Kapitza Conductance between Gaseous Atomic Hydrogen and Liquid Helium

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The energy-exchange process between atomic hydrogen gas and liquid ^{4}He has been considered at low temperatures. It is found that previous calculations have substantially underestimated sticking and Kapitza conductance in this system.

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The highly exothermic recombination kinetics of atomic hydrogen make energy exchange a crucial problem of many low-temperature experiments. Because of the finite Kapitza thermal boundary conductance, G_K, substantial temperature steps can develop between the gas sample and the liquid-helium–covered cell walls which confine the gas. The sticking coefficient, s, and accommodation coefficient, a, are also vital and related quantities as they control the kinetics of thermal equilibrium. At a required density and wall temperature for Bose-Einstein condensation (BEC), because of surface recombination, the gas may be so much hotter than the cell walls that BEC cannot be achieved. Recent proposals[1,2] to remove the walls by use of a magnetic trap require low initial temperatures (~ 30 mK) for filling the traps. If s is too large then atoms will stick to the walls and recombine, again thwarting attempts to achieve BEC. In the low-density regime the only experimental value[3] for s is ~ 0.035 at $T \simeq 200$ mK, and the accommodation coefficient is measured[4,5] to be 0.2 to 0.3 for $T = 0.2$–0.5 K. At low temperatures ($T < 50$ mK) calculations[6-9] predict quickly vanishing sticking coefficients and Kapitza conductance: Zimmerman and Berlinsky[6] find $s = 0.049T^{1/2}$, Statt[7] finds $a = 0.063T^{1/2}$, and Kagan, Shlyapnikov, and Glukhov[9] find

$$G_K = 5.6T \text{ mW/K}\cdot\text{cm}^2\cdot(10^{18} \text{ atoms/cm}^3).$$

These authors used a Morse potential instead of the $1/z^n$ form ($n = 3$ or 4) for the long-range part of the surface interaction. (The Morse potential has the distinct advantage that scattering solutions and transition matrix elements can be done analytically.) Whereas at higher temperatures the analytical form used to represent the surface potential is not of critical importance,[10] for energy and temperature scales relevant to the H-He system, this results in substantial quantitative and qualitative differences; as a result, energy-exchange parameters have been greatly underestimated, possibly by orders of magnitude in the temperature region of interest. This is shown in the present calculations; moreover, simple and important relations between a, s, and G_K are presented.

The sticking coefficient and Kapitza conductance of atomic hydrogen on superfluid ^{4}He reported here are for one-ripplon creation in the distorted-wave Born approximation (DWBA). The low-density limit is assumed which means that quantum statistics and high surface coverages are not considered. This allows one to treat the system as one hydrogen atom at temperature T_g interacting with bulk helium with a free surface at temperature T_s.

The Kapitza conductance $G_K(T_g,T_s)$ is defined as $\dot{Q}/(T_g - T_s)$, where $\dot{Q}$ is the heat flux from gas to surface. The energy accommodation coefficient, a, can be defined through the relation

$$G_K(T_g,T_s) = 2k\nu_0(T_g)a(T_g,T_s), \tag{1}$$

where $\nu_0(T)$ is the wall collision rate of the gas $[L^{-1}(kT/2\pi m)^{1/2}$ for one particle of mass m in a box of length $L]$. Equations for heat and particle fluxes can be written in a standard manner[11] in terms of rate equations yielding two distinct channels: nonsticking (NS) and sticking and evaporation (SE). On the assumption that the adsorbed atoms equilibriate with the surface and that a steady-state situation has been reached in which the gas and adsorbate densities are stationary, the accommodation coefficient for the SE channel can be expressed as[12]

$$a^{\mathrm{SE}}(T_g,T_s) = s(T_g,T_s)\left\{1 + \frac{1}{2}\,\frac{\tau(T_g,T_s) - \tau(T_s,T_s)}{T_g - T_s}\right\}, \tag{2}$$

where

$$\tau(T,T_s) = T\,\partial\ln[s(T,T_s)]/\partial\ln T.$$

For small gas-surface temperature differences Eq. (2) linearizes to

$$a^{\mathrm{SE}}(T)=s(T)\left\{1+\frac{1}{2}\beta^2\frac{\partial^2 \ln s(T)}{\partial\beta^2}\right\},\quad \beta=\frac{1}{kT}. \tag{3}$$

The above implies that a knowledge of the sticking coefficient alone is sufficient to obtain all heat-exchange parameters in the SE channel. In fact, the low-temperature results of Refs. 7 and 9 can be simply obtained from the sticking coefficient calculated by Zimmerman and Berlinsky.[6]

For one-ripplon excitations the sticking coefficient, which represents the ratio of sticking events to the number of wall collisions, can be expressed as

$$s(T_g,T_s)=\frac{\rho_0^2}{3\alpha\hbar kT_g}\int_0^\infty d\epsilon_q \sum_\kappa e^{-E_\kappa/kT_g}|\langle\kappa|\phi(z|\mathbf{q})|B\rangle|^2$$
$$\times\{[1+n(\epsilon_q|T_s)]S_\parallel(q;E_\kappa-E_B-\epsilon_q|T_g)+n(\epsilon_q|T_s)S_\parallel(q;E_\kappa-E_B+\epsilon_q|T_g)\}, \tag{4}$$

where **q** is a two-dimensional wave vector in the plane of the surface, and $\epsilon_q=\hbar(\alpha/M\rho_0)^{1/2}q^{3/2}$ is the ideal ripplon dispersion relation with $\alpha=0.378$ erg/cm^2 the surface tension and $M\rho_0=0.145$ g/cm^3 the mass density of liquid ^{4}He. Furthermore,

$$S_\parallel(\mathbf{q};\hbar\omega|T)=\left(\frac{\pi\hbar^2}{E_qkT}\right)^{1/2}e^{-(E_q-\hbar\omega)^2/4E_qkT}$$

represents the Fourier transform of the density-density correlation function for motion of the H atom parallel to the surface, with $E_q=(\hbar^2/2m)q^2$. The ripplon occupation numbers $n(\epsilon_q|T_s)$ give the contribution to stimulated ripplon absorption and emission. They can be neglected for surface temperatures below ~100 mK, implying $s(T_g,T_s)\simeq s(T_g,0)$ for $T_s\leqslant 100$ mK. The states $|\kappa\rangle$ of the H atom are continuum states with energy E_κ and satisfy the wave equation for the direction, z, perpendicular to the surface:

$$\left[-\frac{\hbar^2}{2m}\frac{d^2}{dz^2}+V(z)\right]\psi_\kappa(z)=E_\kappa\psi_\kappa(z), \tag{5}$$

The static H-^{4}He surface interaction, $V(z)$, has been calculated by various authors.[13,14] Mantz and Edwards[14] show that atomic hydrogen binds to the helium surface with one bound state ψ_B having an energy $|E_B|\geqslant 0.63$ K. Experimental values[15–18] for $|E_B|$ range between 0.89 and 1.15 K and seem to be consistent with a single bound state. The long-range part of $V(z)$ should behave as $-c_3/z^3$ and eventually as $-c_4/z^4$ when relativistic retardation effects become important. Another important feature is the limiting value of the repulsive core strength, i.e., the penetration energy μ_0 of the H atom into the bulk liquid. It has been calculated at a fixed ^{4}He bulk density ρ_0 by Guyer and Miller[13] to be 37 K and recently by Kürten and Ristig[19] as a function of the density, who found $\mu_0=75$ K at the experimental $\rho_0=0.0218$ Å^{-3}. This quantity has not been given sufficient attention in the past for the scattering problem. In addition to a Morse potential, the present calculations use three potentials all having a $1/z^3$ long-range behavior with three values of μ_0:

$$V_{\mathrm{I,II}}(z)=\frac{\mu_0}{e^{\beta z}+1}-\frac{c_3z^3}{z^6+z_0^6}\quad(-\infty<z<\infty),$$

with $\mu_0=37$ K, $\beta=0.544$ Å^{-1}, $c_3=911$ K Å^{-3}, $z_0=3.8$ Å ($E_B=-0.872$ K) for V_{I}, and $\mu_0=75$ K, $\beta=1.00$ Å^{-1}, $c_3=645$ K Å^{-3}, $z_0=3.81$ Å ($E_B=-0.828$ K) for V_{II}, and

$$V_{\mathrm{III}}(z)=Ae^{\beta(Z_m-z)}-(c_3/z^3)f(z)\quad(0\leqslant z<\infty),$$
$$V_{\mathrm{III}}(z)=\mu_0-V_{\mathrm{III}}(|z|)\quad(-\infty<z\leqslant 0),$$

with $\mu_0=2930$ K, $\beta=1.415$ Å^{-1}, $A=4567$ K, $c_3=670.25$ K Å^{-3}, $Z_m=4.2$ Å, $Z_0=2.90$ Å ($E_B=-0.670$ K); $f(z)$ attenuates the singularity at the origin caused by the c_3/z^3 term. V_{I} and V_{II} show the sensitivity of the relevant quantities to a variation of μ_0 for a physically expected "soft" surface; V_{III} has a considerably steeper repulsion than I and II. For meaningful comparison, all potentials are constrained to the same well parameters as Ref. 6 (~4.5 K minimum at 4.2 Å).

The H-ripplon coupling $\phi(z|\mathbf{q})$ describes the interaction of the H atom with a sinusoidally distorted liquid surface. Although microscopic calculations containing all necessary correlations are not yet available, one can obtain an *effective* H-He *pair* interaction $\phi^{\mathrm{eff}}(r)$ using $V(z)$ and the surface profile of the liquid. For a step-function profile one can easily show that

$$\phi(z|\mathbf{q})=\int dx\int dy\,\phi^{\mathrm{eff}}((x^2+y^2+z^2)^{1/2})e^{i\mathbf{q}\cdot\mathbf{r}},$$

with

$$\phi^{\mathrm{eff}}(r)=(2\pi\rho_0r)^{-1}\partial^2V(r)/\partial r^2.$$

The matrix elements appearing in the expression for s [Eq. (4)] were calculated by numerical integration. Equation (5) for ψ_B and ψ_κ was integrated to 600 Å in order to obtain reasonable convergence.

Results for the sticking coefficient for $T_s=T_g$ are shown in Fig. 1. The asymptotic $T^{1/2}$ dependence is due to the limiting low-energy behavior of the matrix

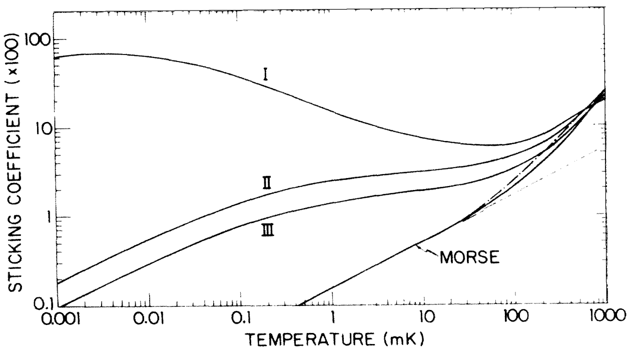

FIG. 1. Sticking coefficients for potentials I,II,III as defined in text and for the Morse potential of Ref. 6. The dashed line represents a pure $T^{1/2}$ dependence. The dash-dotted curve represents the results of Zimmerman and Berlinsky (Ref. 6). (The deviation from the present Morse calculation for temperatures above 50 mK is due to the particular definition of s used by these authors.)

elements $[\langle\kappa|\phi(z|\mathbf{q})|B\rangle \sim E_\kappa^{1/2}]$ and is present for all potentials. Note, however, that the temperature for the *onset* of this dependence is strongly potential dependent and can be in the microkelvin region for hydrogen. Furthermore, even though $V_{\rm III}$ has both a lower binding energy and a steeper repulsion than the Morse potential, it yields significantly higher sticking probabilities. The results for $V_{\rm I}$ display a maximum at very low temperatures. This is not due to an increase in the value of matrix elements with decreasing energies, but rather to a locally amplified contribution to the summation over κ in Eq. (4) arising from a *weak* but still decreasing energy dependence. For this potential one should suspect some breakdown of unitarity in the DWBA. However, it is reasonable to expect large values to persist when unitarity corrections are incorporated. In any case it is clear that for temperatures of present interest parameters for the heat exchange are substantially higher for dispersion potentials and that large uncertainties will remain until the surface interactions are accurately known. Alternatively, the surface potential may be determined by means of energy-dependent scattering experiments.

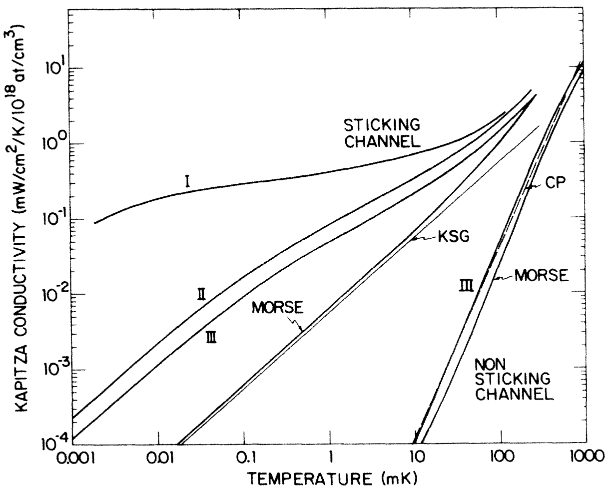

FIG. 2. Kapitza conductance for the sticking and nonsticking channels for various potentials. The results of Kagan, Shlyapnikov, and Glukhov (Ref. 9) (KSG) and Castaing and Papoular (Ref. 8) (CP) are also shown.

Similarly, the accommodation coefficient for the nonsticking channel can be expressed as

$$a^{\rm NS}(T_g,T_s)=\frac{\rho_0^2}{3\alpha\hbar kT_g}\int_0^\infty d\epsilon_q\,\epsilon_q n(\epsilon_q|T_s)\frac{e^{\Delta\beta\epsilon_q}-1}{2k(T_g-T_s)}$$
$$\times\sum_\kappa\sum_{\kappa'}e^{-E_\kappa/kT_g}|\langle\kappa|\phi(z|q)|\kappa'\rangle|^2S_{\parallel}(\mathbf{q};\epsilon_q+E_\kappa-E_{\kappa'}|T_g), \quad (6)$$

where $\Delta\beta=1/kT_s-1/kT_g$.

Figure 2 shows the resulting Kapitza conductivities for both sticking and nonsticking channels as obtained from evaluation of Eqs. (1), (2), (4), and (6). The sticking channel dominates as was predicted by Kagan, Shlyapnikov, and Glukhov.[9] Their results agree with the low-temperature limit of the Morse potential; the dispersion potentials give substantially larger conductivities. The potential dependence for the nonsticking channel is much weaker and the results essentially agree with earlier calculations of Castaing and Papoular[8] who had not explicitly taken any potential into account.

Experimental values for the sticking and accommodation coefficients are available for temperatures ranging from 200 to 500 mK.[4,5,20] The accommodation coefficient in the low-density regime ranges from 0.2 to 0.3 and is consistently higher than the sticking coefficient which is of the order of 0.035. This difference cannot be explained here because the calculated values for a turn out to be very close to those for s in the whole temperature range [the term enclosed in braces in Eqs. (2) and (3) is always of order unity]. Neither can the negative slope of $a(T)$ found in Ref. 5 be reproduced for the temperature range in question.

The low-temperature limit of s has been studied previously by various authors and has given rise to some controversy. Goodman and Garcia[21] maintain that for long-range potentials $s(0,0)$ approaches a finite nonzero limit even in the DWBA. This is not corroborated by the present results. Going beyond the DWBA, Knowles and Suhl[22] have offered a polaron model for enhanced sticking at low energies, but polarization corrections calculated by Brenig and coworkers[23] show a vanishing sticking coefficient at zero energies. It is clear that these aspects should be pursued further for the atomic-hydrogen–helium system.

It has been shown that heat exchange of atomic hydrogen with a liquid-helium surface (especially below 30 mK) is much more effective than previously thought. This is mainly due to both the long-range character and the softness of the surface potential. The calculations presented here were performed on "similar" potentials in order to study various features of the surface interaction, and hard comparison or predictions will have to await an accurate knowledge of this interaction. Further improvements include polarization and unitarity corrections. Calculations for ^{3}He surfaces should also be of interest.

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[1] H. F. Hess, Bull. Am. Phys. Soc. **30**, 854 (1985).

[2] R. V. E. Lovelace, C. Mehanian, T. J. Tommila, and D. M. Lee, Nature (London) **318**, 30 (1985).

[3] R. Jochemsen, M. Morrow, A. J. Berlinsky, and W. N. Hardy, Phys. Rev. Lett. **47**, 852 (1981).

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

[5] K. Salonen, S. Jaakkola, M. Karhunen, E. Tjukanow, 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.

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

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

[8] B. Castaing and M. Papoular, J. Phys. (Paris), Lett. **44**, L-537 (1983).

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

[10] For example, M. D. Stiles and J. W. Wilkins, Phys. Rev. Lett. **54**, 595 (1985).

[11] T. McMullen, J. Low Temp. Phys. **51**, 33 (1983).

[12] V. V. Goldman, to be published.

[13] R. A. Guyer and M. D. Miller, Phys. Rev. Lett. **42**, 1754 (1979).

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

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

[16] M. Morrow, R. Jochemsen, and W. N. Hardy, Phys. Rev. Lett. **46**, 1985 (1980), and **47**, 455(E) (1981).

[17] R. W. Cline, T. J. Greytak, and D. Kleppner, Phys. Rev. Lett. **47**, 1195 (1981).

[18] H. P. Godfried, E. R. Eliel, J. G. Brisson, J. D. Gillaspy, C. Mallardeau, J. C. Mester, and I. F. Silvera, Phys. Rev. Lett. **55**, 1311 (1985).

[19] K. E. Kürten and M. L. Ristig, Phys. Rev. B **31**, 1346 (1985).

[20] 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.

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

[22] T. R. Knowles and H. Suhl, Phys. Rev. Lett. **39**, 1417 (1977), and **40**, 911(E) (1978).

[23] W. Brenig, Z. Phys. B **36**, 227 (1980); J. Böheim, W. Brenig, and J. Stutzki, Z. Phys. B **48**, 43 (1982).