Spin-Polarized Atomic Deuterium: Stabilization, Limitations on Density, and Adsorption Energy on Helium

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Current limitations on the density of stabilized spin-polarized deuterium $(D_{\downarrow})$ to $\sim 10^{14}$ atoms/cm^3 are shown to arise from condensation on the ^{4}He surface and subsequent recombination. A new technique for measuring the adsorption energy is employed to provide the first such measurement for $D_{\downarrow}$ on helium, yielding $\epsilon_a/k_B = 2.5 \pm 0.4$ K.

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We have recently reported the stabilization of a gas of spin-polarized atomic hydrogen, H$_{\downarrow}$, in a cell in which all surfaces were coated with superfluid ^{4}He.[1] Although densities greater than 10^{16} atoms/cm^3 have already been achieved for H$_{\downarrow}$,[2] attempts to achieve higher densities for spin-polarized atomic deuterium, D$_{\downarrow}$, have yielded densities $n \lesssim 10^{14}$ atoms/cm^3. In this Letter we show that the limitation is due to a fundamental problem: the adsorption of D$_{\downarrow}$ on the ^{4}He surface and subsequent recombination to D$_2$. The same process can ultimately prevent achievement of the higher densities necessary for Bose-Einstein condensation in H$_{\downarrow}$. We have developed a method for measuring the adsorption energy and find $\epsilon_a/k_B = 2.5 \pm 0.4$ K for D$_{\downarrow}$ on ^{4}He. We have also attempted to stabilize H$_{\downarrow}$ and D$_{\downarrow}$ using pure ^{3}He surfaces and detected no measurable density. However, a mixture of ^{3}He-^{4}He, which is known to phase separate and to provide a ^{3}He surface, is effective.

D$_{\downarrow}$ is a spin-$\frac{1}{2}$ or -$\frac{3}{2}$ particle and is expected to behave as a Fermi gas.[3] For temperatures lower than the Fermi temperature, $T_F = \hbar^2/2mk_B \times (6\pi^2 n/g)^{2/3}$ (g is the nuclear spin degeneracy) it becomes a degenerate quantum gas. At even lower temperatures it is conjectured to display superfluidity with several phases due to the various possible pairings of the fermion spin,[4] in some analogy with ^{3}He. Densities of order 10^{20} atoms/cm^3 are required to observe these properties with the existing low-temperature technology.

We briefly describe our experimental technique, which is similar to that of Ref. 1. Atomic D originating in a room-temperature microwave discharge enters our cryostat via a warm Teflon tube and is abruptly cooled by an accommodator. This is a short length of cylindrical tubing cooled to $T \simeq 5$ K by a weak thermal contact to a ^{4}He bath[5] and covered with solid D$_2$. The gas flows into a cold ($T \gtrsim 270$ mK) stabilization cell, which we call the HSC (hydrogen stabilization cell), sitting in a solenoidal magnetic field which has a value of 8.0 T in the present experiments. This field polarizes and confines the atoms to the HSC.[2,6] Confinement is further aided by a mixture helium vapor compressor (HEVAC) located between the accommodator and the HSC. Before loading with D$_{\downarrow}$, the HSC and HEVAC walls are covered with a film of ^{4}He which extends almost to the accommodator. The density of H$_{\downarrow}$ confined in such a cell decays exponentially, as a result of thermal leakage,[2,6] with a time constant $\tau \sim 2 \times 10^3$ sec for the current conditions; we expect about the same value for D$_{\downarrow}$. The number of D$_{\downarrow}$ atoms in the cell are measured by precipitating rapid recombination on the surface of a bolometer and measuring the subsequent temperature rise of the HSC; calibration is done electronically.[1,2]

We have found the density of D$_{\downarrow}$ to be limited for two reasons. First, in the presence of a decay process, the maximum density in the HSC depends on the loading flux of atoms. The flux of D$_{\downarrow}$ was substantially lower than that of H$_{\downarrow}$. This was found to be due to condensation and recombination on the surfaces of the accommodator region. By heating the accommodator up by several degrees Kelvin, the D$_{\downarrow}$ flux could be increased by a factor 5–10. This implies a larger adsorption energy of D on D$_2$ than H on H$_2$.[7] A similar study for H$_{\downarrow}$ yielded an increased flux of 2–3. However, even with the increased flux of D$_{\downarrow}$ the maximum density was limited to $\sim 10^{14}$ atoms/cm cm^3, at least two orders of magnitude lower than what we have thus far achieved for H$_{\downarrow}$.

A second and more fundamental limitation was found by studying the temporal decay of D$_{\downarrow}$ samples. We have measured the decay rate of the number of atoms in the HSC, $N = N_g + N_s = Vn_g + An_s$, where N_g is the number in the gas phase and N_s in the surface phase, V is the volume, and A is the area available to the D$_{\downarrow}$. A theoretical

expression for the decay of atoms is found from the solution of the rate equation

$$-dN/dt = -V\,dn_g/dt - A\,dn_s/dt$$
$$= Vn_g/\tau + VK_v n_g^3 + AK_s n_s^2, \qquad (1)$$

where K_v and K_s are the volume and surface recombination rate constants. For the measured decay times of ~ 10 sec the first term is negligible; we also expect the second term of (1) to be unimportant, although K_v has never been measured. Assuming that the surface coverage and gas density remain in thermodynamic equilibrium during decay, we can use the low-density–high-temperature form of the adsorption isotherm for a boson or fermion gas,[8] $n_s = n_g \lambda \exp(\epsilon_a/k_B T)$, where ϵ_a is the adsorption energy and $\lambda = (2\pi\hbar^2/mk_B T)^{1/2}$ is the thermal de Broglie wavelength. Substituting this in Eq. (1) and using the fact that $N = N_g + N_s \simeq N_g$, we find

$$-dN/dt$$
$$= [K_s A V^{-2}\lambda^2 \exp(2\epsilon_a/k_B T)]N^2 \equiv K_s^{\text{eff}} N^2, \qquad (2)$$

which has the solution $N^{-1} - N_0^{-1} = K_s^{\text{eff}}(t - t_0)$. The decay of N was studied by filling the HSC to a known level, waiting for a time t, and then measuring the remaining number of atoms. The results for two different temperatures are shown in Fig. 1. The data have been corrected for a small hydrogen impurity that is present in our D_2 sample and arises from water that gets into the discharge and is dissociated, but could be minimized by judicious cold trapping. The $H_\downarrow$ impurity is long lived and thus the sample in the HSC decays rapidly to the $H_\downarrow$ level which ranged from ~ (0–20)%. The data could be fitted with a second-order process after correcting for the H impurity; a possible third-order term was zero to within experimental error. We conclude that the gas-phase density of $D_\downarrow$ is limited by adsorption and recombination on the ^{4}He surface. Uang and Stwalley[9] have recently suggested that a $D_\downarrow$ impurity in $H_\downarrow$ (the argument also applies for $H_\downarrow$ in $D_\downarrow$) has a giant spin-flip cross section relative to $H_\downarrow$-$H_\downarrow$ cross sections and would be destructive, leading to rapid recombination. Our measurements show that the two gases easily coexist at the densities studied here.

From these studies we have been able to make the first determination of the adsorption energy of a hydrogen isotope on ^{4}He. The slopes of the decay curves such as given in Fig. 1 yield K_s^{eff}, which we determined for several temperatures. The intrinsic rate constant K_s is expected to vary

as $T^{1/2}$ at high temperature, coming to a nonzero value at $T = 0$. For our temperature regime we treat K_s to be independent of temperature. The slope of a plot of $\ln(TK_s^{\text{eff}})$ vs $1/T$, as shown in Fig. 2, yields $2\epsilon_a$ and we find $\epsilon_a/k_B = 2.5 \pm 0.4$ K. A $T^{1/2}$ dependence of K_s would increase the value by 4%. This value is substantially larger than the 1.11 K calculated by Guyer and Miller[10] or the 1.39 K due to Mantz and Edwards.[11]

The ^{4}He used for surface coverage was taken from an ultrahigh-purity lecture bottle. The ^{3}He impurity was unknown. We varied the quantity condensed in the cell by about a factor 4 so that the film was probably undersaturated. Since ^{3}He impurities would reside on the surface, this should vary a ^{3}He impurity effect. Any differences due to film thickness and impurity were within the noise level of our measurements.

Let us consider some of the implications of these results. First, using the adsorption isotherm and $n_g = 1 \times 10^{13}$ atoms/cm^3 sec at $T = 0.4$ K,
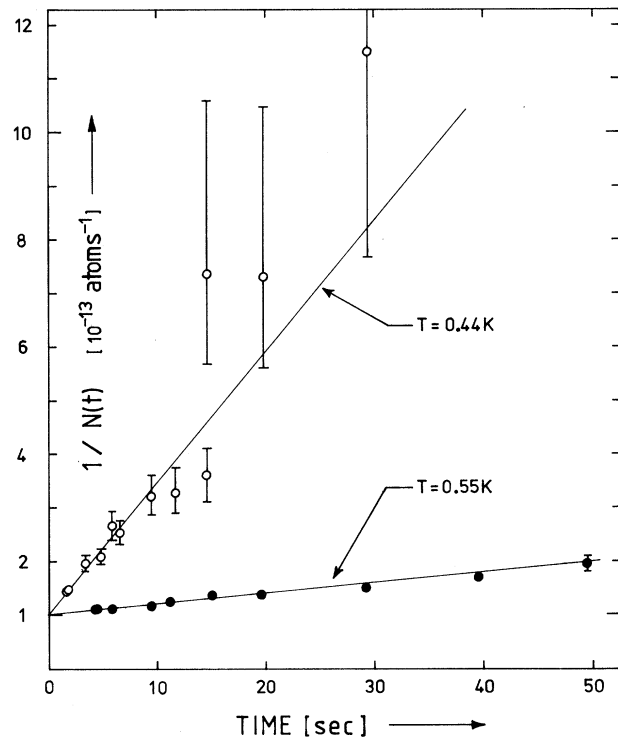

FIG. 1. Decay of the total $D_\downarrow$ particle number N in the HSC as a function of time (t) plotted as $1/N(t)$ vs t. All data are normalized to an initial value of $N = 1 \times 10^{13}$ atoms/cm^3. Open circles $T = 0.44$ K; closed circles $T = 0.55$ K. The lines represent a least-squares fit to the data for which a pure second-order decay process is assumed.
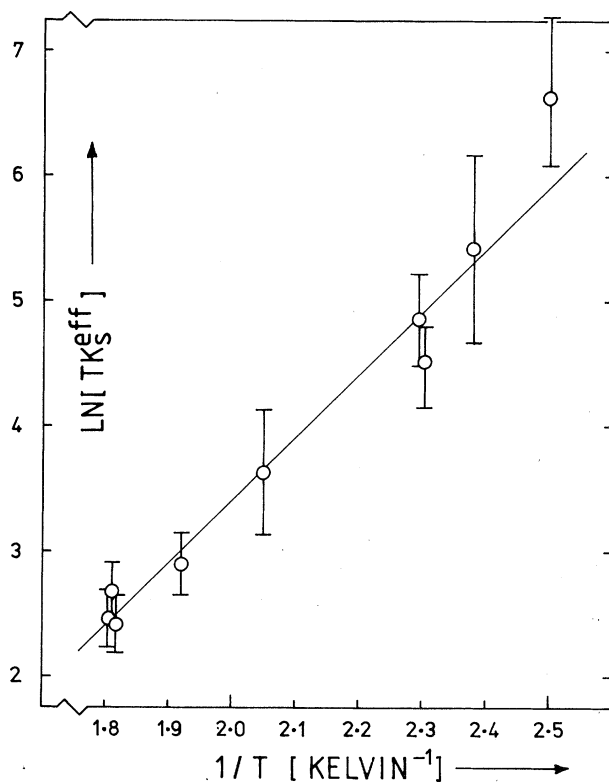
FIG. 2. Logarithmic plot of TK_s^{eff} vs $1/T$ allowing determination of the adsorption energy with application of a weighted—least-squares-fit procedure to a straight line. The value of the logarithm is given to within an additive constant.

we find $n_s = 1 \times 10^9$ atoms/cm^2. This is a much lower limiting coverage than has been anticipated. Silvera and Goldman[8] have shown that at 8 T the critical coverage for static stability is 1.5 $\times 10^{14}$ atoms/cm^2 for H$_\downarrow$ and we expect about the same value for D$_\downarrow$. The present results show that this condition for metastability is of much less importance than the rates for dynamical three-body decay from the metastable spin-polarized state to the molecular state. Second, since at present there is no adequate theory for the dynamical decay processes, we cannot directly assess the implications for H$_\downarrow$. We would not expect K_s to be the same for H$_\downarrow$ and D$_\downarrow$, because the molecules have different energy levels.[12] We can make a rough check of this. Calculated values of ϵ_a/k_B for H$_\downarrow$ on ^{4}He are 0.1 K (Ref. 10) and 0.6 K.[11] If we take a value of 1 K and assume K_s to be the same for both H$_\downarrow$ and D$_\downarrow$, we estimate $K_s^{eff} \simeq 3.4 \times 10^{-16}$/sec at $T = 0.37$ K and a half-life of 30 sec for an initial value $N_g \simeq 10^{14}$ atoms. However, under these conditions we did not ob-

serve experimentally[2] any significant deviation from an exponential decay with time constant $\tau \simeq 2000$ sec, indicating that for H$_\downarrow$ the values for K_s and/or ϵ_a should be considerably smaller. This result is only to be taken as an exercise to show that limitations on H$_\downarrow$ may be quite different. We note that to achieve Bose-Einstein condensation in H$_\downarrow$, a coverage $n_s \simeq 10^{13}$–10^{14} atoms/cm^2 must be contended with.[8] A *rough* estimate of the maximum coverage of H$_\downarrow$ on helium can be made as follows. The zero-field value of K_v for H-H-He collisions is 2.8×10^{-31} cm^6/sec.[13] We estimate (see last referral, Ref. 1) this to be reduced by a factor $\eta \simeq 8 \times 10^{-6}$ in a field of 10 T. From the rate equation, $dn/dt = - (\eta K_v n_{He}) n^2 + \varphi$ (which resembles a surface rate equation) for a flux $\varphi = 10^{16}$ cm^3/sec, we find a maximum density 1.7×10^{17} atoms/cm^3 (with $n_{He} = n$). Since final states in the recombination process on a surface are probably high energy and free particlelike, we treat the liquid helium as a dense gas of surface density $(10^{22})^{2/3}$ atoms/cm^2. Arguing that the dominant consideration of volume or surface recombination is the interparticle distance, we can scale the maximum volume density to find a maximum surface coverage of H$_\downarrow$ to be $\sim 5 \times 10^9$ atoms/cm^2. We note that for densities of order 10^{16} atoms/cm^3 nonexponential-decay limitations were observed.[2] Assuming $\epsilon_a/k_B = 1$ K, we calculate $n_s = 3 \times 10^{10}$ atoms/cm^2. As a third point, we note that we have a large amount of copper sinter in our HSC which provides a large value for $A \sim 400$ cm^2 in the expression for K_s^{eff}. This can be reduced by 50–100 cm^2 to enhance lifetimes and densities.

To relieve the surface problem it may be necessary to improve upon the use of ^{4}He as a surface. The adsorption energy is very sensitive to the surface density profile of the helium. ^{3}He is expected to have a more extended profile than ^{4}He and thus a lower H$_\downarrow$ or D$_\downarrow$ adsorption energy. We have lined our HSC with pure ^{3}He and found no measurable density of H$_\downarrow$ or D$_\downarrow$. We estimate our detection limit to be 10^8–10^9 atoms/cm^3. Evidently the problem is that the tubing between the accommodator and the HEVAC is not covered or protected and the H recombines in this region, never reaching the HSC. The superfluid properties of ^{4}He appear to be necessary. We have also used a ^{3}He-^{4}He mixture. This we found to be an effective coverage for stabilization. The components phase separate with the dilute-superfluid phase covering the vital surfaces. ^{3}He is expected to reside on top of the ^{4}He-rich phase to pres-

ent a ^{3}He surface to the H$_\downarrow$. At our current experimental temperature range the thermal response of the HSC is sluggish compared with pure ^{4}He because of the substantially higher vapor pressure. Lower temperatures will relieve this problem.

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[1]I. F. Silvera and J. T. M. Walraven, Phys. Rev. Lett. 44, 164 (1980), and J. Phys. (Paris), Colloq. 41, C7-137 (1980), and in Proceedings of the European Physical Society Conference on Condensed Matter, Antwerpen, 1980 (to be published).

[2]J. T. M. Walraven, I. F. Silvera, and A. P. M. Matthey, Phys. Rev. Lett. 45, 449 (1980); R. Cline, T. J. Greytak, D. Kleppner, and D. A. Smith, J. Phys. (Paris), Colloq. 41, C7-151 (1980).

[3]J. H. Freed, J. Chem. Phys. 72, 1414 (1980).

[4]A. J. Leggett, J. Phys. (Paris), Colloq. 41, C7-19 (1980).

[5]I. F. Silvera and J. T. M. Walraven, Phys. Lett. 74A, 193 (1979).

[6]J. T. M. Walraven and I. F. Silvera, Phys. Rev. Lett. 44, 168 (1980), and J. Phys. (Paris), Colloq. 41, C7-147 (1980).

[7]The measured adsorption energy of H on H$_2$ is 38 ± 8 K, S. B. Crampton, J. Phys. (Paris), Colloq. 41, C7-249 (1980).

[8]I. F. Silvera and V. V. Goldman, Phys. Rev. Lett. 45, 915 (1980).

[9]Y. H. Uang and W. C. Stwalley, Phys. Rev. Lett. 45, 627 (1980).

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

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

[12]W. C. Stwalley, Phys. Rev. Lett. 37, 1628 (1976).

[13]W. N. Hardy, M. Morrow, R. Jochemsen, B. W. Statt, P. R. Kubik, R. M. Marsolais, A. J. Berlinsky, and A. Landesman, Phys. Rev. Lett. 45, 453 (1980).

Origin of Low-Temperature Tunneling States in Glasses

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The low-temperature tunneling levels observed in glasses are discussed in terms of the free-volume model. As the system falls out of thermodynamic equilibrium near its glass transition temperature T_g, liquid clusters are frozen in. This Letter proposes that voids are formed within the liquid clusters as they in turn freeze at lower temperatures. Approximately 10^{-4} such centers form per atom, 1% of which contribute to the heat capacity below 1 K.

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Anderson, Halperin, and Varma[1] and Phillips[2] proposed to explain the linear temperature dependence of the specific heats of glasses at low-temperature via tunneling centers. They assumed that in any glassy system a certain number of atoms, or groups of atoms, has accessible two nearly equivalent equilibrium configurations corresponding to the minima of asymmetric double-well potentials and tunnels between them. The model explains many other experimental observations.[3-6] Nevertheless, there is no successful microscopic description of a tunneling center.

Instead, tunneling is treated via a model Hamiltonian for two-level systems representing the ground states in the two local energy wells. Here we explore the origin of the tunneling states.

The tunneling states are commonly associated with a small group of atoms undergoing a local rearrangement.[6] The number of atoms involved is assumed to be reasonably small to minimize the distance between states in configuration space. However, the larger the number of atoms, the easier it is to find two ground states of roughly equivalent energies. It is believed that this com-