

Thermal accommodation of atomic hydrogen with a liquid- ^4He surface

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The accommodation coefficient is calculated for H atoms scattering from a liquid- ^4He surface. Sticking collisions are found to dominate the accommodation process. Calculated results are approximately a factor of 2 smaller than reported experimental results. A simple model describing the cooling of a flux of H atoms by a cold liquid-He-coated container is also presented.

I. INTRODUCTION

Samples of spin-polarized atomic hydrogen ($\text{H}\downarrow$) gas have been studied to date while confined to liquid-He-coated containers.¹ Most experiments have used ^4He where the binding energy of H to the ^4He surface is $E_B \simeq 1$ K, whereas only a few have used ^3He where $E_B \simeq 0.3$ – 0.4 K. Typical sample temperatures in ^4He -coated vessels have been limited to $T \gtrsim 0.1$ K. At lower temperatures the surface $\text{H}\downarrow$ density is sufficiently high to severely limit the sample lifetime due to recombination events on the surface. Recombination has so far been the chief limiting factor in the quest for Bose-Einstein condensation (BEC) of $\text{H}\downarrow$ gas. The compression experiments^{2–4} performed to date, responsible for the highest $\text{H}\downarrow$ densities achieved so far ($\approx 4.5 \times 10^{18}$ cm^{-3}), are about one to one and a half orders of magnitude away from the BEC transition. Three body dipolar recombination in the gas and on the surface is responsible for limiting the density, due to short sample lifetimes, and for limiting the temperature, due to the large amount of heat released during molecular hydrogen formation. To avoid this problem a possible avenue of research is to work with low density samples at much lower temperatures.

One of the problems associated with lower temperatures is that of thermal accommodation. Hot $\text{H}\downarrow$ gas must be cooled to the ambient temperature of the sample cell. As discussed by Kagan *et al.*⁵ this involves, in principle, the following two types of wall collisions. The first is inelastic collisions with the ^4He surface. This channel is quite inefficient, as mentioned in Ref. 5, when compared to the second channel involving sticking collisions. Here the H atoms adsorb onto the surface in a single bound state where they thermalize (under some circumstances) and are then desorbed. A measure of how well a gas thermalizes with a surface is given by the accommodation coefficient a , defined as

$$a = (Q_i - Q_r) / (Q_i - Q_s), \quad (1)$$

where Q_i is the average incident energy flux, Q_r is the average reflected energy flux, and Q_s is the average energy flux that would result if the reflected atoms had completely thermalized with the surface. Note that in the case of sticking events the "reflected" flux refers to atoms desorbed from the surface.

Experimental and theoretical efforts to determine the

accommodation coefficient between H and ^4He surfaces are apparently in conflict. Salonen *et al.*⁶ measure $a = 0.2 \pm 0.1$ for $0.2 \text{ K} < T < 0.5 \text{ K}$ and Salonen *et al.*⁷ measure $0.2 < a < 0.3$ for $T \simeq 0.4 \text{ K}$. Kagan *et al.*⁵ calculate in the low-temperature limit $a_s \simeq 0.06T^{1/2}$, significantly lower than the experimental results. Castaing and Papoular⁸ have used a classical approach to calculate the inelastic contribution to the accommodation coefficient and find $a_{\text{in}} \simeq 0.11T^2$, significantly larger than the results obtained here. In this paper, a microscopic calculation of the accommodation coefficient is presented which resolves most of this discrepancy. No assumption is made regarding the temperature difference between the gas and the surface, which proves useful when making comparisons with the experimental data of Ref. 7. This calculation is based on the model Zimmerman and Berlinsky⁹ (ZB) used to calculate the sticking probability s for H atoms striking a ^4He surface. It was found in that calculation that the low-temperature limit for s was inadequate much above 10 mK. This proves to be the case for a as well. The following section describes the calculation for both channels of accommodation. Results are presented in Sec. III and a discussion follows in Sec. IV.

II. CALCULATION

Following the treatment in ZB, the liquid ^4He is taken to be an incompressible fluid with a sharp density profile:

$$\rho(\mathbf{r}, z) = \rho_0 \Theta(z + h(\mathbf{r})), \quad (2)$$

where $\rho_0 = 0.145 \text{ g/cm}^3$, \mathbf{r} is a two-dimensional (2D) vector parallel to the surface of area A , z is normal to the surface, and $\Theta(x)$ is the unit step function. Ripplons, the elementary surface excitations of ^4He , cause fluctuations in the height of the surface:

$$h(\mathbf{r}) = (1/\sqrt{A}) \sum_{\mathbf{q}} h_{\mathbf{q}} e^{i\mathbf{q}\cdot\mathbf{r}}, \quad (3)$$

where

$$h_{\mathbf{q}} = (\hbar q / 2\rho_0 \omega_{\mathbf{q}}) (r_{\mathbf{q}}^{\dagger} + r_{-\mathbf{q}}). \quad (4)$$

$r_{\mathbf{q}}^{\dagger}$ is the ripplon creation operator and the ripplon dispersion relation is taken to be

$$\omega_{\mathbf{q}}^2 = gq + (\gamma/\rho_0)q^3, \quad (5)$$

where $\gamma = 0.378 \text{ erg/cm}^2$ is the ^4He -surface tension. The

effect of g , which is due to either gravitational acceleration or van der Waals attraction to the substrate, can safely be ignored in this calculation as long-wavelength ripples do not contribute to sticking.

The effective H-atom- ^4He -surface interaction, taken to be the Mantz and Edwards (ME) potential,¹⁰ can be modeled as a sum over individual H-atom- ^4He -atom potentials $V(r)$ as follows:

$$U(\mathbf{R}, Z) = n_{\text{He}} \int d^2r \int_{-\infty}^{+\infty} dz V(|\mathbf{R}-\mathbf{r}|^2 + (Z-z)^2)^{1/2} \times \Theta(z-h(\mathbf{r})), \quad (6)$$

where n_{He} is the ^4He number density. Expanding $U(\mathbf{R}, Z)$ in terms of h_q yields

$$U(\mathbf{R}, Z) = \sum_{n=0}^{\infty} \frac{(-1)^n}{n!} \prod_{j=1}^n \sum_{q_j} h_{q_j} e^{iq_j \cdot \mathbf{R}_j} \frac{\partial^n U_q}{\partial Z^n} \delta_{q, \sum_j q_j}, \quad (7)$$

where

$$U_q(Z) = n_{\text{He}} \int d^2r e^{iq \cdot \mathbf{r}} \int_{-\infty}^0 dz V[r^2 + (Z-z)^2]^{1/2}. \quad (8)$$

The net energy flux into the surface $Q_i - Q_r$ can be re-

$$P(k_i, \sigma_i; k_f, \sigma_f) = \frac{2\pi}{\hbar} \left| \left\langle \phi_{\sigma_i}(Z) \left| \frac{\partial U_q^h}{\partial Z}(Z) \right| \phi_{\sigma_f}(Z) \right\rangle \right|^2 A^{-1} (\hbar\omega_q / 2\rho_0\omega_q) \times \{ \delta(E(k_i, \sigma_i) - E(k_f, \sigma_f) - \hbar\omega_q) [1 + n_q^r] + \delta(E(k_i, \sigma_i) - E(k_f, \sigma_f) + \hbar\omega_q) n_q^r \}, \quad (11)$$

where n_q^r is the ripplon (Bose) population factor. Now the effective interaction potential is given by

$$U_q^h(Z) = n_{\text{He}} \int d^2r e^{iq \cdot \mathbf{r}} \int_{-\infty}^{+\infty} dz V[(r^2 + (z-Z)^2)^{1/2}]^{1/2} \text{erfc}[z / (2\langle h^2 \rangle)^{1/2}], \quad (12)$$

where $\langle h^2 \rangle$ is the mean-square surface fluctuation. The complimentary error function term is the equivalent of the Debye-Waller factor for neutron scattering.

Estimates of $(\langle h^2 \rangle)^{1/2} \sim 2 \text{ \AA}$ (Refs. 11 and 12) compared to the extent ($\sim 10 \text{ \AA}$) of the H-surface wave function indicate that it is safe to set $\langle h^2 \rangle$ to zero in (12), thus obtaining the same effective interaction potential as used by ZB, Eq. (8). A Morse potential is used to describe the ME effective surface potential as was done in ZB:

$$U_0(Z) = \epsilon(e^{-2\beta(Z-Z_0)} - 2e^{-\beta(Z-Z_0)}). \quad (13)$$

This allows the matrix elements for H-atom adsorption to be calculated analytically (see ZB for results) which are then used to determine the sticking contribution to the accommodation coefficient a_s . Unfortunately the matrix elements for inelastic scattering could not be evaluated analytically. The low-energy limit is proportional to σ , the incident normal wave vector, and can be evaluated once [for $U_0(z)$] in order to determine the low-temperature inelastic contribution to the accommodation coefficient a_{in} . This is acceptable since $a_{\text{in}} \ll a_s$ in the temperature range of interest.

The sticking contribution to the accommodation coeffi-

lated to the transition rate $P_{i,f}$ for an H atom incident on the surface in state i reflecting from the surface in state f as follows:

$$Q_i - Q_r = n_g L \sum_{i,f} P(E_i) P_{i,f}(E_i - E_f), \quad (9)$$

where $P(E)$ is the thermal Boltzmann factor, L is the box normalization length normal to the surface, and n_g is the gas density. First-order time-dependent perturbation theory is used to calculate $P_{i,f}$:

$$P_{i,f} = (2\pi/\hbar) |\langle i | V_{\text{int}} | f \rangle|^2 \delta(E_i - E_f). \quad (10)$$

The H-atom wave function is given by $\Psi_{k,\sigma}(\mathbf{R}, Z) = A^{-1/2} \exp(i\mathbf{k} \cdot \mathbf{R}) \phi_{\sigma}(Z)$, where $\phi_{\sigma}(Z)$ is the surface normal wave function determined from Schrödinger's equation with interaction potential $U_0(Z)$. Unbound surface states have kinetic energy $\hbar^2\sigma^2/2m$ whereas the bound surface state has energy $-E_B$. Now $|i\rangle = \Psi(\mathbf{R}, Z) |\Phi\rangle$, where $|\Phi\rangle$ is the surface ripplon state and $E_i - E_f = E(k_i, \sigma_i) - E(k_f, \sigma_f) \pm \hbar\omega_q$, where $\hbar\omega_q$ is the total energy exchanged with the ripplon bath. Expanding the matrix element in terms of r_q^{\dagger} and summing over all cross terms yields the following first-order term in $U(\mathbf{R}, Z)$, the single ripplon emission/absorption transition rate:

cient will be considered first with the inelastic contribution to follow. An added complication to this problem is the fact that while Q_i depends on the gas density n_g , Q_r depends on n_s , the surface density. During the accommodation process the surface and gas densities are out of thermal equilibrium. In steady state however, the two densities can be related by equating the incident particle flux J_i and the reflected (desorbed) particle flux J_r (ignoring recombination). Inserting (11) and (12) into (9) and performing the thermal average over \mathbf{k} yields the following:

$$J_i = \left\{ \frac{\Lambda^{-3}(T_g)}{2\pi(\gamma\rho_0)^{1/2}} e^{\beta_g \mu_g} \right\} \beta_g \times \int d\sigma \int dq q^{-1/2} \tilde{n}_q |M_{\text{st}}(q, \sigma)|^2 \times \exp \left[-\beta_g \frac{2m}{\hbar^2} \left[\frac{\sigma^4}{4q^2} + \sigma^2 h(q) + f(q) \right] \right], \quad (14a)$$

where

$$h(q) = \frac{1}{2} + \frac{m}{\hbar^2 q^2} (E_B - \hbar\omega_q), \quad (14b)$$

and

$$f(q) = q^2/4 + m/\hbar^2 (E_B - \hbar\omega_q) + (m/\hbar^2 q)^2 (E_B - \hbar\omega_q)^2, \quad (14c)$$

with

$$(1/L) |M_{st}(q, \sigma)| = \left| \left\langle \phi_\sigma \left| \frac{\partial U_q}{\partial Z} \right| \phi_B \right\rangle \right|,$$

$$\tilde{n}_q = 1 + n_q^r, \quad \beta_x = 1/(k_B T_x),$$

$\Lambda(T_x)$ is the thermal deBroglie wavelength and μ_g is the chemical potential of the gas. It has been assumed that Boltzmann statistics apply for both gas and surface atoms. The expression for J_r is similar to (14) except that $T_x = T_s$ instead of T_g in the prefactor and the exponential. In order to equate the incident and reflected particle flux the prefactor in $\{ \}$ will be replaced by the prefactor for J_i scaled by χ , which is chosen such that $J_i = J_r$. Thus the surface density is related to the gas density through χ .

The expressions for Q_i and Q_r are similar to those for J_i and J_r where the following term is included in the integrand of (14a):

$$\beta_x \left[\frac{1}{2} + \beta_x \frac{2m}{\hbar^2} \left[\frac{\sigma^4}{4q^2} + \sigma^2 h(q) + f(q) \right] \right], \quad (15)$$

with $T_x = T_g$ in calculating Q_i and $T_x = T_s$ for Q_r . These four integrals are evaluated numerically and inserted into (1) to obtain a_s . As a check on the numerical integration the low-temperature limit has been calculated for a_s :

$$a_s^{lt} = \frac{5m^2 k_B}{(\gamma\rho_0)^{1/2} \hbar^4 (\bar{v}/T^{1/2})} (M_{st}^0)^2 \frac{\sqrt{2}}{(f''(q^*))^{1/2}} (q^*)^{-1/2} T^{1/2}, \quad (16)$$

where $M_{st}^0 = \lim_{\sigma \rightarrow 0} M_{st}(q, \sigma)/\sigma$, \bar{v} is the average thermal velocity, and q^* is determined by the low-temperature energy-conservation $E_B = (\hbar^2/2m)q^{*2} + \hbar\omega_{q^*}$. Numerical results will be presented in the next section.

Calculating the inelastic contribution to the accommodation coefficient proceeds in a similar fashion (with no need for χ). In this case however, the matrix element could not be calculated analytically. Instead, the low-energy limit has been determined and used to obtain the low-temperature expression for a_{in} :

$$a_{in}^{lt} = \frac{m^3 k_B^2}{2\hbar^6 \sigma_0} (M_{in}^0)^2 T^2, \quad (17)$$

where

$$M_{in}^0 = \lim_{\sigma_i, \sigma_f \rightarrow 0} M_{in}(q, \sigma_i, \sigma_f)$$

and

$$(1/L) M_{in}(q, \sigma_i, \sigma_f) = \langle \phi_{\sigma_i} | \partial U_0 / \partial Z | \phi_{\sigma_f} \rangle.$$

It should be noted at this point that H atoms which do

not stick to or scatter inelastically from the ^4He surface scatter elastically and hence do not contribute to thermal accommodation.

III. RESULTS

Numerical evaluation of the accommodation coefficient is presented in this section. The dominant sticking contribution is considered first, followed by the weaker inelastic scattering contribution.

Three sets of parameters for the effective surface potential have been used in determining a_s . Two sets are the same parameters used by ZB to calculate the sticking probability. A good fit to the ME potential is obtained with $\beta = 0.52 \text{ \AA}$, $\epsilon = 4.48 \text{ K}$, and $Z_0 = 4.2 \text{ \AA}$. A plot of a_s as a function of temperature is shown in Fig. 1 (solid line) using these parameters. One problem with this potential is that its binding energy $E_B = 0.70 \text{ K}$ is considerably lower than the experimentally measured value. Increasing the binding energy by increasing the well depth will also increase the H-ripplon coupling and hence a_s . Choosing $\epsilon = 5.52 \text{ K}$ yields the dash-dotted line in Fig. 1. Although the values for a_s now agree much better with experiment, the values for s are overly large. As a compromise the third parameter set was chosen to be $\beta = 0.52 \text{ \AA}$, $\epsilon = 5.52 \text{ K}$, and $Z_0 = 4.2 \text{ \AA}$ with $V_{int} = \partial U_0(Z)/\partial Z$, thereby artificially lowering the coupling. The resulting accommodation coefficient is also plotted in Fig. 1 (dashed line). All of these curves are calculated in the limit of equal gas and surface temperatures.

Comparing these results with their low-temperature limits indicates the necessity of performing the calculation for arbitrary temperatures. The low-temperature limit for $\epsilon = 4.48 \text{ K}$ is $a_s^{lt} = 0.063 T^{1/2}$ and for $\epsilon = 5.52 \text{ K}$ with zero q coupling is $a_s^{lt} = 0.055 T^{1/2}$. The difference between a_s and a_s^{lt} at higher temperatures ($> 10 \text{ mK}$) is due to both stimulated ripplon emission and an increase in the matrix element over the linear σ dependence.

Experiments measuring the accommodation coefficient require a finite temperature difference between the gas

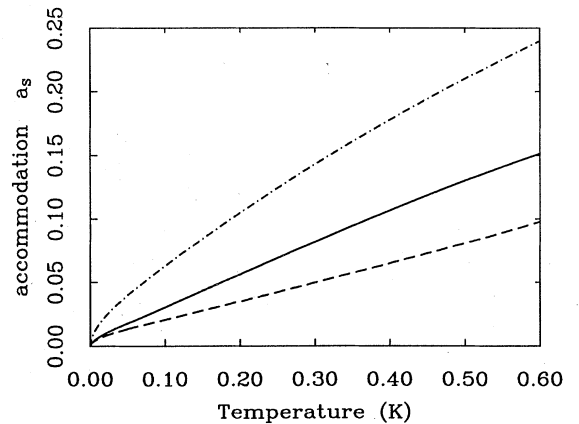


FIG. 1. Calculated values of $a_s(T)$. Solid line used $\epsilon = 4.48 \text{ K}$ with full q coupling dependence. Dash-dotted line used $\epsilon = 5.52 \text{ K}$ with full q coupling and dashed line used $\epsilon = 5.52 \text{ K}$ with zero q coupling.

TABLE I. Comparison of data from Ref. 7, a_{expt} , and this calculation, a_s . In both cases $T_s=440$ mK and full q coupling is used. $\epsilon=4.48$ K in case 1 and $\epsilon=5.52$ K in case 2.

T_g (mK)	a_s (case 1)	a_s (case 2)	a_{expt}
329	0.09	0.16	0.34
378	0.10	0.17	0.28
403	0.11	0.18	0.23

and surface. If this temperature difference becomes overly large, the preceding results are invalid. For this reason a_s is calculated for the gas and surface temperatures reported in Ref. (7) and are presented in Table I.

Now the inelastic contribution to the accommodation coefficient will be discussed. Asymptotic approximations of the Whittaker functions describing the unbound surface wave functions are used to calculate the matrix element M_{in}^0 where $q=0$. Inserting this value of M_{in}^0 into (17) yields $a_{\text{in}}=0.011T^2$. For all temperatures of interest, $a_{\text{in}} \ll a_s$.

IV. DISCUSSION

The results of the preceding section will now be compared with the available experimental results. Several comments regarding inelastic scattering will follow. Finally, a simple model is used to simulate the cooling of an incident flux of H atoms by a He-coated container using the calculated accommodation coefficient. The implications these results have on future low-temperature experiments will be discussed.

The calculated results are approximately a factor of 2 lower than the experimental results of Refs. 6 and 7. The larger error bars on the first measurement⁶ do not allow for a check on the temperature dependence. Both experiments favor the larger of the calculated results. This is contrary to the case of the sticking coefficient where the experimental data agreed better with the lower calculated results (see ZB). Note that the integral in a_s weights the high-energy part of the coupling more than the integral involved in the sticking coefficient. This suggests a stronger coupling for large q than that used in these models.

Data presented in Ref. 7 is summarized in Table I. The calculated results are almost independent of T_g with constant T_s . Nevertheless there does not seem to be any way to explain the experimentally observed rise in a as T_g is lowered.

As pointed out in Ref. 7, experimental values of a are considerably larger than s . This is consistent with the calculated results. It is not necessary to invoke other scattering mechanisms, such as inelastic scattering, to account for this fact. In particular, the results presented here indicate that inelastic scattering due to single ripplon emission or absorption is totally insignificant in the accommodation process. Higher-order ripplon processes are also negligible, both for sticking and inelastic scattering. For example the ratio of double ripplon emission to single ripplon emission for sticking is $\approx \beta^2 \hbar / [6\pi(\gamma\rho_0)^{1/2}](q^*)^{3/2} \approx 0.2$.

Thermal accommodation of atomic hydrogen gas at low temperatures will be inhibited by recombination on the surface which limits the number of atoms available for desorption. Clearly a larger contribution to a from inelastic scattering would be desirable. Scattering of ^4He atoms from ^4He surfaces¹¹ also indicate that inelastic scattering is insignificant when compared to elastic scattering. In the case of ^4He -atom scattering, most of the atoms are absorbed into the liquid whereas most of the H atoms are elastically scattered from the liquid. A possible way to increase inelastic scattering is suggested by the experiment of Echardt *et al.*,¹³ where ^3He impurities, residing in a surface state on the ^4He surface for temperatures < 100 mK, were responsible for a considerable fraction of inelastic scattering. In the case of H scattering however, the H-atom wave function is repelled by the surface whereas the ^3He surface-state wave function is very close to the surface (see Ref. 10). The resulting small overlap suggests a small effect on inelastic scattering of H from ^3He impurities on the ^4He surface.

Knowles and Suhl¹⁴ have shown that polaronic effects are responsible for large accommodation coefficients for He scattering from metal surfaces at low temperatures. This is unlikely to apply to H scattering from ^4He surfaces, as suggested in Ref. 7, because of the weak coupling to the surface excitations. As pointed out in Refs. 15 and 16, H polaronic effects are unimportant.

In conclusion, the results presented here portray a reasonably accurate picture of the accommodation process for H atoms with a liquid ^4He surface. With the advent of more precise temperature-dependent data for a and s various models for the coupling might be tested. These models would also have to be consistent with the measured binding energy.

Finally, a simple model is used to describe the effect the calculated accommodation coefficient has on cooling a constant flux ϕ of atoms, initially at temperature T_{g0} by a liquid-He-coated container (1-cm cube) at temperature T_{s0} . A dilute mixture of ^3He in liquid ^4He is assumed to cover the container walls with $E_B=0.4$ K and a Kapitza resistance of $R=60T^3$ WK⁻¹ between the He and Ag sintered walls (1 mm thick). The temperature of the He film T_s is then determined solely by the heat of recombination due to three-body dipolar recombination on the surface.^{17,18} The heat of recombination is assumed to be totally absorbed by the He film. A balance between the flux of atoms sticking to, recombining on and desorbing from the surface is determined from ϕ and the low-temperature limits of J_i and J_r . The gas temperature T_g is calculated by taking into account the energy flux associated with ϕ and the heat accommodated by the He surface, again using the low-temperature limit for a_s . The calculated values of a_s and s for a ^4He surface are used. Experiment¹⁹ indicates that the sticking coefficient for ^3He surfaces is about a factor of 2 less than that for ^4He surfaces. Hence this assumption is likely to overestimate the accommodation provided by the $^3\text{He}/^4\text{He}$ film.

Several numerical simulations were performed to find the steady-state conditions during constant loading. Initial conditions are: zero atoms in the cell with $T_s=T_{s0}$. As the atoms enter the cell the surface and gas densities

increase to steady-state values while T_s increases as well. The initial simulation used $T_{s0}=10$ mK, $T_{g0}=100$ mK and $\phi=10^{12}$ atom/s. The final surface and gas temperatures are $T_s=14$ mK and $T_g=74$ mK with $n_s=6.8\times 10^{11}$ cm $^{-2}$ and $n_g=1.2\times 10^{10}$ cm $^{-3}$. Steady-state conditions are reached when the incident flux equals the recombination rate, determining the surface density. At low surface temperatures this results in a disappointingly low gas density. In addition, accommodation via sticking collisions with the walls is largely ineffective in cooling the gas leaving $T_g \gg T_s$. Raising the flux to 10^{13} s $^{-1}$ increases n_g by a factor of 10, $T_g=84$ mK while $T_s=23$ mK and $n_s=1.4\times 10^{12}$ cm $^{-2}$.

These dismal results prompted the suggestion of using an intermediate cooling stage to assist in the accommodation at low temperatures. This intermediate stage is taken to have a surface temperature of 25 mK and it is assumed that the flux of atoms leaving this stage, into the 10-mK sample cell, is fully thermalized. With $\phi=10^{13}$ s $^{-1}$, the steady-state values for the intermediate stage are $T_{si}=28$ mK, $T_{gi}=44$ mK, $n_{si}=1.3\times 10^{12}$ cm $^{-2}$, $n_{gi}=7.7\times 10^{11}$ cm $^{-3}$. Now taking the flux of atoms into the 10-mK cell to be 10^{12} s $^{-1}$ yields $T_g=43$ mK, $T_s=14$ mK, $n_s=6.8\times 10^{11}$ cm $^{-2}$, and $n_g=2.0\times 10^{10}$ cm $^{-3}$. Evidently the intermediate accommodation stage is largely inef-

fective in reducing the heat load on the gas in the sample cell. Reducing the flux into the second stage to 10^{11} s $^{-1}$ decreases T_g to 40 mK.

The results of this simple model do not bode well for experiments trying to achieve Bose condensation below 100 mK. Cooling the gas below 40 mK with liquid-He-coated walls does not seem feasible using reasonable values for the H flux. This may pose a problem for the two recently proposed magnetic trapping schemes^{20,21} which rely on a source of atoms at 20 or < 1 mK (Refs. 20 and 21, respectively). Future experiments measuring the accommodation coefficient below 100 mK are clearly needed. As pointed out in Ref. 21, an alternative is to transport cold H atoms on the He surfaces, the problem then being of how to eject them from the surface without significantly warming them.

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