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Influence of the substrate on the low-temperature limit of the sticking probability of hydrogen atoms on He films

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We show that the sticking probability s of hydrogen atoms on the surface of He films in the limit of small incident energy may be determined by the dielectric properties of the substrate, and that considerably higher values of s may be found than in the case of bulk He.

The hydrogen (H) atom lends itself ideally to the study of quantum reflection phenomena off the surface of liquid He. The adsorption potential $U_0(z)$ of a H atom at the surface of liquid He has only one bound state with a binding energy of 1 K. Here z is the distance of the atom to the surface. For vanishingly low energy of the incident H atom, the surface is predicted to behave like a perfect mirror.^{1,2} For finite but sufficiently small incident energy the probability to scatter out of the specularly reflected beam should be dominated by a process in which the atom is adsorbed under emission of a ripplon^{1,2} and desorbed after a residency time which depends exponentially on temperature. This sticking probability s is proportional to the component of the momentum perpendicular to the surface ($s \sim \sqrt{T}$). The contribution of direct (i.e., without adsorption) inelastic scattering by absorption or emission of thermal ripplons is negligible,^{3,4} except at very small momentum transfer (small angle scattering).⁵

The most extensive set of measurements of s to date were performed by Berkhout *et al.*⁶ in the temperature range 0.1 – 0.5 K. These authors did not observe the $s \sim \sqrt{T}$ behavior, indicating that the low-temperature limit was not reached in these experiments. In another set of experiments the dominance of specular reflection at low temperatures was demonstrated by focusing a beam of H atoms with a concave He-coated mirror onto a small aperture.⁷

Zimmerman and Berlinsky¹ (ZB) calculated s using a Morse potential to approximate the atom surface interaction:

$$U_m(z) = \varepsilon[e^{-2\beta(z-z_0)} - 2e^{-\beta(z-z_0)}]. \quad (1)$$

This form for the potential has the advantage of allowing analytical expressions to be obtained, and moreover it closely resembles the variational expression for $U_0(z)$ obtained by Mantz and Edwards.⁸ ZB found the $s \sim \sqrt{T}$ behavior to apply at temperatures below approximately 0.05 K, just below the experimental range of Berkhout *et*

al. A similar approach was followed by Kagan, Shlyapnikov, and Glukhov³ to calculate the accommodation coefficient. These authors used a modified Morse potential with the correct asymptotic behavior:

$$U(z) \rightarrow -C_3/z^3, \quad (2)$$

where¹² $C_3 = 219.7 \text{ K } \text{\AA}^3$. Comparison of these results with those of ZB shows the influence of the algebraic tail to be relatively modest for this value of C_3 . Goldman⁹ investigated the influence of the form of the potential on s numerically. Varying both C_3 and the steepness of the repulsive core as measured by the penetration energy μ_0 , he showed that the value of s at low temperature could be larger than the ZB result by several orders of magnitude, and the limiting behavior may in some cases be only reached when T is less than 1 μK . A close inspection of Goldman's potentials suggests that it is the value of C_3 rather than the value of μ_0 which dominates the limiting value of s at low T . The importance of the attractive Van der Waals tail of the potential for quantum reflection was also stressed by Böheim, Brenig, and Stutski,¹⁰ who used a simplified potential with a hard core repulsion.

Recently, Doyle *et al.*¹¹ published a paper in which measurements of the sticking probability are presented for incident energies in the range 100 μK to 10 mK. These authors report finding anomalously high, and almost temperature-independent values for s (see Fig. 1). This result seems to be consistent with the calculations of Goldman, but only if a value for C_3 is assumed that is more than four times higher than the known exact value. In this paper we shall argue that for thin He films the low-energy limit of s may indeed be dominated by the long-range behavior of the effective atom-surface potential as suggested by Goldman's calculations, but that this is not an intrinsic property of the H-He system but rather of the underlying substrate.

We are interested in the low-energy behavior of the sticking coefficient and consequently we shall confine ourselves to the case where the energy of the incident atom is negligible compared to the binding energy ε_a of the H

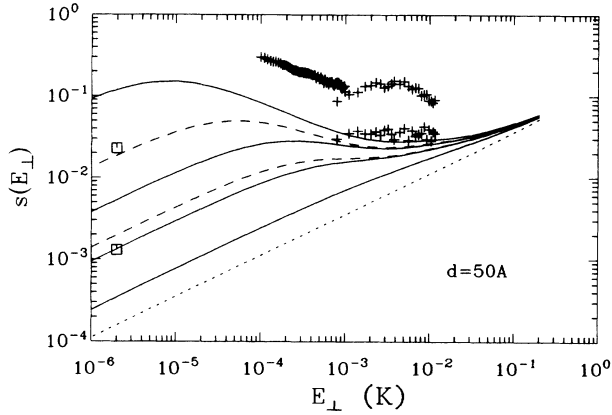


FIG. 1. The sticking probability $s(E_{\perp})$ vs the perpendicular component of the energy of the incident atom for $d = 50 \text{ \AA}$ and varying C_s . Increasing values of s correspond to increasing C_s . Solid curves: $C_s = 0$, $C_s = 12$, $C_s = 20$, and $C_s = 28$. Dashed curves: $C_s = 12$ and $C_s = 20$ but neglecting retardation. Squares: analytical results for $C_s = 12$, and $C_s = 20$. The dotted curve is the result for the Morse potential without the algebraic tail. The crosses are the experimental results from MIT, the two sets of data above 1 mK correspond to two different interpretations of the raw data (Ref. 11).

atom on the He surface. In this case the dominant process is one in which an adsorbed H atom and a ripplon with opposite momenta $\pm \hbar q_0$ are created where q_0 satisfies the energy conservation requirement: $\hbar^2 q_0^2 / 2m + \hbar \omega_{q_0} = \varepsilon_a$, where the ripplon frequency is given by $\omega_q^2 = (\gamma / \rho_0) q^3$. In this limit, the expression found by ZB using first-order perturbation theory, for s of an H atom of incident energy E , reduces to

$$s(E_{\perp}) = \left(\frac{m q_0}{2 \rho_0 \gamma} \right)^{1/2} \frac{1}{\left[\frac{3}{2} \hbar (\gamma q_0 / \rho)^{1/2} + \hbar^2 q_0 / m \right] E_{\perp}^{1/2}} \times \left| \left\langle B \left| \frac{dU_{q_0}}{dz} \right| E_{\perp} \right\rangle \right|^2. \quad (3)$$

Here E_{\perp} is $E \cos(\theta)$ where θ is the angle of incidence of the atom, $\langle z | E_{\perp} \rangle$ and $\langle z | B \rangle$ are the wave functions of the atom in the initial and the bound state, respectively, γ and ρ_0 are the surface tension and the He mass density, and m is the mass of the H atom. The function U_q appearing in Eq. (3) is defined in Ref. 1 and reduces to the static potential U_0 for $q = 0$. The function $|E_{\perp}\rangle$ in the matrix element in Eq. (3) is normalized to unity in a box of length one and the factor $E_{\perp}^{1/2}$ in the denominator appears due to normalization to a unit incident flux.

In order to investigate the influence of the substrate let us first discuss the physical picture. In first approximation (neglecting retardation effects), the replacement of the bulk He below a distance d from the surface by a substrate can be represented by adding the following potential to the one describing the interaction of an H atom with an infinitely thick layer of He:

$$U_d(z) \approx \frac{-C_s C_3}{(z+d)^3}, \quad (4)$$

where C_s is a numerical constant such that $C_s + 1$ is the

ratio of the Van der Waals attraction of the H atom due to the substrate and the infinitely thick He film, respectively. For common substrates C_s is in the range 10–25. First we note that for these values of C_s and reasonable film thickness ($d > 50 \text{ \AA}$) the potential energy near the surface due to the substrate is of the order of a few mK, very small compared to the binding energy of 1 K. Hence, the shape of the wave functions appearing in the matrix element of Eq. (3), as well as the quantity dU_q/dz , is practically unaltered in the range in which $|B\rangle$ is localized ($z \lesssim 10 \text{ \AA}$), and the effect of the substrate on s is entirely due to the altered normalization of the wave function $|E_{\perp}\rangle$ of the incident particle.

Neglecting for the moment the algebraic tail of the H-He potential we may write the Schrödinger equation as

$$-\frac{\hbar^2}{2m} \psi''(z) + (U_m + U_d) \psi(z) = \frac{\hbar^2 k^2}{2m} \psi(z). \quad (5)$$

Here k is the perpendicular component of the wave vector of the incident atom. For small incident energy the right-hand side of Eq. (5) may be set equal to zero in the region of z which we are interested in and the solution of Eq. (5) may be written as

$$\psi(z) = \psi_m(z) \psi_d(z), \quad (6)$$

where ψ_d is the solution to the Schrödinger equation for the pure substrate potential U_d and $(z-a)\psi_m(z)$ is the solution for the pure Morse potential U_m . Here $a \approx 10 \text{ \AA}$ is the characteristic radius of interaction of the H atom with the He film (ka is the phase shift for the wave function in the potential U_m). The representation Eq. (6) holds when $a \ll d$ and implies $\psi_m \approx 1$ for $z > a$. The function ψ_d takes the form

$$\psi_d(z) = \pi k (z+d)^{1/2} (b C_s)^{1/2} \times \left\{ \gamma J_1 \left[2 \left(\frac{b C_s}{z+d} \right)^{1/2} \right] - N_1 \left[2 \left(\frac{b C_s}{z+d} \right)^{1/2} \right] \right\}. \quad (7)$$

Here J_1 and N_1 are Bessel and Neumann functions and $b \equiv 2m C_3 / \hbar^2 \approx 9 \text{ \AA}$. The following choice for γ ensures that $\psi_d \sim z - a$ for $a < z \ll d$:

$$\gamma = N_1 \left[2 \left(\frac{b C_s}{a+d} \right)^{1/2} \right] / J_1 \left[2 \left(\frac{b C_s}{a+d} \right)^{1/2} \right]. \quad (8)$$

When the film thickness tends to infinity ($U_d = 0$) we can approximate ψ for $1/k \gg z > a$ as

$$\psi(z) = k(z-a)\psi_m(z). \quad (9)$$

Expanding Eq. (7) for small $(z-a)/d$, using Eq. (6) and comparing with Eq. (9), the ratio of the sticking probabilities for finite and infinite film thickness is given as the square of the ratios of the wave functions in these two cases:

$$\frac{s}{s_{\infty}} = \left(\frac{b C_s}{a+d} \right) J_1^{-2} \left[2 \left(\frac{b C_s}{a+d} \right)^{1/2} \right]. \quad (10)$$

Replacement of U_m by a potential with the correct asymptotic tail Eq. (2) in the Schrödinger equation Eq. (5) makes the ratio Eq. (10) somewhat smaller.

The enhancement factor Eq. (10) due to the substrate can also be calculated for a fully retarded substrate potential [$U_d \sim 1/(z+d)^4$], yielding qualitatively similar results, but rather than performing this exercise we have numerically solved Eq. (3) using a potential which takes into account the retardation effects in a more realistic manner:

$$U_{0,d}(z) = U_m(z) - \frac{C_3}{z^3} f(\beta[z - z_*]) + U_d(z), \quad (11)$$

with

$$f(x) = \frac{1}{2} \tanh(x) + \frac{1}{2}. \quad (12)$$

We used the values $\varepsilon = 5.14$ K, $\beta = 0.52 \text{ \AA}^{-1}$, and $z_0 = 4.2 \text{ \AA}$ in Eq. (1). The well depth ε is chosen slightly larger than the value used by ZB in order to obtain the correct binding energy of 1 K. The value of z_* is determined by $U_m(z_*) = -C_3/z_*^3$. For $C_3 = 219.7 \text{ K \AA}^3$ we have $z_* \approx 13 \text{ \AA}$. For U_d we use the form

$$U_d(z) \approx \frac{-C_s C_3}{(z+d)^3} g\left(\frac{d_0}{z+d}\right), \quad (13)$$

where the function g , which takes into account the retardation effects, is given by

$$g(1/x) = (1/x)\{1 - (1+x)\exp[-2x]\}. \quad (14)$$

We used a value of 200 \AA for the retardation length d_0 . Equation (11) is a generic form which should apply reasonably well to a wide range of substrates. For comparison with a specific experimental situation in which the dielectric properties of the substrate are known, qualitatively similar but slightly more accurate results can be obtained by using an expression due to Lifshitz.^{13,14} The latter procedure was successfully applied to describe the experimentally observed thickness of He films adsorbed on SrF_2 .¹⁵

In Fig. 1 we show the results of the numerical calculation using Eqs. (3) and (11) for $d = 50 \text{ \AA}$ and various values of C_s (solid curves). The curve for $C_s = 0$ corresponds to bulk He. The dotted line represents the result for the pure Morse potential. For comparison with the analytical results we also performed the calculation neglecting the retardation effects (dashed curves) by setting $g = 1$ in Eq. (13). The analytical results (squares) were obtained by applying Eq. (10) at $E_{\perp} = 2 \times 10^{-5}$ K where the low-energy limit is valid. In Fig. 2 the results are shown for $C_s = 20$ and varying film thickness. The agreement between the numerical calculations and Eq. (10) is quite satisfactory. The dotted curve for $C_s = 28$

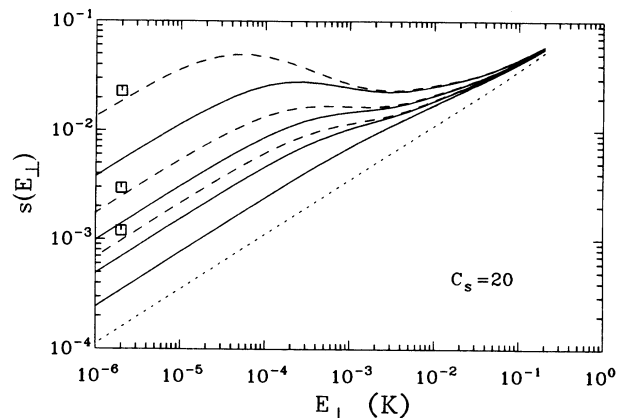


FIG. 2. Same as Fig. 1 but for $C_s = 20$ and varying d . Here decreasing values of s correspond to increasing film thickness. Solid curves: $d = 50 \text{ \AA}$, $d = 70 \text{ \AA}$, $d = 100 \text{ \AA}$, and $d = \infty$. Dashed curves: $d = 50 \text{ \AA}$, $d = 70 \text{ \AA}$, and $d = 100 \text{ \AA}$, neglecting retardation. Squares: analytical results for $d = 50 \text{ \AA}$, $d = 70 \text{ \AA}$, and $d = 100 \text{ \AA}$. The dotted curve is the result for the pure Morse potential.

(nonretarded case) is omitted from Fig. 1 because for this case the vicinity of a resonance renders the present perturbative treatment questionable.

In Fig. 1 we also included the recent MIT results.¹¹ These data have been plotted as a function of E rather than E_{\perp} . Neither the substrate material nor the film thickness are given in the MIT paper, but the results seem to be in qualitative agreement with the present analysis if a large value for C_s and a small value for d is assumed. In particular the latter assumption seems reasonable in view of the long sample cell and the concomitantly high elevation above the liquid level ($> 65 \text{ cm}$). For more quantitative comparison the dielectric properties the substrate used in their experiment would have to be specified.

In conclusion we can say that in order to measure the low-energy sticking coefficient of H atoms on liquid He, the experiments should be performed on sufficiently thick films and preferably on bulk liquid. For thin films and large substrate potentials values of s may be found which exceed the result for bulk He by several orders of magnitude.

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