Measurement of Pressure of Gaseous \( \text{H}_2 \): 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 \( K_5 \) and the adsorption energy \( \epsilon_a \) of \( \text{H}_2 \) on \(^4\text{He}\) are \( K_5 B^2 / T^{1/2} = (2.6 \pm 0.8) \times 10^{-7} \text{ cm}^2 \text{ T}^2 \text{ K}^{-1/2} \text{ sec}^{-1} \) 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 \( \text{H}_1 \) on a dilute \(^4\text{He} - \text{He} \) 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 \(^4\text{He}\).\(^{1,2}\) 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 \( \text{H}_1 \). \( \text{H}_1 \) is predicted to undergo Bose-Einstein condensation (BEC) if sufficiently high densities can be achieved.\(^6\) However, processes which limit the highest densities\(^{3,5}\) of \( \text{H}_1 \) have not been identified. In this Letter, we provide the first experimental proof that magnetic fields stabilize \( \text{H}_1 \) against recombination to \( \text{H}_2 \).

We show surface recombination to be the dominant recombination process in our system. We also measure the adsorption energy of \( \text{H}_1 \) on \(^4\text{He}\) finding \( \epsilon_a / k_B = 0.89 \pm 0.07 \text{ K} \). This is consistent with the theoretical estimate of Mantz and Edwards\(^7\) and with very recent zero-field measurements of Morrow et al.\(^8\)

The large value of \( \epsilon_a \) combined with surface recombination rates create a serious barrier against achieving conditions for BEC. Measurements with a very dilute \(^3\text{He} - \text{He}\) surface coverage yield a significant and promising reduction of \( \epsilon_a \). Although \( \text{H}_1 \) is predicted to be the only elemental material that remains a gas\(^6\) at \( T = 0 \text{ K} \), there have only been indirect determinations of its state. We have directly measured the pressure, \( p \), of \( \text{H}_1 \) using a sensitive capacitance manometer providing the most direct evidence that \( \text{H}_1 \) is a gas for \( T \geq 0.27 \text{ K} \), our lowest accessible temperature. For densities studied, \( n_e \sim 1 \times 10^{16} \text{ atoms/cm}^3 \), we have shown that \( \text{H}_1 \) behaves as a perfect gas, i.e., \( p = n_e k_B T \).

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 \(^4\text{He}\). The cell is in the center of a solenoidal magnetic field of maximum value 11 T which serves to polarization the hydrogen and confine it in the cell by magnetic compression. The \( \text{H}_1 \) is further confined by a helium vapor compressor (HEVAC) located in the fill tube. In earlier experiments the \( \text{H}_1 \) 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 \( \text{H}_1 \) 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-\( \mu \text{m} \)-thick gold-plated Kapton sheet attached with Stycast-1286 epoxy. The capacitance was measured with an ac bridge and calibrated at low temperature against a Barocell pressure gauge with use of \(^4\text{He} \) gas. The manometer would normally measure the pressure due to \( \text{H}_2 \) and \(^4\text{He} \) in the chamber. A superleak connecting the vacuum side of the membrane to the \( \text{H}_1 \) side equalized the \(^4\text{He} \) pressure, so only the \( \text{H}_1 \) pressure was measured. This could also serve as a detector of superfluidity in \( \text{H}_1 \) 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 \( \text{H}_1 \) 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 \( \text{H}_1 \) 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
The density of \( D_1 \) which also obeys Eq. (1) is limited by surface recombination. For low densities the surface adsorption isotherm\(^{11,12} \) is \( n_z = n_z \lambda \exp(\epsilon_z/k_B T) \), where \( \lambda = (2\pi \hbar^2/m k_B T)^{1/2} \) is the thermal de Broglie wavelength. After substituting this \( n_z \) into Eq. (1), the third term becomes \( K_z \epsilon_z^2 \), with \( K_z = K_z(A/V) \exp(2\epsilon_z/k_B T) \). A measurement of \( K_z \) as a function of temperature was used to determine \( \epsilon_z \) for \( D_1 \) on \(^4\)He. The density limiting processes in \( H_1 \) 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 \( H_1 \) 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 \( H_1 \) 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

\[
\rho(t) = \frac{\rho(0) \exp(C_C t)}{1 + [1 - \exp(C_C t)] \rho(0) C_C / C_1}
\]

where \( C_1 \) and \( C_2 \) are the first- and second-order rate constants. With use of \( \rho = n_z k_B T \), the density decay constants \( 1/\tau \) and \( K_z \) are calculated. No evidence was found for the presence of a significant third-order term. Separate measurements were made to establish that \( \rho \propto N \), by using the bolometric technique to measure \( N \), and that \( \rho \propto T \). 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 \( \epsilon_z \) for \( H_1 \) on \(^4\)He we plot \( K_z \epsilon_z^2 T^{1/2} \) vs \( 1/T \) as shown in Fig. 2(a). The factor \( T^{1/2} \) arises from the \( T^{-1} \) dependence of \( \lambda^2 \) and the \( T^{1/2} \) dependence expected for \( K_z \). This has been established by Morrow et al.,\(^8 \) who measured \( \epsilon_z \) by two independent techniques: decay of the density,
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 \pm 0.02 K by the decay technique and 0.92 \pm 0.04 K from the hyperfine shift. This may be compared with \( D_1 \) on \(^3\)He 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: \( K_s(B) = \gamma(B)K_s(0) \). A simple model for this dependence proposes \( \gamma = 1/B^2 \). \(^{14,15}\) The stabilized \( H_1 \) consists of atoms in two hyperfine states \( |a\rangle = - |4\uparrow\rangle + \epsilon |\downarrow\rangle \) and \( |b\rangle = |\downarrow\rangle \) (\( \uparrow \), electron spin; \( \downarrow \), proton spin), where \( \epsilon = \alpha/4\mu_B B \) 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, we\(^{14} \) find a probability \( \gamma = \frac{1}{4} \) at zero field and \( \gamma = \frac{2}{8} \) 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 \( (K_s)^{-1} \) vs \( B \). The dotted line relates to our best value for \( K_s: K_s B^2/T^{1/2} = (2.6 \pm 0.8) \times 10^{-7} \) cm\(^2\) K\(^{-1/2}\) sec\(^{-1}\). Morrow et al. observed \( K_s(0) = 4.5 \times 10^{-16} \) cm\(^3\) sec\(^{-1}\) in zero field at \( T = 0.5 \) K. Comparison with our results may be made by scaling \( A/V \) and using \( \gamma = \frac{1}{4} \).

We calculate \( K_s(0) = 1.2 \times 10^{-15} \) cm\(^3\) sec\(^{-1}\), 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 \( n_s^{sat} = \epsilon_s/2a_s = 6 \times 10^{13} \) cm\(^{-2}\), where \( a_s \) 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 shown\(^ {14} \) that coverages up to \( 1.8 \times 10^{14} \) cm\(^{-2}\) will have static stability. The limitations which we have measured have again shown\(^9 \) that only dynamic stability is important. For example, with high flux results of Cline et al.\(^3 \) with \( n_s = 7.5 \times 10^{15} \) cm\(^{-3}\) obtained at 0.3 K, the adsorption isotherm yields \( n_s = 4.6 \times 10^{11} \) 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\% \(^3\)He-\(^4\)He mixture, where we observed a reduction of \( K_s \) by a factor of 6.1. If we attribute this completely to a change in adsorption energy, we find \( \epsilon_s/k_B = 0.59 \) 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 \(^3\)He which are not compensated by the superleak. A possibility exists that the surface of the mixture is saturated with \(^3\)He and little additional reduction of \( \epsilon_s \) will be found with increasing \(^3\)He concentration, although a reduction of the bulk density will also reduce \( \epsilon_s \).

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. We acknowledge useful discussions with V. V. Goldman and R. Sprink. We thank W. S. Truscott for his advice in designing the pressure gauge,
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 $2.1 \times 10^{15}$ Ni atoms/cm$^2$ 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. 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. With a coverage of $1 \times 10^{16}$ Ni atoms/cm$^2$, approximately $6 \times 10^{15}$ Si atoms/cm$^2$ 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.

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. 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.