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Measurement of Pressure of Gaseous H_1 : Adsorption Energies and Surface Recombination Rates on Helium

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Pressure decay measurements are used to study adsorption and recombination of atomic hydrogen on liquid helium surfaces below 1 K. Recombination is suppressed by five orders of magnitude by applying a 10-T magnetic field. The surface-recombination-rate constant (K_s) and the adsorption energy (ϵ_a) of H_1 on ^4He are $K_s B^2/T^{1/2} = (2.6 \pm 0.8) \times 10^{-7} \text{ cm}^2 \text{ T}^2 \text{ K}^{-1/2} \text{ sec}^{-1}$ and $\epsilon_a/k_B = 0.89 \pm 0.07 \text{ K}$. An estimate of $\epsilon_a/k_B = 0.59 \text{ K}$ is obtained for H_1 on a dilute ^3He - ^4He mixture.

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We have recently shown that atomic hydrogen can be stabilized for periods of hours in a low-temperature cell when all surfaces are covered with a superfluid film of ^4He .^{1,2} It has subsequently been created in three other laboratories.³⁻⁵ In a magnetic field B the hydrogen is spin polarized and referred to as H_1 . H_1 is predicted to undergo Bose-Einstein condensation (BEC) if sufficiently high densities can be achieved.⁶ However, processes which limit the highest densities^{2,3} of H_1 have not been identified. In this Letter, we provide the first experimental proof that magnetic fields stabilize H_1 against recombination to H_2 . We show surface recombination to be the dominant recombination process in our system. We also measure the adsorption energy of H_1 on ^4He finding $\epsilon_a/k_B = 0.89 \pm 0.07 \text{ K}$. This is consistent with the theoretical estimate of Mantz and Edwards⁷ and with very recent zero-field measurements of Morrow *et al.*⁸

The large value of ϵ_a combined with surface recombination rates create a serious barrier against achieving conditions for BEC. Measurements with a very dilute ^3He - ^4He surface coverage yield a significant and promising reduction of ϵ_a . Although H_1 is predicted to be the only elemental material that remains a gas⁶ at $T=0 \text{ K}$, there have only been indirect determinations of its state. We have directly measured the pressure, p , of H_1 using a sensitive capacitance manometer providing the most direct evidence that H_1 is a gas for $T \approx 0.27 \text{ K}$, our lowest accessible temperature. For densities studied, $n_g \lesssim 1 \times 10^{16} \text{ atoms/cm}^3$, we have shown that H_1 behaves as a perfect gas, i.e., $p = n_g k_B T$.

The experimental techniques have been described in some detail elsewhere.^{1,2} Here we shall give a brief description with detail of new developments. Atomic hydrogen from a microwave discharge flows via a tube into a cell at

temperatures as low as 0.27 K in which all surfaces are covered with superfluid ^4He . The cell is in the center of a solenoidal magnetic field of maximum value 11 T which serves to polarize the hydrogen and confine it in the cell by magnetic compression. The H_1 is further confined by a helium vapor compressor (HEVAC) located in the fill tube. In earlier experiments the H_1 was detected by a bolometric technique: simple and extremely sensitive but with the disadvantage that it destroys the sample, resulting in tedious time-consuming measurements. We have now developed a sensitive capacitance manometer which measures the pressure of H_1 and is nondestructive. The gauge, which is an integral part of the sample cell, is shown in Fig. 1(a). The flexible membrane is a 12.5- μm -thick gold-plated Kapton sheet attached with Stycast-1266 epoxy. The capacitance was measured with an ac bridge and calibrated at low temperature against a Barocell pressure gauge with use of ^4He gas. The manometer would normally measure the pressure due to H_1 and ^4He in the chamber. A superleak connecting the vacuum side of the membrane to the H_1 side equalized the ^4He pressure, so only the H_1 pressure was measured. This could also serve as a detector of superfluidity in H_1 if it occurs. The sensitivity of the manometer was measured to be 11.1 pF/Torr with a minimum detectable pressure of 10^{-6} Torr corresponding to a resolution $\Delta c/c = 1.7 \times 10^{-7}$. A bolometer detector is also built into the cell to destroy the H_1 in establishing the capacitance base line. The cell has a volume $V = 1.7 \text{ cm}^3$ and an area $A = 32 \text{ cm}^2$.

Our current experimental objectives are to study the properties of H_1 in contact with helium surfaces and in high magnetic fields to understand the limitations and conditions for achieving the higher densities required for BEC and superfluidity of this Bose gas. The highest densities

FIG. 1. (a) Schematic drawing of pressure gauge in stabilization cell. (b) Typical experimental run showing buildup and decay of pressure; for discussion, see text.

currently achieved are 10^{16} – 10^{17} atoms/cm³.^{2,3} The critical temperature for BEC in a noninteracting Bose gas is $T_c = 3.31\hbar^2(n_g/g)^{2/3}/mk_B$ where g is the spin degeneracy. For $g=1$ and $n_g=10^{17}$ atoms/cm³, $T_c=3.5$ mK. Densities one to two orders of magnitude higher will be required to observe BEC in the more easily accessible 50-mK range. Limitations on the buildup of density can be determined by studying the decay of the density. An isothermal study of the temporal decay of pressure provides the decay of the density which obeys the approximate rate equation⁹

$$\frac{dn_g}{dt} = -(n_g/\tau) - K_v n_g^3 - (A/V)K_s n_g^2 + \varphi/V. \quad (1)$$

Here the first term represents the loss due to thermal escape of atoms out of an open ended cell with time constant^{2,3,10} τ ; the second and third are volume and surface recombination loss, with rate constants K_v and K_s , respectively. n_s is the surface density and φ is the incoming flux. We assume $N = N_g + N_s \approx N_g$. A careful study of the pressure decay will provide the rate constants and allow an estimate of the maximum attainable densities. Recently two of us⁹ have shown that the

density of D_4 which also obeys Eq. (1) is limited by surface recombination. For low densities the surface adsorption isotherm^{11,12} is $n_s = n_g \lambda \exp(\epsilon_a/k_B T)$, where $\lambda = (2\pi\hbar^2/mk_B T)^{1/2}$ is the thermal de Broglie wavelength. After substituting this n_s into Eq. (1), the third term becomes $K_s^{\text{eff}} n_g^2$, with $K_s^{\text{eff}} \equiv K_s(A/V)\lambda^2 \exp(2\epsilon_a/k_B T)$.¹³ A measurement of K_s^{eff} as a function of temperature was used to determine ϵ_a for D_4 on ^4He . The density limiting processes in H_4 could not be identified in a similar manner, since the bolometric detection technique, used earlier in Ref. 9, is not well suited to study the slow decay processes observed in this system. The development of the capacitance pressure gauge has allowed such a study. By choosing A/V large, T sufficiently small, and B sufficiently high, the volume recombination term can be rendered negligible at moderate densities.

A typical pressure fill-decay plot is shown in Fig. 1(b). The cell is filled for a period of 5–10 min with H_4 starting at point A. At point B the discharge is turned off and the cell cools to point C where it is then stabilized in temperature to within 1 mK. The pressure decay is then observed for periods up to 1 h to point D where the remaining H_4 is destroyed with the bolometer. The data are sampled with a Nicolet-1174 transient recorder and transmitted to a PDP/11 computer. It is fitted to a combined first- and second-order decay process using a nonlinear least-squares fitting procedure to a function of the form

$$p(t) = \frac{p(0) \exp(C_1 t)}{1 + [1 - \exp(C_1 t)] p(0) C_2 / C_1},$$

where C_1 and C_2 are the first- and second-order rate constants. With use of $p = n_g k_B T$, the density decay constants $1/\tau$ and K_s^{eff} are calculated. No evidence was found for the presence of a significant third-order term. Separate measurements were made to establish that $p \propto N$, by using the bolometric technique to measure N , and that $p \propto T$. The latter relationship was less accurately established, because of uncertainties in the temperature dependence of the density in our system.¹⁰ The constants were determined as a function of temperature for $0.34 < T < 0.54$ K at $B = 7.9$ T and magnetic field for $5 < B < 10$ T at $T = 0.385$ K.

To determine ϵ_a for H_4 on ^4He we plot $K_s^{\text{eff}} T^{1/2}$ vs $1/T$ as shown in Fig. 2(a). The factor $T^{1/2}$ arises from the T^{-1} dependence of λ^2 and the $T^{1/2}$ dependence expected for K_s .⁹ This has been established by Morrow *et al.*,⁸ who measured ϵ_a by two independent techniques: decay of the density,

FIG. 2. (a) Logarithmic plot of $K_s^{\text{eff}} T^{1/2}$ vs $1/T$. The adsorption energy is determined from the slope. (b) Field dependence of the rate of surface recombination plotted as $(K_s^{\text{eff}})^{-1}$ vs B^2 for $T = 0.385$ K.

as is done here, and shift of the zero-field hyperfine resonance transition as a function of temperature. From the slope of the plot in Fig. 2(a), we find $\epsilon_a/k_B = 0.89 \pm 0.07$ K. Morrow *et al.* found 0.95 ± 0.02 K by the decay technique and 0.92 ± 0.04 K from the hyperfine shift. This may be compared with D_1 on ^4He which has a value of $\epsilon_a/k_B = 2.6 \pm 0.4$ K when we use the $T^{1/2}$ temperature dependence of K_s of Ref. 9.

We have also determined the magnetic field dependence of the surface rate: $K_s(B) = \gamma(B)K_s'$. A simple model for this dependence proposes $\gamma \sim 1/B^2$.^{14,15} The stabilized H_1 consists of atoms in two hyperfine states $|a\rangle \approx -|\uparrow\downarrow\rangle + \epsilon|\uparrow\uparrow\rangle$ and $|b\rangle = |\uparrow\downarrow\rangle$ ($\uparrow$, electron spin; $\uparrow$, proton spin), where $\epsilon \approx a/4\mu_B B$ is the admixture of the electron spin-up state into the ground state (a is the zero-field hyperfine splitting). By assuming that only atomic collisions in which at least one atom is in the $|a\rangle$

state can lead to recombination, we¹⁴ find a probability $\gamma = \frac{1}{4}$ at zero field and $\gamma = \frac{1}{2}\epsilon^2$ for our present high-field regime. This corresponds to a reduction of the zero-field rate by $2\epsilon^2 = 1.3 \times 10^{-5}$ at 10 T. A confirmation of this model is found in Fig. 2(b), where we plot $(K_s^{\text{eff}})^{-1}$ vs B^2 . The dotted line relates to our best value for K_s : $K_s B^2 / T^{1/2} = (2.6 \pm 0.8) \times 10^{-7} \text{ cm}^2 \text{ T}^2 \text{ K}^{-1/2} \text{ sec}^{-1}$. Morrow *et al.* observed $K_s^{\text{eff}} = 4.5 \times 10^{-16} \text{ cm}^3/\text{sec}$ in zero field at $T = 0.5$ K. Comparison with our results may be made by scaling¹⁶ A/V and using $\gamma = \frac{1}{4}$. We calculate $K_s^{\text{eff}} = 1.2 \times 10^{-15} \text{ cm}^3/\text{sec}$, in reasonable agreement with the observed value.

These results have serious implications for observing BEC in current geometries. It has been shown^{11,12} that to attain BEC the surface must be saturated to a value $n_s^{\text{sat}} = \epsilon_a/2\alpha_2 = 6 \times 10^{13} \text{ cm}^{-2}$, where α_2 is taken from a linear expansion for the two-dimensional interaction energy in terms of the density. There are two conditions for stability: static and dynamic. Silvera and Goldman have shown¹¹ that coverages up to $1.8 \times 10^{14} \text{ cm}^{-2}$ will have static stability. The limitations which we have measured have again shown⁹ that only dynamic stability is important. For example, with use of the high-flux results of Cline *et al.*³ with $n_s = 7.5 \times 10^{16} \text{ cm}^{-3}$ obtained at 0.3 K, the adsorption isotherm yields $n_s = 4.6 \times 10^{11} \text{ cm}^{-2}$. This is much lower than the static stability value or n_s^{sat} .

More promising are some preliminary measurements on a 0.2% ^3He - ^4He mixture, where we observed a reduction of K_s^{eff} by a factor of 6.1. If we attribute this completely to a change in adsorption energy, we find $\epsilon_a/k_B \approx 0.59$ K. Unfortunately, in our temperature range we were unable to do a useful measurement with a saturated mixture because of fluctuations in the background pressure of ^3He which are not compensated by the superleak. A possibility exists that the surface of the mixture is saturated with ^3He and little additional reduction of ϵ_a will be found with increasing ^3He concentration, although a reduction of the bulk density will also reduce ϵ_a .

We also note that the presence of the first-order loss of atoms from our cell prevented us from establishing evidence for the interesting relaxation-limited recombination process proposed by Statt and Berlinsky,¹⁵ which may also allow achievement of higher surface densities in high fields.

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Lattice-Location Experiment of the Ni-Si Interface by Thin-Crystal Channeling of Helium Ions

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A thin-crystal channeling experiment has been devised to locate preferential sites of monolayers of Ni atoms deposited onto Si crystalline substrates. The Ni scattering yield shows dips or peaks in the angular scan measurements when channeled along the major crystallographic axes and planes of Si. The results can be explained by a dispersed Ni-Si interface with 2.1×10^{15} Ni atoms/cm² occupying the tetrahedral interstitial voids of Si.

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Numerous techniques such as low-energy electron diffraction, Auger-electron spectroscopy, X-ray photoemission (XPS), ultraviolet photoelectron spectroscopy, and ion channeling have been applied to the study of metal-semiconductor interfaces.¹⁻⁶ Recently, such studies have revealed that interfaces between metals and semiconductors such as Si, GaSb, InP, and GaAs are chemically reactive even at room temperatures and that the interfaces are far from being abrupt as modeled by various theoretical studies. For the Ni-Si system, the existence of an intermixed interface between as-deposited Ni atoms and the crystalline substrate has been observed by ion channeling for clean Si surfaces prepared under ultrahigh-vacuum conditions as well as chemical-

ly cleaned Si surfaces.⁶ With a coverage of 1×10^{16} Ni atoms/cm², approximately 6×10^{15} Si atoms/cm² are nonregistered with respect to the crystalline (111) substrate. An XPS study has also confirmed the room-temperature reactivity between Ni and Si and the Ni atoms were found to be in a Si-rich environment.^{5,7}

In the present study, a thin-crystal channeling experiment has been devised to locate the lattice sites of monolayers of Ni atoms deposited onto Si crystalline substrates. Ion-beam channeling has been applied to locate preferential sites of impurities in bulk crystals.⁸ By making use of the "flux-peaking" effect of ion channeling, the relative position of the impurity atoms with respect to the host's crystalline matrix can be determined