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Compression of Spin-Polarized Hydrogen to High Density

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A new technique enabled the authors to compress doubly polarized hydrogen by up to five orders of magnitude and study the sample in very small volumes at constant density up to $2\times10^{18}/\mathrm{cm}^3$. The first determination of the bulk three-body dipolar recombination rate was made, $K_V^{3b}=4(1)\times10^{-39}\ \mathrm{cm}^6\ \mathrm{s}^{-1}$. The authors also identified a new process, bulk electronic dipolar $b\rightarrow c$–state relaxation with rate constant $G_V^{bc}\exp(2\mu_B B/k_B T) = 8(4)\times10^{-16}\ \mathrm{cm}^3\ \mathrm{s}^{-1}$. Samples were very delicate and too rapid a compression could result in an explosion.

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Since spin-polarized atomic hydrogen (H↓) was first stabilized there has been an intense effort to obtain high enough densities or low enough temperatures to observe degenerate properties of this boson gas.[1] A prime goal is to observe Bose-Einstein condensation (BEC), which, in the limit of weakly interacting bosons, should occur at $T_c = 3.31\hbar^2 n^{2/3}/mk_B$ (=74 mK for a density of $n = 10^{19}/\mathrm{cm}^3$; m is the hydrogen mass). H↓ is a gas with atoms in two hyperfine states: the ground state $|a\rangle = |\downarrow\Downarrow\rangle - \epsilon|\uparrow\Uparrow\rangle$ and the pure spin state $|b\rangle = |\downarrow\Downarrow\rangle$ (here ↑ and ⇑ denote electron and nuclear spin-$\frac{1}{2}$ projections and $\epsilon = a/4\mu_B B$ is the hyperfine mixing parameter with a the hyperfine constant, μ_B the Bohr magneton, and B the magnetic field). Recently, a gas of hydrogen with almost all atoms in the state $|b\rangle$, called doubly polarized hydrogen (H↓⇓), has been produced[2-5] as follows. Collisions involving an atom with a spin-up admixture can result in recombination. Thus, a-a and a-b, but not b-b, collisions can lead to H_2 formation. Because of the slow $b\rightarrow a$ relaxation rate, G^{ba}, a gas of H↓ evolves to H↓⇓ by this preferential recombination. The lifetime of H↓⇓ is substantially longer than H↓ since it is controlled by G^{ba}, not the recombination rate constant. Densities as high as $10^{17}\ \mathrm{cm}^{-3}$ of H↓⇓ have been produced by preferential decay; this gas can potentially be compressed to very high density.

In this Letter, we describe a new experimental approach which has enabled us to increase the maximum densities by more than an order of magnitude to $2\times10^{18}/\mathrm{cm}^3$. The volume of the gas of H↓⇓ can be compressed by a factor 10^5. All earlier hydrogen studies have used fixed-volume cells and the decay of the density was studied. Here we have developed a new type of cell which enables us to study high-density samples for periods of order minutes at constant hydrostatic pressure and temperature, observing the volume of the compressed hydrogen "bubble" decay away. We can measure volumes as small as $3\times10^{-3}\ \mathrm{mm}^3$. Such small volumes have important technical advantages for obtaining BEC. The temperature is limited by the heating rate $\frac{1}{2}D\,dN/dt$ ($D = 4.6$ eV is the dissociation energy of H_2, and $N = nV$ is the number of atoms in volume V). For our smallest volume, and densities of $10^{19}/\mathrm{cm}^3$, there are only 3×10^{13} atoms in the bubble, or about 10 μJ of potential recombination energy which can easily be accommodated by a moderate-size dilution refrigerator without excessive warming. Furthermore, the small bubble diameter reduces the thermal gradient which can develop in the bubble as a result of thermal boundary resistance.

A number of new and interesting phenomena were observed by studying the volume decay rate as a function of magnetic field and temperature. This includes the first measurements of previously undetected decay mechanisms: three-body dipolar recombination and bulk electronic dipolar relaxation from the pure $|b\rangle$ state to the electron-spin-reversed state, $|c\rangle = |\uparrow\Downarrow\rangle + \epsilon|\downarrow\Uparrow\rangle$. The compressed bubbles of hydrogen were very delicate and under certain circumstances their lives were terminated by a spontaneous explosion (rapid recombination). Explosions could also be induced by a rapid compression. A technical cryogenic problem prevented us from obtaining

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temperatures lower than 600 mK for pure ^{4}He films. For ~1% mixtures of ^{3}He-^{4}He, measurements were made down to 200 mK. Since these bubbles were susceptible to explosion and possibly out of thermal equilibrium, quantitative results are only given for the pure ^{4}He case. The highest densities achieved were $2\times10^{18}/\text{cm}^3$ for ^{4}He and $1\times10^{18}/\text{cm}^3$ for ^{3}He-^{4}He, under the assumption of equilibrium.

The cell is shown in Fig. 1 and consists of a central cell (CC) in the bore of a superconducting magnet and an outer leg partially filled with liquid helium, containing a weight which can be vertically translated to displace the helium. The CC has upper and lower chambers separated by a midsection. The heart of the cell is a capacitive volume gauge mounted in the bottom of the midsection. The capacitor is made of two concentric spherical sections (radius of curvature, 10 mm) with a spacing of 0.9 mm. A reference capacitor plate was incorporated for added stability. The three sections are electrically isolated and floating with respect to ground. The measurement plates are made of silver sinter (0.1 μm grain size) to carry away the recombination heat. The thermometer is attached to the sinter block. The

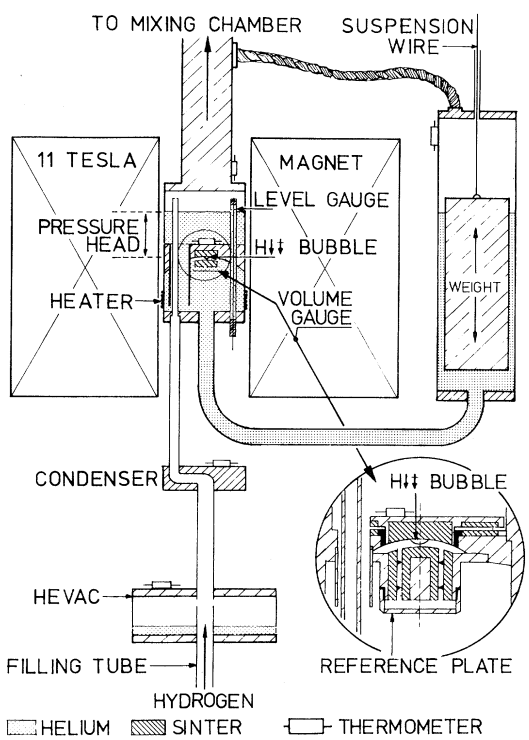

FIG. 1. Schematic diagram of the experimental apparatus.

capacitance of the capacitor increases substantially (5.7%) when it is filled with helium. The volume of an H bubble (assumed to have a negligible dielectric constant) is proportional (for small volumes) to the change of capacitance due to the displaced helium. Hydrogen is loaded into the CC from a room-temperature discharge. (The lower part of the cell is described elsewhere.)[1] With the weight up and the helium level in the CC down, the hydrogen diffuses into both the upper and the lower chambers. As the weight is lowered, the helium rises in the CC until it reaches the bottom of a tube concentric with the H↓ fill tube, sealing the 3-cm^3 volume of the lower chamber from the upper chamber. Any further increase of the He level compresses the hydrogen in the lower chamber. The gas is forced in between the capacitor plates, guided by the curved lower surface of the midsection. The pressure $p = p_s + p_k$ is determined by the hydrostatic head p_k of the helium and a surface-tension contribution p_s. The p_k is measured with a coaxial capacitance level meter. A maximum head of 12 mm was available (1-mm ^{4}He corresponds to $p_k = 1.4$ Pa or ~10^{-2} Torr) which could be measured with a resolution of 10^{-2} mm. The height of the helium in the CC could be modulated with an ~0.5-Hz ac heating; the thermomechanical effect (fountain effect) of ^{4}He leads to a level (i.e., pressure) and a volume modulation, giving directly the compressibility, providing a method for future detection of BEC.

To analyze the decay of the volume, we must know the shape of the bubble. For large volumes, it will conform to the top capacitor plate and have the shape of a skullcap, whereas for small volumes, the surface tension will dominate the buoyant forces and the shape will approach a sphere. For simplicity, we use the small-volume approximation and shall correct for the resulting systematic error. If we write the decay equation in the form $\dot{N} = -Vf(n)$, where $f(n)$ is polynomial in n, then since $\dot{N} = n\dot{V} + \dot{n}V$ and $\dot{n} = \dot{n}_k - \frac{1}{3}n_s\dot{V}/V = \frac{1}{3}n_s\dot{V}/V$ (where $n_k \equiv p_k/k_B T$, $n_s \equiv p_s/k_B T$, and p_s is assumed to scale with $V^{-1/3}$ since the surface tension is inversely proportional to the radius),

$$\dot{V}/V = -f(n_k + n_s)/(n_k + \tfrac{2}{3}n_s); \quad n_k \gtrsim n_s . \qquad (1)$$

A characteristic decay rate τ^{-1} may be defined for any volume (i.e., any n_s) by plotting V versus time on a semilogarithmic scale and determining the local derivative: $\tau^{-1} = d(\ln V/V_0)/dt = \dot{V}/V$. In Fig. 2 we show some typical decay curves.

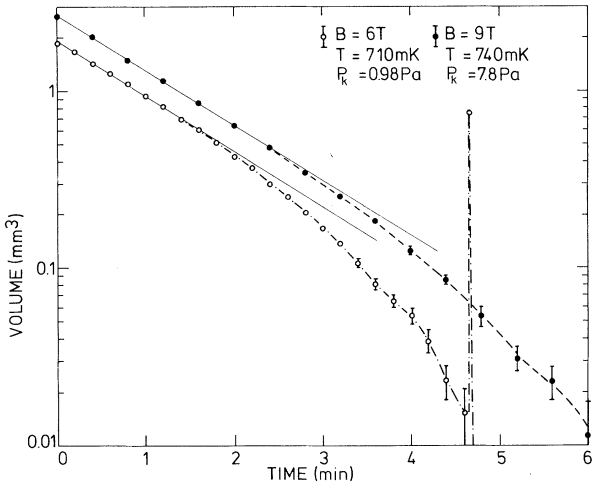

FIG. 2. Typical decay curves for ^{4}He surfaces. The low-field decay shows an explosion. The data points are taken from a recorder trace. The error bars are a measure of the noise. The dotted lines represent a guide to the eye.

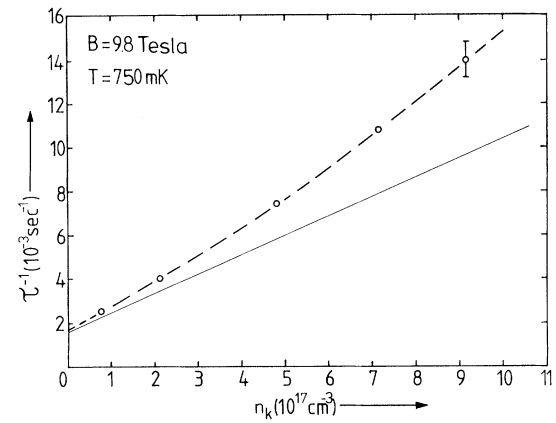

FIG. 3. Decay rate vs density. The dashed curve represents a second-order fit to the data used for extrapolation to zero density. The solid line corresponds to bulk relaxation, G_V.

The curvature for $V < 1$ mm^3 is interpreted as compression due to surface tension. The lower curve is terminated by an explosion. By plotting τ^{-1} as a function of n_k for a fixed volume (1 mm^3) as shown in Fig. 3, we observe curvature, pointing to a new fundamental decay process (third order in n). We first correct all data points for systematic error in the density determination (due to errors in the absolute helium level and the spherical assumption) by fitting with a second-order polynomial in n_k and extrapolating to $\tau^{-1} = 0$ to determine the systematic error Δn, which is used to correct the raw data; i.e., $n \equiv n_k + n_s + \Delta n$. The quantity $f(n)$ must have second- and third-order terms in the density. We assume the second-order term to be nuclear relaxation for a pure b-state gas so that n is the b-state density. In this case, the limiting recombination process is three-body dipolar volume recombination ($\dot{N} = -VK_V^{3b}n^3$) predicted by Kagan, Vartanyants, and Shlyapnikov[6] to have a value $K_V^{3b} = 4\times10^{-39}$ cm^6 sec^{-1} for $B = 10$ T and $T = 0$ K. We then have

$$f(n) = 2G_V^{ba}n^2 + K_V^{3b}n^3. \qquad (2)$$

A computer fit leads to $G_V^{ba}T^{-1/2}(1+16.68/B)^{-2} = 6.6(6)\times10^{-22}$ cm^3 K$^{-1/2}$ s^{-1} in agreement with the double-polarized picture and earlier experiments.[2,3,5] We also find $K_V^{3b} = 4(1)\times10^{-39}$ cm^6 s^{-1} for $T = 750$ mK and $B = 9.8$ T, in agreement with Kagan, Vartanyants, and Shlyapnikov. Here the error bars are statistical and do not include possible deviations from thermal equilibrium, to be discussed later.

A study of the magnetic field dependence of second-order decay (see Fig. 4) shows that the term G_V^{ba} alone is inadequate, implying that a new second-order process becomes important at lower fields. As spin exchange cannot relax the $|b\rangle$ state in H↓↑, the most likely candidate is the bulk electronic dipolar $|b\rangle \to |c\rangle$ relaxation discussed by Kagan, Vartanyants, and Shlyapnikov.[6] In Fig. 4 the solid line corresponds to the quoted value for G_V^{ba}; the dashed line is obtained from an estimate by Lagendijk[7] for the G_V^{bc} process (Kagan, Vartanyants, and Shlyapnikov did not evaluate the energy average of the cross section). This new process has an exponential B/T dependence, as established by temperature variation. From the

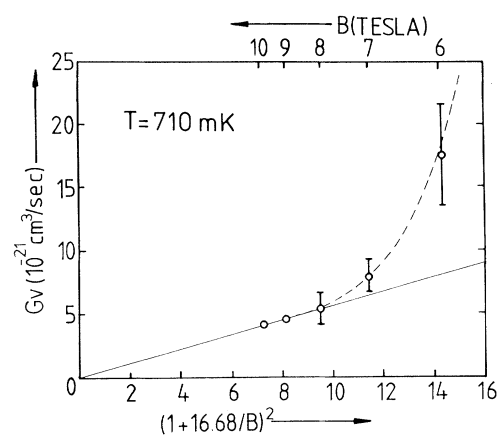

FIG. 4. The bulk relaxation rate constant $G_V = G_V^{ba} + G_V^{bc}$ (dashed line). The solid line is G_V^{ba}.

field dependence, we determined $G_V^{bc} \exp(1.34B/T) = 8(4)\times 10^{-16}$ cm^3 s^{-1}, in good agreement with theory.

At our current high densities, H↓↑ is a very delicate gas and a rapid increase in the pressure can result in an explosion of the bubble. This result is extremely important in relation to future experiments to observe BEC where rapid compressions are required in view of the expected short lifetime of the sample. For $B/T \lesssim 10$ T/K and small volume, the sample explodes spontaneously. However, for $B/T > 10$ T/K, it was possible to follow the decay of the bubble to the minimum detectable volume. The explosions may be understood if we assume the sample to be driven out of equilibrium by too rapid of a compression. Then the exponential temperature dependence of the $b \to c$ relaxation process can generate the required positive feedback to explain the occurrence of an explosion. "Spontaneous" explosions may possibly result from compression by surface tension in shrinking bubbles. On two occasions, we also observed an explosion to result from a decompression, a phenomenon which does not fit in the proposed picture.

We conclude with a discussion of thermodynamic equilibrium upon which the analysis of decay is based. The gas can be out of thermal equilibrium because of Kapitza resistance, with interfacial temperature differences ΔT_{K1} between H↓↑ and He,[8] and ΔT_{K2} between He and the cell walls. Our experiment and cell were carefully designed to minimize this problem by use of small bubbles, high temperature, and silver sinter. The worst situation is when the bubble volume-to-surface ratio is large; i.e., for large volume. If it is out of thermal equilibrium, then $V \to V_{eq}$; as the bubble shrinks, $V \to V_{eq}$ and curvature should be observed in Fig. 2 for large V, instead of the straight line. We estimate $\Delta T_{K2} \ll 1$ mK. For ΔT_{K1} we make a calculation for the highest density in Fig. 3, $n = 10^{18}$ cm^{-3}: $\dot{Q} = D\dot{N}/2 = \alpha(nvA/4) \times 2k\Delta T_{K1}$. Using the low-temperature (conservative) value of Salonen *et al.*[9] for the accomodation coefficient, $\alpha = 0.2$, we find the $\Delta T_{K1} \approx 3$ mK or $\Delta T_{K1}/T \lesssim 4\times 10^{-3}$, supporting our contention that samples are close to thermal equilibrium.

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