PHYSICAL REVIEW B VOLUME 38, NUMBER 13 1 NOVEMBER 1988

Spin-polarized hydrogen in high magnetic fields

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(Received 13 June 1988)

We have nearly tripled the range of magnetic-field strength over which bulk three-body recombination has been studied in spin-polarized atomic hydrogen. Analysis of our data (8–20 T) shows for the first time a qualitative agreement with theoretical predictions for the field dependence of dipolar recombination. Our results for the three-body exchange recombination are over an order of magnitude lower than a previously published value, and thus in reasonable agreement with theory. We discuss the prospects for observing Bose-Einstein condensation in very high magnetic fields.

The observation of Bose-Einstein condensation (BEC) in a nearly ideal Bose gas has been a vigorously pursued goal since the stabilization of atomic hydrogen.[1] Because the critical temperature T_c for BEC is proportional to $n^{2/3}$ (the gas density $n = 1.6\times10^{19}$ atoms/cm^3 corresponds to $T_c = 100$ mK), efforts were made to raise the density, resulting in an increase of more than four orders of magnitude. This increase stopped abruptly at about 5×10^{18} cm^{-3} because of the rapid three-body recombination of hydrogen, both in the bulk gas and especially adsorbed on the confining helium surfaces.[2–4] This recombination channel not only limited the density, but also heated the gas far above T_c. To advance further towards the goal, two different approaches were followed. Hess *et al.*[5] devised a magnetic trap to isolate the hydrogen from the helium walls. This intrinsically low-density system requires extremely low temperatures for BEC. We have pursued a high-density approach with an effort to suppress the three-body recombination by substantially increasing the magnetic field above values used in earlier studies. Although the theory of Kagan, Vartanyantz, and Shlyapnikov[6] and de Goey and co-workers[7,8] predicted the three-body recombination rate constant to increase with field, peaking at about 15 T and then falling sharply, all earlier experiments[2–4,9] have shown a rate constant that decreases weakly with increasing field from 4.5–9 T, so that the magnetic-field dependence was a major puzzle and an open problem. We have studied the bulk three-body recombination rates in fields up to 20 T at the Francis Bitter National Magnet Laboratory (FBNML). Our data qualitatively support the existing theoretical description in the high-field range implying that much larger fields are required to approach the high-density BEC conditions. A more sophisticated theory is required to understand both the low- and high-field behavior; we comment more on this at the end of this Brief Report.

Our experimental technique is to generate a high-density sample of atomic hydrogen by compressing the volume of a low-density sample by several orders of magnitude, leaving a small bubble of hydrogen gas immersed in a bath of superfluid ^{4}He. Loss of atoms due to recombination of H_2 causes the volume of the gas bubble to shrink. We measure the temporal decay of the volume under constant hydrostatic pressure and temperature to obtain information about the recombination rate constants. Similar compression schemes have been used by previous investigators at fields under 10 T.[2,4]

A schematic representation of our cell is shown in Fig. 1. The cell body is constructed out of 1266 epoxy, strategically imbedded with a great number of very fine copper wires to allow good thermal conduction while suppressing eddy current heating from the magnetic-field ripple. The cell is filled with an adequate quantity of liquid ^{4}He, the level being controlled with a displacer. The latter is a

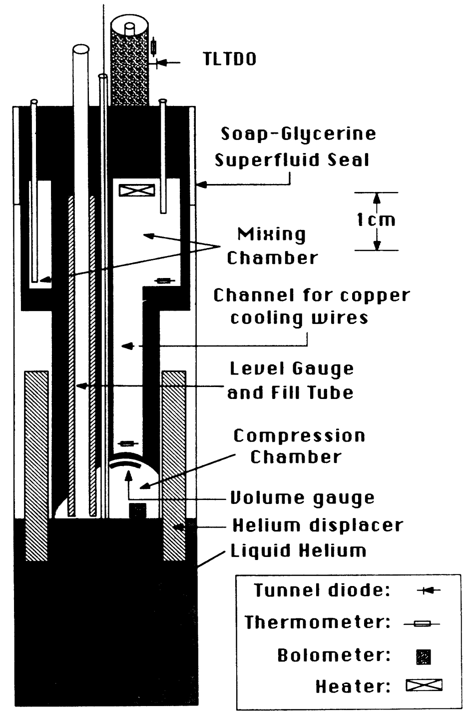

FIG. 1. Schematic diagram of the compression cell.

solid cylindrical annulus attached to a semirigid wire that runs up to room temperature and can be raised until the helium level drops below the lower end of the hydrogen fill tube. Atomic hydrogen from a room-temperature source can then flow down the fill tube and enter the compression space. When a significant number of atoms have accumulated in the compression space, the displacer is lowered, raising the helium level until the end of the fill tube is sealed off. Thereafter the hydrogen gas is compressed into an increasingly smaller volume, increasing the density. The spherically curved roof of the compression space forces the hydrogen to eventually end up as a small bubble squeezed between the capacitor plates (0.5 mm spacing) which form the volume gauge. As the H atoms decay, the volume of helium between the capacitor plates increases and the capacitance changes due to the dielectric constant of the liquid helium. These changes, which can be directly related to the volume changes, are monitored by a transmission-line tunnel-diode oscillator[10] (TLTDO) operating at ~200 MHz. The hydrostatic pressure head is measured by the level gauge, a split-cylinder capacitive sensor that also serves as an extension of the fill tube.

The temperature of the cell is stabilized by monitoring a carbon thermometer which is imbedded in the upper plate of the volume gauge. This thermometer was recently calibrated *in situ* against ^{3}He vapor pressure up to 20 T. We believe that our thermometry is accurate within 5%. Since it has been previously shown[11] that even small amounts of ^{3}He can drastically alter the effective surface recombination rates, we have waited until after the data were collected to do the calibration. This resulted in a 100-mK shift down from extrapolations of earlier zero-field calibrations on a second thermometer, and thus the true measurement temperatures were not quite optimum for the study of a pure bulk effect.

Decays are frequently terminated by spontaneous explosions as the hydrogen bubble shrinks to a very small volume. Rapid time resolution of the decay curve often reveals pronounced oscillatory structure prior to explosion. The period of oscillation is about 1 sec. We suspect that these oscillations may be driven by the thermal energy released during recombination. We have not attempted to analyze regions of the decay curves that show oscillations.

There are several different decay processes occurring between atoms in the four hyperfine states of hydrogen, and the populations of these states change with time. At low densities, recombination is dominated by two-body collisions that occur in the two-dimensional (2D) layer of gas adsorbed on the surface of the helium. This is described by a recombination rate constant K_{ij}^{eff}, where i,j designates the hyperfine states of the two atoms and "eff" means the effective rate for processes both on the surface and in the bulk. The wave function of the lowest-lying hyperfine state (the "*a* state") contains a small admixture of the reversed electron spin; this results in preferential decay of the *a* state so that the sample becomes enriched with atoms in the second-lowest energy level, the pure spin-down *b* state. This state is highly stable,[12] limited only by slow relaxation, with rate constant G_{ij}^{eff} down into the *a* state, before recombination can take place. At higher densities, however, the collision of three hydrogen atoms, both on the surface and in the bulk, becomes more probable; such events allow the electron spins to be flipped due to the magnetic dipole-dipole interaction, and recombination results. In addition, since in our experiment the polarization ratio N_a/N_b (where N_i is the number of atoms in the ith state) develops from 1.0 to about 0.15, there is a contribution to three-body decay which involves both *a* and *b* atoms, mediated by the exchange interaction, as suggested by Kagan *et al.*[3] The goal of our data analysis is to determine the three-body dipolar rate constant, K_{bbb}^{eff}, and the three-body exchange rate constant, $C_{\text{ex}}^{\text{eff}}$. We model the decay of the hydrogen bubble with the following set of coupled differential rate equations:

$$\begin{aligned}\frac{\dot N_a}{V} = &-(2+\xi\eta)\dot r_a-\xi\eta(\dot r_a+\dot r_b)[n_a/(n_a+n_b)]\\ &-2K_{aa}^{\text{eff}}n_a^2-K_{ab}^{\text{eff}}n_an_b-G_{ab}^{\text{eff}}(n_a+n_b)(n_a-n_b)\\ &-C_{\text{ex}}^{\text{eff}}(\tfrac{8}{3}n_a^3+\tfrac{4}{9}n_bn_a^2)-\tfrac{2}{3}\xi\eta K_{bbb}^{\text{eff}}(n_b-n_a)n_an_b\ ,\end{aligned} \tag{1}$$

$$\begin{aligned}\frac{\dot N_b}{V} = &-(2+\xi\eta)\dot r_b-\xi\eta(\dot r_a+\dot r_b)\left[\frac{n_b}{n_a+n_b}\right]-K_{ab}^{\text{eff}}n_an_b\\ &+G_{ab}^{\text{eff}}(n_a+n_b)(n_a-n_b)-C_{ex}^{\text{eff}}(\tfrac{2}{9}n_bn_a^2-\tfrac{2}{3}n_a^3)\\ &+\tfrac{2}{3}\xi\eta K_{bbb}^{\text{eff}}(n_b-n_a)n_an_b\ ,\end{aligned} \tag{2}$$

$$\frac{\dot V}{V} = (\dot N_a+\dot N_b)/(N_a+N_b)\text{Cor}(V,P)\ , \tag{3}$$

with

$$\dot r_a = K_{bbb}^{\text{eff}}(n_a^3+\tfrac{2}{3}n_bn_a^2+\tfrac{1}{3}n_an_b^2)\ ,$$

$$\dot r_b = K_{bbb}^{\text{eff}}(n_b^3+\tfrac{1}{3}n_bn_a^2+\tfrac{2}{3}n_an_b^2)\ ,$$

and

$$\text{Cor}(V,P) = 1+V/P(dP/dV)\ ,$$

where V is the volume, P is the pressure, and $n_iV=N_i$.

These rate equations, which we now describe are discussed in detail in Refs. 1 and 2. The two sparsely populated upper hyperfine levels (*c* and *d* states) are dynamically pumped by the three-body dipolar recombination process; we take this into account directly in our model by using the calculated values[8] for the fraction of recombination events that flip the third body into one of the upper levels ξ. We assume that atoms in the upper levels (electron spin reversed) recombine rapidly upon collision with atoms in the lower levels, and that the collision probability scales linearly with the density of the target atom. We approximate the effect of the small fraction of upper level atoms that relax back down before recombining by including the factor $\eta \approx 0.80$.

The last terms in Eqs. (1) and (2) are included for completeness but have no significant effect on the final results of our data analysis; they are due to two-body spin exchange collisions which cause a rapid rearrangement of the spin states before upper-level atoms recombine with lower-level atoms. Spin exchange preserves the equality $n_a/n_b=n_d/n_c$, even in the presence of dynamical pumping by three-body recombination. The inclusion of these last terms makes our equations equivalent to those of Ref. 3 in the $\eta = 1$ limit.

The effect of the surface tension of the ^{4}He on the pressure of the hydrogen bubble is accounted for by using the "hemispherical miniscus model" used in Ref. 4. The fact that the surface tension contribution to the pressure increases as the hydrogen volume decreases is accounted for in Eq. (3) by the factor "Cor."[2]

We perform a three-parameter least-squares fit ($K^{\rm eff}_{bbb}$, $C^{\rm eff}_{\rm ex}$, and initial N_a/N_b ratio) to each decay curve, using an exact numerical solution to the above coupled rate equations. K_{ij} are taken from Ref. 13; $G_{\rm eff}$ is taken from Ref. 2. We average the results of four decays at 8 T, nine decays at 10 T, seven decays at 12 T, six decays at 15 T, five decays at 17 T, and four decays at 20 T. Figure 2(a) shows the weighted mean values for our results for the three-body dipolar rate constant along with recent theoretical results of Stoof, de Goey, and Verhaar.[14] Figure 2(b) shows our results for the three-body exchange rate constant along with the theoretical predictions of Kagan *et al.*[6] The error bars shown are one-standard-deviation statistical errors. We have tried to remove any sources of systematic error from the data; however remaining uncertainties in fixed parameters (e.g., temperature of hydrostatic pressure) may contribute a systematic error approximately as large as the statistical errors shown. For the dipolar rate, we stress that the 15 and 17 T points are significantly higher than all other points, as was the total three-body decay rate at these field values. The fit parameters $C^{\rm eff}_{\rm ex}$ and N_b/N_a are highly correlated variables for many of our decays and the subsequent error in $C^{\rm eff}_{\rm ex}$ is large. In fact, the weighted means presented in Fig. 2(b) for 8 and 15 T were each essentially determined by a single decay curve since the errors associated with the others were so large. Although the relative error bars are too large to determine the field dependence, our results for the magnitude of $C^{\rm eff}_{\rm ex}$ are in reasonable agreement with theory and differ significantly from that of Bell *et al.*[3] whose value was over an order of magnitude larger.

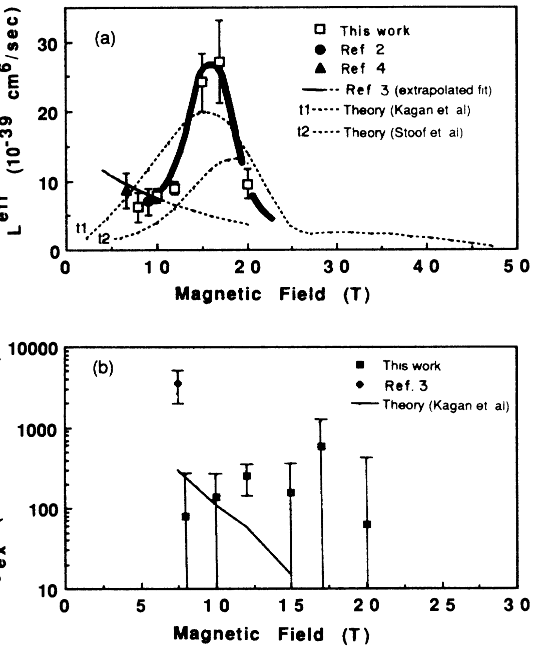

FIG. 2. (a) Field dependence of the effective three-body bulk dipolar recombination rate constant, $L^{\rm eff} \equiv 2K^{\rm eff}_{bbb}(1+\xi\eta)$. Our new high-field data are shown by the open squares; the thick solid line is a guide to the eye. The solid/dashed curve sloping monotonically downward to the right is an extrapolation of the fit of Bell *et al.* (Ref. 3) to their data below 10 T. Other dashed curves show theoretical results of Stoof *et al.* (Ref. 14) in the approximation of Kagan *et al.* (Ref. 6) (upper) and using the exact initial state and distorted final state wave function (lower); both curves are in the $\eta = 1$ limit, and neglect dipole-exchange (Ref. 16) contributions. Also shown are experimental values from Refs. 2 and 4. (b) Field dependence of three-body exchange recombination rate constant, $C^{\rm eff}_{\rm ex}$. Solid curve is theory of Kagan *et al.* (Ref. 6). Truncated error bars extend to zero. Also shown is the experimental value from Ref. 3.

We believe that the three-body rate constants which we have measured are dominated by bulk three-dimensional (3D) events; however, there may be a small contribution due to the surface atoms. Previously published values[3,9] for the surface rates at 8 T indicate that the surface contribution may be as much as 16% at the lowest temperatures accessed in this experiment. The bulk three-body recombination rates are predicted to have a very weak temperature dependence, whereas the effective surface rates are strongly temperature dependent. Half of the data from both the 15 and 20 T sets were taken at 500 mK and the other half at 545 mK; there was no statistically significant difference between the two, lending credence to the suggestion that the three-body decay constants that we have measured are dominated by the bulk rate constants. The 10, 12, and 17 T data were all taken at 500 mK. The 8 T data were taken at 600 mK in a superconducting magnet.

The field dependence of our data displays the characteristic peak predicted by theory for dipolar recombination into the final molecular state with vibrational quantum number $v = 14$, rotational quantum number $J = 3$. Recombination involves the flipping of either one or two spins on the three atoms involved in a collision. For fields below 27 T, the dominant channel is believed to be the two-spin-flip process (2SF channel). Simple energy conservation considerations require that this channel be rapidly quenched when the magnetic field is raised sufficiently that the Zeeman energy change is greater than the recombination energy; thereafter decay only occurs via the weaker single spin-flip channel (1SF). The most recent spectroscopic data for the $v = 14$, $J = 3$ level of hydrogen[15] put the cutoff field at 27 T for 2SF and 54 T for 1SF. Currently, the highest laboratory dc magnetic field available in a large volume is 31 T, and one expects dc fields of order 40 T in the next generation of magnets. In going to higher magnetic fields the decay rate into the deeper $v = 14$, $J = 1$ state of H_2 grows, and an important question remains as to the strength of this channel above the 54 T cutoff of the $v = 14$, $J = 3$ level. The calculations of Stoof *et al.*[14] are being extended to higher fields and are clearly of great importance. An extension of the measurements to the 30 T region to examine the 27 T 2SF cutoff might well help clarify the situation. Apparently a substantial decrease in the recombination rate is still required to enable a straightforward high-density observation of BEC. Moreover, the dominant heating process at low temperature is the three-body surface recombination for which theory and experiment disagree in low fields, and which has not yet been studied in high fields.

Finally, we are left with reconciling the measurements below 10 T with theory. An attractive possibility is that recombination in low fields is dominated by the dipole-exchange mechanism, recently proposed by de Goey *et al.*,[16] which falls off rapidly with increasing field.

We would like to thank P. Wolff and L. Rubin for their hospitality at the Francis Bitter National Magnet Laboratory (FBNML). Useful contributions were made by C. Mallardeau in the early stages of the development of the experiment. We also thank G. Schmiedeshoff and P. Emery for assistance. We thank T. J. Greytak for a discussion of the rate equations. The financial support of the U.S. Department of Energy (DOE) Grant No. DE-FG02-85ER45190 is acknowledged. The Boston University Low Temperature Group is supported by National Science Foundation (NSF) Grant No. DMR-85-13626.

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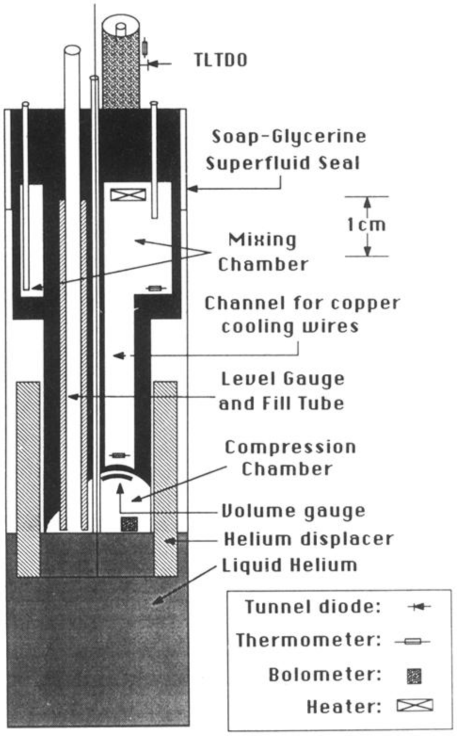

FIG. 1. Schematic diagram of the compression cell.