ture-dependent hysteresis is not understood. While the physics of this system is essentially determined by fluid mechanics and electrostatics, the nonlinearity of the problem also results in many phenomena relevant to microscopic two-dimensional electron systems.

The authors appreciate numerous discussions with C. C. Grimes and P. M. Platzman.

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$$I(\vec{r}+\gamma\nabla a)/I_0=[1+\gamma(a_{xx}+a_{yy})+\gamma^2(a_{xx}a_{yy}-a_{xy}^2)]^{-1},$$

$\gamma=l/(n-1)$, where I_0 is the intensity when $a=0$, l is the distance from the liquid surface to the screen, n is the index of refraction of the liquid helium, and the subscripts x and y stand for the partial derivatives with respect to those quantities. The derivatives of a must be evaluated at $\vec{r}$ $(\equiv x\hat{x}+y\hat{y})$.
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Interaction of Hydrogen Atoms with Helium Films: Sticking Probabilities for H on ^{3}He and ^{4}He, and the Binding Energy of H on ^{3}He

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(Received 3 April 1981; revised manuscript received 28 July 1981)

Magnetic resonance at 1420 MHz in zero magnetic field and for $0.06<T<0.5$ K has been used to determine the binding energy of H on liquid ^{3}He, the rate constant for recombination and the frequency shift for H on ^{3}He, and the sticking probability for H on ^{3}He and ^{4}He. The binding energy for H on liquid ^{3}He is found to be 0.42 ± 0.05 K, and the sticking probabilities are 0.035 for H on ^{4}He and 0.016 for H on ^{3}He.

PACS numbers: 67.40.-w, 67.70.+n, 82.65.My, 95.70.+i

Because hydrogen atoms bind strongly to any solid surface, all low-temperature experiments on atomic hydrogen gas[1-6] must be performed in containers which are coated with a liquid-helium film. Hydrogen also sticks to liquid helium, but the binding energy is much smaller than for solids. Earlier magnetic-resonance studies in our laboratory[4] have shown that the binding energy E_B for H on liquid ^{4}He is 1.15 ± 0.05 K. A similar but somewhat lower result, $E_B=0.89\pm0.07$ K, has been reported by Matthey, Walraven, and Silvera.[5] In this Letter we show that the binding energy of H on liquid ^{3}He is about 0.4 K in agreement with very recent results of van Yperen *et al.*[6] at high magnetic field. It would appear that this value represents the minimum possible binding energy for H atoms on the walls of a container.

The minimum value of E_B has important consequences for experimental attempts to observe Bose-Einstein condensation in a high-density gas of spin-polarized H. Because the surface state lies below the continuum, there is a strong tendency at low temperature for H atoms to reside on the surface. The surface density is eventually limited by repulsive interactions between H atoms, and only then can the addition of more atoms lead to a macroscopic occupation of the lowest-energy bulk states of the gas. The saturated surface density n_w^s is proportional to E_B, $n_w^s=\gamma E_B$, where[7,8] $\gamma=(0.5-1.0)\times10^{14}$ K^{-1} cm^{-2}. The main obstacle to the achievement of Bose-Einstein condensation then lies in the fact that such high surface densities are unstable against the recombination process $H+H\rightarrow H_2$ on the wall.

We also report measurements of the wall recombination cross section λ for H on ^{3}He in zero magnetic field which we find to be nearly equal to that for H on ^{4}He. Direct comparison to the high-

field results of van Yperen *et al.*[6] provides a quantitative test of the hyperfine mixing theory of the magnetic field dependence of λ,[5,9,10] which predicts that $\lambda(B)=2\epsilon^2\lambda(0)$, where $\epsilon=a/2\hbar\gamma_e B\approx(253\ \mathrm{G})/B$. For $B=80$ kG, the experimental ratio $\lambda(0)/\lambda(B)$ is very close to the factor of 5×10^4 predicted by this theory. It is then an elementary exercise to calculate the lifetime of an atom in high field on a saturated ^{3}He surface. For plausible conditions, $T=0.1$ K and $B=100$ kG, this time is less than a millisecond.

One can also envision the use of H atoms at low temperatures as a probe of the elementary excitations of ^{3}He and ^{4}He films via atomic scattering experiments. An important parameter in such experiments is the sticking probability α which is the probability that an atom incident on the surface will scatter into a surface state. By analyzing the temperature dependence of the magnetic-resonance line shape we have obtained the first measurements of α for H on ^{3}He and ^{4}He films.

1420-MHz hyperfine-resonance measurements were performed in the apparatus used for our earlier work.[4] An error in the calibration of the Ge resistance thermometer, which affected our earlier results, has been corrected by recalibrating the thermometer *in situ* against a National Bureau of Standards SRM 768 superconducting fixed-point thermometer together with a cerium magnesium nitrate thermometer. For the ^{3}He runs, the sample consisted of 0.1 atm of H_2 and 0.5 atm of pure ^{3}He at STP. Recombination data were obtained by monitoring the decay in amplitude of the resonance signal as a function of time after the discharge pulse which created the atoms. In all cases the decay of the atom density followed a second-order rate equation $dn_H/dt=-Kn_H^2$. The values for the recombination rate constant K, plotted as $\ln K\sqrt{T}$ vs $1/T$ for $0.06<T<0.5$ are shown in Fig. 1.

For $1/T>5$ K^{-1} recombination is dominated by the wall process H + H + wall $\rightarrow$ H_2 + wall. Since the fraction of atoms on the wall is always much smaller than 1 in our experiments, the rate constant for wall recombination[4] can be written as

$$K=\lambda v_s(A/V)\Lambda^2\exp(2E_B/k_BT), \qquad (1)$$

where $v_s=(32k_BT/3\pi m_H)^{1/2}$ and λ is the two-dimensional analog of a collision cross section. The thermal de Broglie wavelength $\Lambda=h/(2\pi m_H k_B T)^{1/2}$ has the value $(17.4\ \text{Å})/T^{1/2}$, and A and V are, respectively, the surface area and volume of the container ($A/V=4$ cm^{-1}). The solid line in Fig. 1 is a fit to the data with slope $2E_B$, where E_B

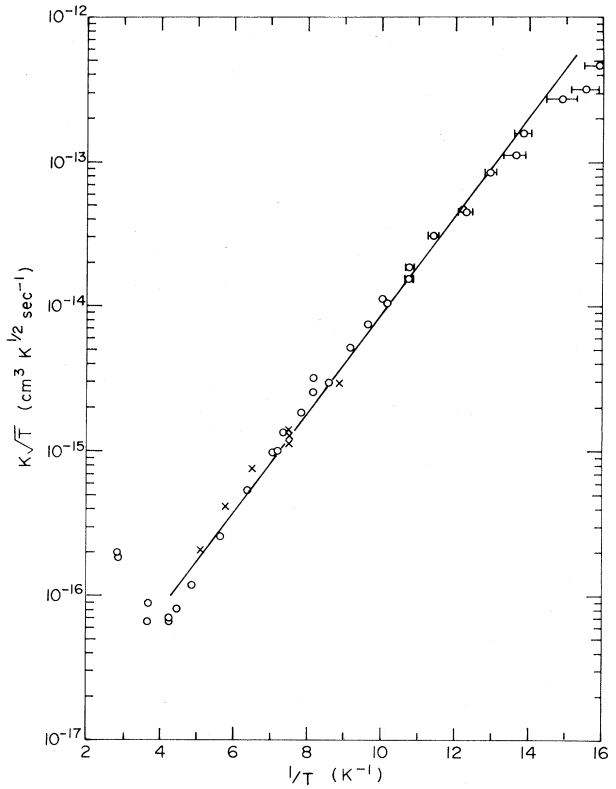

FIG. 1. The recombination rate of H on liquid ^{3}He plotted as $KT^{1/2}$ vs $1/T$. The solid line is a fit to the data with use of a wall recombination cross section $\lambda=0.18$ Å and a binding energy for H on ^{3}He $E_B=0.39$ K, as described in the text. The increased recombination for $1/T<4$ is due to three-body collisions involving ^{3}He atoms in the gas as discussed in Ref. 2.

$=0.39\pm0.04$ K, and a surface recombination cross section $\lambda=0.18\pm0.03$ Å. The horizontal error bars for $1/T>11$ indicate the uncertainty in our thermometer calibration.

The crosses in Fig. 1 are the results of van Yperen *et al.*[6] at high magnetic field ($B=80$ kG) on the surface of a mixture of ^{3}He and ^{4}He. In order to plot them on the same scale as our zero-field results we have divided the high-field results by the expected ratio of the cross sections $2\epsilon^2$ and renormalized according to the ratio of A/V for the two experiments which leads to an overall factor of 10^4. In spite of the fact that a fit to their data gives the slightly lower value $E_B=0.34\pm0.03$ K it is clear that the high-field data agree quite well with our zero-field results both in temperature dependence and in absolute magnitude.

Further measurements would be required to determine whether the apparent slight curvature

of the data results from a real temperature dependence of λ as proposed by Greben, Thomas, and Berlinsky[10] for gas-phase recombination.

Next we consider the magnetic-resonance lineshape data. The atoms in the bulk undergo essentially no collisions, and hence their frequency is given by the unperturbed hyperfine frequency f_0. For a homogeneous surface the frequency of an atom on the wall is $f_0 - \Delta_s$. It is reasonable to assume that the distribution of times *on* the wall, and hence the distribution of phase shifts, is described by a Poisson distribution. This is equivalent to saying that the atom thermalizes in a small fraction of its residency time. Under these conditions, the free induction decay (FID) may be written as

$$S(t) = S(0)\exp(i\omega_0 t)\sum_{n=0}^{\infty} p(n,t)/(1-i\varphi)^n, \qquad (2)$$

where $p(n,t)$ is the probability that an atom has stuck to the wall n times in the interval $(0,t)$, $\omega_0 = 2\pi f_0$, and φ is the average phase shift per sticking $\varphi = 2\pi\tau_s\Delta_s$, with τ_s the average time on the wall. If the distribution of times *between* wall stickings also obeys Poisson statistics with an average time τ_B, then the sum in Eq. (2) can be done exactly. The result[4] is

$$S(t) = S(0)\exp[i(\omega_0 - \Delta\omega)t]\exp(-t/T_2); \qquad (3)$$

$$\Delta\omega = (1/\tau_B)\varphi/(1+\varphi^2)$$

$$\approx 2\pi\Delta_s(\tau_s/\tau_B) \quad \text{for} \quad \varphi \ll 1, \qquad (4a)$$

$$1/T_2 = (1/\tau_B)\varphi^2/(1+\varphi^2). \qquad (4b)$$

The ratio $\tau_s/\tau_B = (\Lambda A/V)\exp(E_B/k_B T)$ is equal to the ratio of the number of atoms on the wall to the number of atoms in the bulk which is always small. The average time between *stickings* τ_B is determined by the average time between wall *collisions* τ_c and the sticking probability $\alpha = \tau_c/\tau_B$. For our apparatus we use $\tau_c = 7.4\times10^{-5}T^{-1/2}$ sec as determined by computer simulation.

Equations (4) imply a maximum frequency shift $\Delta\omega_{\max} = 1/2\tau_B$ and a maximum decay rate $1/T_2 = 1/\tau_B$. Each of these quantities yields an independent measure of τ_B and hence of α. Figure 2 shows data for H on ^{4}He, extending our earlier results[4] to lower temperature. A good fit to both $\Delta\omega$ and $1/T_2$ is obtained using Eqs. (4) with $E_B = 1.15$ K, $\Delta_s = 49$ kHz, and $\alpha = 0.04 \pm 0.004$.

Analogous results for H on ^{3}He are shown in Fig. 3. In the inset we show the exponential behavior of $\Delta\omega\sqrt{T}$ vs $1/T$, which is expected for $\varphi \ll 1$ as indicated in Eq. (4a). A value $E_B = 0.43 \pm 0.02$ K was found for the binding energy and Δ_s

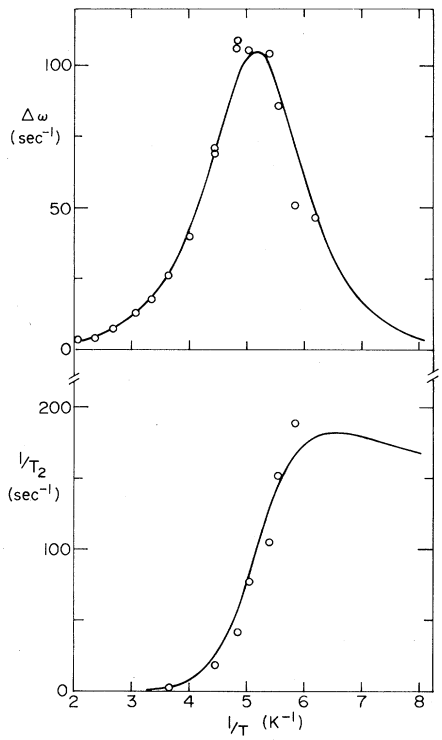

FIG. 2. The hyperfine frequency shift $\Delta\omega$ and linewidth $1/T_2$ of the magnetic-resonance signal of H atoms on a ^{4}He surface. The solid lines are calculated from Eqs. (4) with the parameters $E_B = 1.15$ K, $\Delta_s = 49$ kHz, and sticking coefficient $\alpha = 0.035$ for H on ^{4}He.

$= 23 \pm 2$ kHz. The dashed line in Fig. 3 uses these parameters, and also $\alpha = 0.011$. There is a noticeable discrepancy for $1/T > 11$ K^{-1}. An important feature of the ^{3}He data, which differs from the ^{4}He data, is the occurrence of nonexponential free induction decays, resembling a sum of two or more exponentials, at low temperature ($T < 100$ mK). Such line shapes are not easily attributed to "extra" relaxation mechanisms since these would normally just add to $1/T_2$. In terms of the model described by Eq. (2), a very plausible interpretation of the nonexponential FID's is to attribute them to a nonexponential distribution of times between sticking. If α were close to 1 this would not be surprising since the distribution of arrival times at the wall must certainly be sensitive to the shape of the container. For small α, a nonexponential distribution of times between stickings could arise if α depended strongly on the angle of incidence, especially if nonsticking collisions were elastic and specular.

If the distribution of times between stickings is modeled by a sum of two exponentials, Eq. (2)

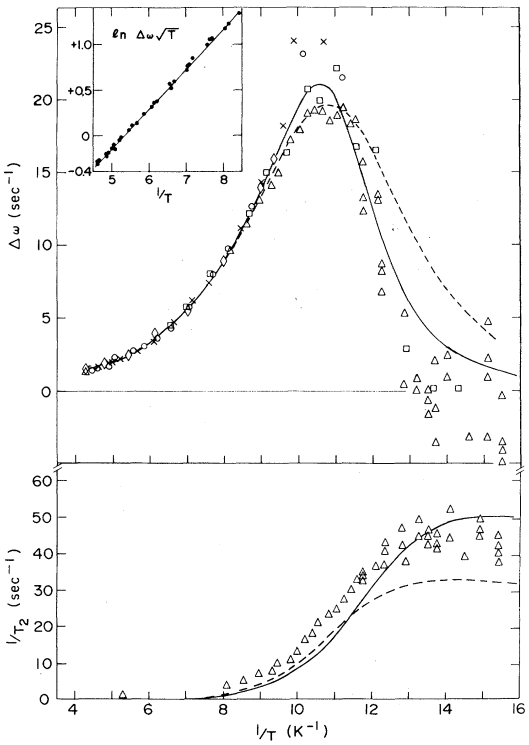

FIG. 3. The hyperfine frequency shift $\Delta\omega$ and the linewidth $1/T_2$ of the magnetic-resonance signal of H atoms on a ^{3}He surface. The fits are discussed in the text.

can be evaluated to obtain the resonance line shape in terms of the parameters of this distribution together with α, Δ_s, and E_B. The parameters of the distribution function are obtained from the lowest-temperature FID's. Δ_s and E_B are obtained from the high-temperature ($\varphi \ll 1$) data and α is chosen to give the best overall fit. Frequency shifts $\Delta\omega$ and initial decay rates $1/T_2$ obtained from this model are shown in Fig. 3 by the solid lines. This fit uses $\Delta_s = 23$ kHz, $E_B = 0.435$ K, and $\alpha = 0.016$. We estimate the uncertainty in α to be about $\pm 30\%$.

In summary, we find that the binding energy of H on ^{3}He is 0.42 ± 0.05 K, and this small value of E_B has allowed us to perform magnetic-resonance measurements down to 60 mK. The cross section λ for surface recombination on ^{3}He is about the same as for ^{4}He [$\lambda(^3\text{He}) = 0.18 \pm 0.03$ Å and $\lambda(^4\text{He}) = 0.20 \pm 0.03$ Å] and the magnetic field dependence, obtained by comparison with the results of Ref. 6, is as predicted by the hyperfine-mixing theory of recombination.[10] This again raises the question of how the lowest two hyperfine levels come to equilibrium at high magnetic field, since unless T_1 for these levels is surprisingly short it should limit the recombination rate. van Yperen *et al.*[6] find no evidence for such a T_1 bottleneck. However, further investigation, particularly by high-field magnetic resonance, would be useful.

Finally, we have measured the sticking probability for H on helium films. We find that α is small and different for the two isotopes [$\alpha(^4\text{He}) = 0.035 \pm 0.004$ and $\alpha(^3\text{He}) = 0.016 \pm 0.005$]. These results suggest that H atoms are potentially the ideal probe for inelastic scattering studies of the elementary excitations of helium films.

This work was supported in part by the Natural Sciences and Engineering Research Council of Canada. One of us (A.J.B.) would like to acknowledge the support of the Alfred P. Sloan Foundation. We thank W. G. Unruh for his solution of the summation in Eq. (2) for general forms of the distribution of τ_B's, and we also acknowledge useful discussions of the magnetic-resonance line shape with S. B. Crampton. Thanks are due to Dr. J. A. Galt of the Dominion Radio Astrophysical Observatory, Penticton, British Columbia, for assistance in the calibration of our frequency standard, and to Bryan Statt for assistance and discussions at various stages of the experiment.

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