thickness. There is an abrupt change in slope at a film thickness of about 4 nm. This is close to the thickness where we believe the pores in the sinter fill. Saturation is reached at the bolometer for a film thickness of 24 nm. We continued to measure $s(T)$ up to a helium depth of 1 mm where the data representing bulk behavior (see Fig. 2) were obtained. For comparison, our bulk value for the sticking probability at 300 μK is 0.013(2).

Our results for $s(T)$ for bulk helium are in excellent agreement with theory. A precise study of the influence of thin film and substrate effects on $s(T)$ would require better characterized substrates and a more accurate method for determining the film thickness than were available for this study. Such improvements should make it possible to examine in detail the effects of retardation on $s(T)$, as predicted by theory [11,12]. Nevertheless, our current measurements show that both the magnitude and the temperature dependence of $s(T)$ for thin films are consistent with the most recent theories of quantum reflection.

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 - [18] See the discussion concerning Fig. 3 of the second paper in Ref. [12].