Magnetic Equation of State of a Gas of Spin-Polarized Atomic Hydrogen

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The magnetic equation of state of spin-polarized atomic hydrogen is reported, relating density to magnetic field ($B \lesssim 10.5$ T), temperature ($T \gtrsim 300$ mK), and atomic flux used to fill the sample cell. Densities greater than 10^{16} atoms/cm^3, more than two orders of magnitude greater than initial observations, have been achieved. Results are also presented on the buildup and decay of the density as a function of external conditions. Preliminary observations on the Fermi fluid, atomic deuterium, are reported.

Spin-polarized atomic hydrogen (H↓) has recently[1,2] been stabilized over periods of minutes at 270 mK and densities of order 10^{14} atoms/cm^3. In order to observe many of the fascinating predicted properties such as Bose-Einstein condensation and the gaseous nature of this many-body system at $T = 0$ K, it is necessary to achieve substantially higher densities. In this Letter we report density increases of more than two orders of magnitude over previous values; no fundamental limitations have yet been encountered. Experimental results are presented on the temporal behavior of H↓ during filling of a hydrogen stabilization cell (HSC) and the subsequent decay after filling. We have also studied the magnetic equation of state of H↓ which provides an important guide for efforts to increase the density. Finally we report some preliminary measurements on the stabilization of the Fermi fluid, spin-polarized atomic deuterium.

One of our principle objectives in this article is to thermodynamically demonstrate that, although exotic, H↓ behaves as a gas of magnetic moment bearing atoms that exists at very low temperatures. An ideal geometry for studying the thermodynamic properties would be an infinite space with no surfaces having a uniform temperature and including a region with a nonzero static magnetic field. The requirement that the chemical potential be uniform in such a system yields a relationship between the equilibrium local magnetization $\vec{M}$, the magnetic field $\vec{B}$, and the temperature T. This is the magnetic equation of state. Since $|\vec{M}| = \mu_B n$ (n is the local gas density and μ_B the Bohr magneton), establishment of the relationship between n, B, and T provides thermodynamic evidence of the gaseous nature of H↓.

In our actual system,[1] the gas is fed into the HSC from a room-temperature source with intermediate cooling states; it can exist in a nonequilibrium steady-state condition with a temperature extending from ~0.3 to 300 K. However, the gas is confined in the HSC by magnetic compression and a miniature low-temperature helium vapor compressor (HEVAC) and the density only suffers slow decay due to leakage or recombination. As a consequence we treat the gas as a quasiequilibrium system. For the density distribution we have[2]

$$n(B) = n(B_0) \exp[-\mu_B(B_0 - B)/kT], \qquad (1)$$

where B_0 is the maximum field. The ratio of the density at field B_0 to that at field B is called the magnetic compression factor, $c_M \equiv \exp[\mu_B(B_0 - B)/kT]$. The time constant τ is determined by the experimental design. In our system (described in Ref. 1)

$$\tau = 4c_H c_M V_{\text{eff}} / K\bar{v}A, \qquad (2)$$

where c_H is the HEVAC compression, $V_{\text{eff}} \equiv N/n(B_0)$ is the effective volume[2] of the HSC (with total number of particles N), A is the cross-sectional area of the fill tube with Clausing factor K, and $\bar{v}$ is the average atomic velocity of the gas. The expression for τ is again only valid in the low-density limit; it neglects any recombination in the cell and thus the decay is fully ascribed to free atomic flow of H↓ towards the HEVAC where an atom has a probability of $1/c_H$ to escape. This model predicts the density to change exponentially towards a new steady-state value when the conditions are changed.

In our present geometry the HSC and the HEVAC are in fields B_0 and $0.56B_0$, respectively. The HSC can be varied in temperature from ~0.27 to 0.8 K; the HEVAC operates at ~0.5 K. Below the HEVAC the temperature rises rapidly to 4.2 K at an accommodator[3] used to cool the H gas generated in a room-temperature discharge. It is quite clear that the configuration differs from the ideal uniform temperature system, yet we believe that our system can be approximated by Eqs. (1) and

(2) with c_M evaluated with respect to the field at the HEVAC. When the discharge is turned on a steady-state density (n_{eff}) of cold H is maintained in front of the HEVAC in spite of surface recombination processes. If we assume these processes to be first order in the density we expect n_{eff} to scale linearly with the incoming flux, φ; we conclude[4] that $n(B_0) \sim c_H c_M \varphi$.

Measurements were made by filling the cell at a constant flux for specified periods. The density was determined by triggering the stable H↓ gas to recombine by means of a bolometer.[1] The recombination energy was mainly dissipated in the cell and caused an easily measured temperature rise of the HSC with a characteristic wave form. The cell was calibrated for density by electronically generating heat pulses in the bolometer to reproduce this wave form. Since after triggering, atoms and recombined molecules could also be blown out of the HSC, our measured densities represent lower bounds.

We shall first discuss our low-density ($n \lesssim 3 \times 10^{14}$ atoms/cm^3) results. In Fig. 1(a) we show a typical curve for the time dependence of filling the HSC. When the discharge is turned off, the density in the cell decays exponentially with a time constant τ as shown in Fig. 1(b). In zero field at $T = 0.27$ K we measured[1] $\tau \equiv \tau_0 = 1.5 \pm 1$ sec. For 4 T, 0.39 K and 8 T, 0.37 K we find 38.6 ± 1 and 1998 ± 37 sec, respectively. The long-
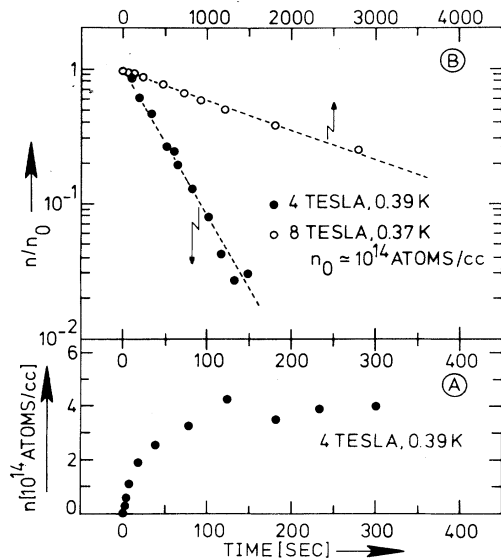
est period of stabilization was 47 min before triggering.

Writing $\tau = \tau_0 (V_{eff}/V_0) \exp[\gamma \cdot \mu_B B_0/kT]$, where V_0 is the geometric volume of the cell and $\gamma \equiv (B_0 - B)/B_0$, enables us to solve for τ_0 and γ using the measured time constants for 4 and 8 T. This yields $\tau_0 = 1.7$ sec and $\gamma = 0.53 \pm 0.05$, where we give the error in γ due to uncertainties in temperature. The value of τ_0 agrees well with our direct measurement at $B_0 = 0$; $\gamma = 0.53$ corresponds to a point within 2 mm of our estimate of the location of the compression side of the HEVAC,[5] suggesting that c_M should be calculated with respect to the field in the HEVAC.[6] In some circumstances we observed almost steplike changes of the time constant by factors of 2 or 3. We believe this may be due to different modes of operation of the HEVAC. During the measurements of τ at 4 and 8 T used to calculate γ, changes of this nature were not observed. At higher densities, $n \gtrsim 10^{16}$ atoms/cm^3, we observed initial decay rates of order five times faster so that simple exponential decay no longer applies. This may imply that recombination is becoming a problem.

One of our objectives has been to determine the magnetic equation of state. In steady-state equilibrium we can write $n_{sat}(B_0) = \varphi \tau_0 c_M/V_0$. Flux was easily measured by filling the HSC for a time $t \ll \tau$ and triggering to determine the number of atoms N or $\varphi = N/t$. In Fig. 2 we plot the satura-

FIG. 1. (a) Density as a function of time during filling of the cell at constant φ, B_0, and T. (b) Decay of $n(B_0)$ in time at constant B and T. The upper scale is for the 8-T data.
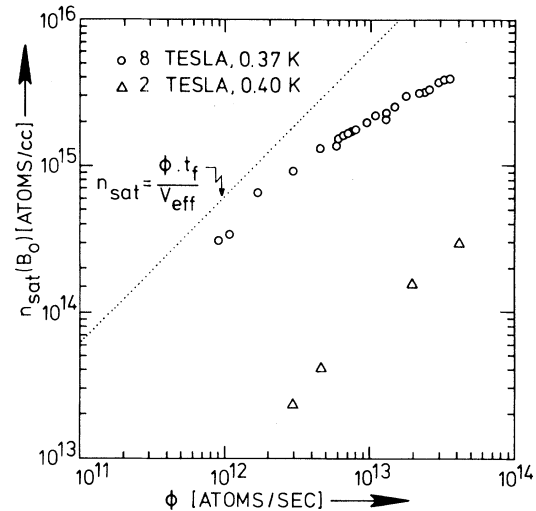

FIG. 2. Saturation density as function of the incoming flux at constant B_0 and T. The dotted line represents the maximum possible density in case of systematic error.

tion density as a function of flux, with B_0 and T held constant. The simple theory suggests a linear relationship which is found for low density (triangles). However, for higher density (circles), we see a deviation. Our detection method only determines a lower bound of $n(B_0)$. We can also set an upper bound by noting that the measured density must be less than $\varphi t_f / V_{eff}$, where t_f is the fill time used to ensure that $n(B_0) = n_{sat}$. This upper bound is given by the dotted line. In Fig. 3 we plot the saturation density versus B_0/T for a constant flux, $\varphi \simeq 1.3 \times 10^{13}$ atoms/sec. The open circles represent data obtained from variation of the magnetic field at constant temperature while the solid circles are obtained by varying the temperature, keeping the field constant. The dashed line represents the predicted behavior where n_{sat} = $\varphi \tau_0 c_M / V_0$ and $\gamma = 0.53$; the dotted line again represents an upper bound as determined by the integrated flux of atoms. The data are corrected for changes in the mode of operation of the HEVAC which were detected by measuring the saturation density at regular time intervals for a reference value of B, T, and φ. The instability of operation was mainly responsible for the scatter in the data. The statistical error in measurement of field, temperature, time, and the wave forms used to determine the density was at most a few percent. However, we estimate that the density calibration, which covered more than three orders of magnitude could have a systematic error, in par-
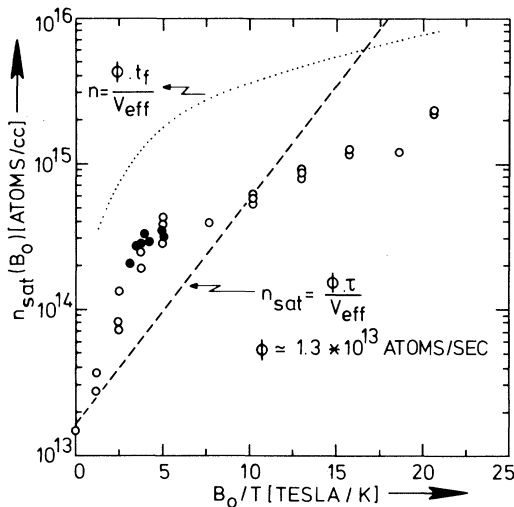
ticular at high density, of order 2. At a density of 10^{15} atoms/cm^3 the measured density could be as much as a factor of 5 lower than the actual density due to leakage of recombination energy (or atoms) out of the HSC.

The highest H↓ density achieved to now is $n(B_0)$ $\geq 1 \times 10^{16}$ atoms/cm^3 in 10.5 T at 0.4 K. As the flux is increased the HSC warms up, probably due to recombination that occurs during loading and sorting out of spin-up and spin-down atoms. Thus our maximum density is limited by our current refrigeration power. The flux itself was never a limit and usually was restricted to (1–10)% of its maximum value.

The presented data clearly show how higher densities can be achieved in the HSC. The simple theory of compression appears to have some validity but is not accurately applicable due to non-uniformities in the temperature. Moreover at higher densities we only observe a lower limit for the density, complicating interpretation. Deviations from the simple theory observed for higher densities may possibly be related to the location of the HEVAC and can be resolved by changing the geometry. If deviations are due to the onset of recombination in the HSC, then the problem is fundamental. However, density decay can also arise from recombination in the tail of the density distribution where a dense gas of helium can promote three-body recombination, or where the walls do not have a proper helium coverage.

We also report an interesting observation concerning the detection of H↓. With a sufficient number of atoms in the cell, after triggering, the cell temperature rises to a temperature T_f and then remains constant, for periods t_b up to 2.6 sec. Simultaneously triggering both bolometers in the HSC in order to double the active surface area did not affect T_f or t_b for a given density. However, simple kinetic considerations suggest that the rate of recombination is proportional to the bolometer area and that the process should be completed within tens of milliseconds.[1] This indicates that another (unidentified) process becomes important at higher densities. A possible explanation is that after triggering, the initial recombination heats the cell to temperatures greater than that of the HEVAC, also vaporizing some helium. At this point the helium vapors begin to flow towards the HEVAC, compressing the H↓ between the HSC and the HEVAC. This is a self-controlled feedback process that keeps the temperature of the HSC constant by recombining the H at a constant rate as long as a sufficient

FIG. 3. Saturation density as a function of B_0/T for constant incoming flux. For densities above $\sim 3 \times 10^{14}$, the data are thought to represent a lower limit. The dotted line represents an upper limit. For the dashed line, see text.

number of atoms are available and for which t_b is independent of the bolometer area.

Two of us (I.F.S. and J.T.M.W.) have injected gas from a deuterium discharge into the HSC. The resultant trigger signals correspond to disappointingly low densities of stabilized gas. This result could be due to a higher surface adsorption energy[7] for D as compared to H. It is quite clear that D behaves differently than H, even at low density. A complete report will be made after further investigations are completed.

tation from H↑ to H↓ for spin polarized H, which is preferable since we stabilize the down ($m_s = -\frac{1}{2}$) spin state.

[1]I. F. Silvera and J. T. M. Walraven, Phys. Rev. Lett. 44, 164 (1980); note that here we have changed the no-

[2]J. T. M. Walraven and I. F. Silvera, Phys. Rev. Lett. 44, 168 (1980).

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

[4]Measurements were mostly done at ~0.4 K, limiting gradients to 25% of the cell temperature so that thermomolecular effects are small.

[5]This enables an improved estimate of $c_H = \frac{1}{4}\tau_0 K\bar{v}A/V_0$. Use of $V_0 = 1.44$ cm^3, $\bar{v} = 93$ m/sec, $A = 0.125$ cm^2, and $K = 0.11$ with $\tau_0 = 1.7$ sec leads to $c_H = 38$. This value differs from that given in Ref. 2 as we have now determined the helium condensation point of the HEVAC (the compression side) to be further from the HSC, changing V_0 and K. In Ref. 2 due to a typographical error K was given as 0.1 instead of 0.18; the calculated estimate, $c_H = 79$, was correct as given.

[6]In Ref. 2 time constants were calculated taking $B = 0$ in the low-density tail.

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