Experiments with Atomic Hydrogen in a Magnetic Trapping Field

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We describe the loading of a minimum-*B*-field trap with neutral atomic hydrogen cooled through thermal exchange with liquid-helium surfaces. We monitor the gas during filling and decay by observing the atoms which are ejected from the trap as a result of spin exchange and magnetic dipolar relaxation. The stability of the gas is unaffected if we change the wall coverage from pure ⁴He to dilute ³He and ⁴He mixtures. The maximum trapped density is 3×10^{14} cm⁻³ at about 100 mK. The limiting decay rate is attributed to dipolar relaxation and found to agree with theory for both magnitude and field dependence.

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Surface-free confinement of neutral gases is opening up exciting new dimensions to condensed-matter and atomic physics. For atomic hydrogen, trapping concepts are particularly challenging because of the potential of H gas to undergo a transition to a Bose-Einstein condensed (BEC) state. Although liquid-helium surfaces provide the weakest conceivable attraction to the H atom and have thus enabled a wealth of interesting experiments,^{1,2} it has been observed $^{3-6}$ that the achievement of BEC would require the rigorous suppression of surface recombination. Once this was recognized, proposals were made to eliminate the influence of surfaces altogether by use of static⁷ or dynamic⁸ trapping technique. The behavior and manipulation of trapped atomic hydrogen (and deuterium) have been discussed by various authors,⁹⁻¹³ for the reason that trapping techniques may open the route to submillikelvin temperatures and to density-to-temperature ratios $n^{2/3}/T \approx 6.3 \times 10^{13}$ cm⁻² K^{-1} as required for BEC. Unfortunately, the Maxwell equations do not allow the construction of static traps for atoms in high-field-seeking spin states (a and b, referred to as $H\downarrow$) but only for the less stable low-field seekers (states c and d, denoted by H^{\uparrow}).¹⁴ Here a, b, c, and d are the hyperfine states of H in its electronic ground state in order of growing energy.

The first observation of trapped neutral atoms was made by Migdall *et al.*¹⁵ using a laser-cooled beam of Na atoms in combination with an anti-Helmholtz magnetic quadrupole trap. Recently, Hess *et al.*¹⁶ succeeded in trapping up to 5×10^{12} H[↑] atoms in a magnetic field minimum. The operating densities were near 10^{13} cm³ at temperatures of about 40 mK, corresponding to $n^{2/3}/T \approx 10^{10}$ cm⁻² K⁻¹. A mixture of all four hyperfine states of H was blown into the trapping region with a single pulse from the dissociator. The field gradients led to rapid spatial separation of H[↑] and H[↓]. In the rarified regime where the escape time of energetic H[↑] atoms was shorter than the interatomic collision time, Hess *et al.* could demonstrate "evaporative" cooling of the gas to below the wall temperature. The gas was studied destructively by our dumping into a magnetic resonance detector the atoms remaining in the trap after a certain holding time.

In this Letter we report on a novel approach to study H[†] in a magnetic confinement geometry. A bolometric detection method is employed which does not destroy the sample in the trap and enables us to arrange experiments much like conventional experiments with $H^{1,1,2}$ The trap is filled with H[†] until a steady state is reached between filling flux and the flux of $H\downarrow$ atoms escaping the trap due to spin-exchange and magnetic dipolar relaxation to the high-field-seeking states. As in the experiments of Greytak and co-workers, no direct thermometry is available for the gas. Our approach is to minimize temperature-related uncertainties by positioning walls and a high-field (4 T) barrier, avoiding the added complexity of evaporative cooling. The H-atom coverage on the surfaces is suppressed by the depth of the trap so much that all second-order surface processes, surface recombination in particular, become insignificant. At the same time the collision rate of the atoms with surfaces remains sufficiently large to enable thermalization with the environment. This method enables us to reach higher densities and to determine accurately the dipolar relaxation rate. We study filling of the trap and relaxational decay for temperatures ranging from 80 to 225 mK. We obtain a maximum density $n_0 = 3 \times 10^{14}$ cm⁻³ at the trap center at $T \approx 100$ mK, corresponding to a total of $N=4\times 10^{13}$ atoms or $n^{2/3}/T \approx 4.5\times 10^{10}$ cm⁻² K^{-1} . The stability of the sample is found to be the same for surfaces covered with either pure ⁴He or with ³He/⁴He mixtures, convincingly demonstrating the suppression of surface adsorption. By observing the $H\downarrow$ emerging from the trap, we determine the rate G_{dd} at which *d*-state atoms are converted into high-field seekers. This conversion-rate constant may be expressed in terms of the event rates for dipole-diple relaxation as defined in Ref. 9: $G_{dd} \equiv 2G_{ddaa}^d + G_{ddac}^d + G_{ddad}^d$. For T = 100 mK and B = 0.05 T, we find $G_{dd} = 1.1(2) \times 10^{-15}$ cm³/s, improving on the result of Hess et al.¹⁶ to be compared

with the theoretical value⁹ $G_{dd} = 1.2 \times 10^{-15}$ cm³/s. In extracting our results, we are sensitive to the field dependence of the relaxation channels.

Our trapping configuration is similar to that advocated by Pritchard¹⁷ and Hess.⁷ The field is generated by a system of eight superconducting coils operated in persistent mode. Four radially oriented racetrack-shaped coils provide a quadrupole field for radial confinement of the gas. At maximum current the quadrupole field is \approx 1.4 T at the surface of the sample cell (r=6.5 mm, see Fig. 1). Axial confinement is achieved with a couple of dipole coils positioned at z = +50 mm and z = -50mm near the ends of the racetracks. These coils produce fields of 1.7 and 1.5 T, respectively. The field strength at the B-field minimum was adjusted to B = 0.05 T by use of a trim coil at z=0. At the lower end of the racetracks, z = -131 mm, there is a 4.4-T cylindrical coil which serves to separate H^{\uparrow} and H^{\downarrow} . The well depth of the trap is defined by $\epsilon_{tr} = \mu_B \Delta B$, where μ_B is the Bohr magneton and ΔB is the difference between the lowest value of the field on any surface of the sample cell and the values at the field minimum. For our configuration $\epsilon_{\rm tr}/k_{\rm B} = 0.92 {\rm K}.$

All surfaces of the experimental cell shown in Fig. 1 are covered with liquid helium. A mixture of H \uparrow and H \downarrow is produced in the high-field zone (B=4 T) by rf discharge at low temperatures ($T \le 650$ mK) with the techniques pioneered by Hardy *et al.*¹⁸ Our dissociator is an all-metal $\frac{1}{4}\lambda$ helical cavity with $Q \approx 300$. It resonates at 718 MHz and is driven with 0.1-W×50- μ s pulse at a repetition rate of 50 Hz. The H \downarrow fraction, favoring the high field, is trapped in the dissociator region, where it is continuously removed by forced recombination on a helium-free bolometer surface (B#1). The



FIG. 1. Schematic drawing of the experimental cell.

low-field-seeking H[↑] flux ($\Phi^{\uparrow} \le 5 \times 10^{12}$ /s) is guided up through a thin-walled german silver (GS) tube (3.6-mm inner diameter) to a low-field region ($B \le 1.5$ T) bounded by the walls of a cylindrical copper cell, symmetrically surrounding the B-field minimum. Thermometry is done against a ³He melting-curve thermometer. We find an optimum H[†] filling flux at a dissociator temperature of 600 mK. No evaporative cooling takes place as the H[†] atoms cannot escape the low-field region and surface adsorption times are much shorter than the lifetime of the sample. We monitor the trapped gas continuously by observing the $H\downarrow$ flux escaping from the trap because of magnetic relaxation. For this purpose a "pumping plate" detector¹⁹ is mounted in high field (B = 4 T) at the annularly shaped lower end of the copper cell. We assume that the recombination heat is detected with an efficiency of $60\% \pm 10\%$.¹⁹ The minimum detectable flux is 2×10^{11} atom/s. A second bolometer (B#2) enables us to trigger recombination of the trapped H[†] and is included for calibration purposes.

We plot our data as $V_{\gamma}\dot{N}/N^2$ versus time t as shown in Fig. 2. N(t) is obtained by integration of the observed flux \dot{N} over the time interval (t,∞) . V_{γ} is the effective volume of the trap with the gas temperature T_g assumed to be equal to the wall temperature T_w , as defined below. To compare with theory, we calculate $\dot{N}(t)$ by averaging the field and temperature-dependent relaxation rates of Stoof, Koelman, and Verhaar⁹ over a thermal density distribution. The contribution due to any individual rate is written as

$$\dot{N} = (\gamma_0 G_0 / V_{\gamma}) N_{h1} N_{h2}, \tag{1}$$

where G_0 is the rate constant for the process under consideration, evaluated for the conditions at the center of the trap, and

$$\gamma_0 \equiv \int \{G(\mathbf{r})/G_0\} [n(\mathbf{r})/n_0]^2 d\mathbf{r}/V_{2e}$$

includes all effects due the field dependence of the rate. The effects associated with the density profiles are incor-



FIG. 2. Typical decay curve plotted as $V_{\gamma}\dot{N}/N^2$. ϕ is the H \uparrow filling flux; the dashed line is a simulation.

porated in the effective volume $V_{\gamma} \equiv V_{1e}^2/V_{2e}$, where $V_{me} \equiv \int [n(\mathbf{r})/n_0]^m d\mathbf{r}$. N_{hi} refers to the total number of atoms in the trap in hyperfine state hi. For our trap, $V_{\gamma}T_g^{-5/2} \approx 180 \text{ cm}^3 \text{ K}^{-5/2}$ and $V_{1e}T_g^{-5/2} \approx 34 \text{ cm}^3 \text{ K}^{-5/2}$. The field dependence gives rise to important corrections. When T_g is varied from 80 to 225 mK, theory predicts γ_0 to increase from 2.1 to 3.5 for the dipolar relaxation processes ($\gamma_0 \equiv \gamma_{dd}$), while γ_0 decreases for spin exchange ($\gamma_0 \equiv \gamma_{cc}$) from 0.28 to 0.08 over the same range. Notice that the temperature dependence of the γ factors reflects the field dependence of the rates as with increasing temperature the atoms sample higher fields in the trap. As spin exchange is very fast and only affects the c-state atoms $(cc \rightarrow aa, ac, bd)$, preferential escape of *c*-state atoms leads to a sample which consists almost exclusively of d-state atoms.⁹ This is observed as an enhanced initial decay (EID) during up to 8 s after the discharge is switched off (see dashed curve in Fig. 2 for a simulation of this effect). Below 100 mK, the EID is dominated by another effect which leads to a reduced initial decay (RID) for up to 15 s, which we attribute to incomplete thermalization to T_w . According to theory the asymptotic decay is described to within 3% by $V_{\gamma} \dot{N} / N^2 \equiv \gamma_{dd} G_{dd}.$

In Fig. 3 we plot the results for the overall secondorder decay rate $\gamma_0 G_0 \equiv V_\gamma \dot{N}/N^2$ vs T obtained by discarding the EID-RID period of 15 s. For ⁴He-covered surfaces, data were taken for 135 < T < 225 mK (open circles). Above 225 mK, systematic errors arise from recombination of H[↑] on the plate due to the far tail of the He[↑]-density distribution extending to high field. Below about 135 mK we had insufficient flux to fill the trap because of surface recombination in the filling tube. To reach lower temperatures, 1% ³He was added to the cell (filled circles in Fig. 3) which is known to reduce the adsorption energy from $\epsilon_a \approx 1$ K to $\epsilon_a \approx 0.4$ K and hence also the surface recombination.^{1,2} However, under these conditions heat conduction by ³He vapor makes the



FIG. 3. The second-order decay rate plotted vs temperature (open circles: ⁴He wall coverage; filled circles: ³He/⁴He coverage). Dashed line: Theoretical result for the bottom of the trap. Solid line: Including the field average. Dotted line: See text.

plate unreliable for T > 160 mK. Near 80 mK, we were limited by refrigerator power. The theoretical result for $\gamma_{dd}G_{dd}$ is given by the solid curve in Fig. 3. The dashed curve corresponds to G_{dd} for B=0.05 T and shows the small intrinsic temperature dependence of the dipolar rate. The good agreement with the solid curve demonstrates the importance of the γ_{dd} average and reflects the increase of the dipolar rate with growing field. After dividing our data by γ_{dd} , we find $G_{dd} = 1.1(2) \times 10^{-15}$ cm³/s for T = 100 mK and B = 0.05 T. In view of the strong temperature dependence of V_{γ} , we infer that the gas in the trap is thermalized to T_w . In principle, the deviation of the data with respect to the solid curve below 100 mK may be explained by our assuming T_g to be 8 mK higher than T_w as illustrated by the dotted curve in Fig. 3. We believe this effect not to be significant because of a 17% systematic error not included in the error bars of Fig. 3 and related to the uncertainty in absolute calibration of the detector. Comparing the open and filled circles, we note that our results are insensitive to the type of helium coverage. Hence the H[†]-surface density {scaling as $\exp[(\epsilon_a - \epsilon_{tr})/k_BT]$ } must be small, as is to be expected for $\epsilon_{\rm tr} \gtrsim \epsilon_a$.

To describe the filling of the trap we assume that initially, i.e., before entering the trap, the internal energy of the gas is driven by $U_i = \frac{3}{2} N k_B T_w + N U_p$, where the average potential energy $U_p \approx \epsilon_{\rm tr}$ is determined by the magnetic field at the site of the last sticking event. Once in the trapping region, most of the potential energy is converted irreversibly into kinetic energy within a few interatomic collision times, after which the gas may be characterized by a well-defined temperature T_g (not necessarily equal to T_w) and has an internal energy given by $U_f \approx 4Nk_BT_g$.²⁰ To fill the trap no volume work is done by the gas against the environment. In the adiabatic limit $(U_i = U_f)$ and starting with $T_w \ll \epsilon_{tr}/k_B$ the gas temperature should therefore increase to $T_g \approx \frac{1}{4} \epsilon_{\rm tr}/k_{\rm B}$, 250 mK for our trap. In practice, some heat will be carried off to the walls during the filling and by atoms that escape after relaxation. In addition, the inelastic nature of the relaxation process gives rise to some internal heating. The rate at which energy is carried off to the walls is given by

$$\dot{Q} = 4(N/\tau_w)k_B(T_g - T_w),$$
 (2)

where

$$\tau_w^{-1} = \frac{1}{8} a \overline{v} (A_{1e}/V_{1e}) \exp\left[-\epsilon_{\rm tr}/k_{\rm B}T_g\right]$$

is the gas-to-surface thermal accommodation rate, $\alpha/T_g \approx 0.5 \text{ K}^{-1}$ is the accommodation coefficient²¹ and $\bar{v} = (8k_BT_g/\pi m)^{1/2}$ the average atomic speed. The effective surface area $A_{1e}T_g^{-1/3} \approx 40 \text{ cm}^2 \text{ K}^{-1/3}$ is obtained by integration of the relative surface coverage over the walls of the sample cell. The exponential temperature dependence of τ_w arises from the ratio of density in the center of the trap to that near the wall and

varies from 1×10^5 at 80 mK to 60 at 225 mK. At 80 mK, τ_w is about 17 s. The interatomic collision rate τ_c^{-1} may be estimated from $\tau_c^{-1} \approx 2^{3/2} \pi a^2 \bar{v} (N/V_{\gamma})$, where a = 0.72 Å is the s-wave scattering length. For $n_0 = 10^{12}$ cm⁻³ and $T_g = 80$ mK, one calculates $\tau_c \approx 3$ s. For our experimental conditions, with central densities up to 3×10^{14} cm⁻³, the overall thermalization rate, $\tau_{th}^{-1} = 1/$ $(\tau_c + \tau_w)$, is either faster than the response time of our detector or limited by the surface accommodation rate. The observed RID's can be explained if we assume that just after switching off the discharge, the gas is at a slightly higher temperature ($\Delta T < 10$ mK) than the walls and cools down to T_w in 10-15 s. Even a small ΔT should show up as a substantial RID because of the strong temperature dependence of V_{γ} . The observed $\tau_{\rm th}$ is roughly in agreement with the above model, but the exponential character of its temperature dependence seems to be absent and if we compare with a computer simulation, the observed RID's are too large. This point deserves further study.

Our results show that even for our relatively high densities the temperature has to be reduced by 3 orders of magnitude to reach BEC. Extrapolating we estimate that our current technique is applicable down to approximately 55 mK. For cooling to the optical quantum limit (2 mK), we propose to use a Lyman- α optical cooling method. Since this leaves the number of particles unaffected, densities over 10^{14} cm⁻³ should be attainable at temperatures below 10 mK. As G_{dd} remains constant while τ_c diverges as $T^{-1/2}$, such initial conditions are vital to aim successfully for BEC with an evaporative cooling method.

During revision of the manuscript we received a copy of unpublished work by Masuhara *et al.*, reporting $T_g = 3 \text{ mK}$ and $n_0 = 7.6 \times 10^{12} \text{ cm}^{-3}$.²²

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FIG. 1. Schematic drawing of the experimental cell.