Observation of Three-Body Recombination in Spin-Polarized Hydrogen

Harald F. Hess, David A. Bell, Gregory P. Kochanski, Richard W. Cline,[a]
Daniel Kleppner, and Thomas J. Greytak
Department of Physics and Center for Material Science and Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

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Three-body recombination in spin-polarized hydrogen has been observed. Doubly polarized hydrogen was compressed by a piston into a thin cell and the temperature and pressure were independently monitored. Measurements were carried out in the temperature range 0.3 to 0.45 K in a magnetic field of 8.0 T at densities up to 1.5×10^{18} cm^{-3}. The results are in good agreement with the theory of Kagan, Vartanyants and Shlyapnikov.

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Kagan, Vartanyants, and Shlyapnikov[1] have predicted that three-body recombination will occur in spin-polarized hydrogen by a dipole interaction between the electronic magnetic moments of the atoms. This process, which occurs both in the bulk and in the surface, is ***not*** suppressed by nuclear polarization or by high magnetic fields. Verifying their prediction experimentally is important because at the high densities needed to observe the Bose-Einstein transition, three-body recombination would be faster than the decay due to two-body relaxation.[2,3] We have observed this three-body process by mechanically compressing a gas of doubly polarized hydrogen (electron and nuclear polarized) and studying the subsequent decay.

In our experiment the pressure and temperature of the gas in a small cell were measured. Figure 1 shows the apparatus. Doubly polarized hydrogen in a cylinder, 22 mm long × 4.8 mm diam, is compressed by a piston and passes through a small connecting tube into a thin pancake-shaped cell, 0.13 mm thick × 6.4 mm diam. The cell is lined with a saturated ^{4}He film; the helium also seals the piston by capillary condensation. The bottom of the cell is covered with 0.07-μm sintered silver to facilitate heat removal. The top of the cell is the 13-μm-thick Kapton diaphragm of a capacitive pressure transducer. A thin-film carbon thermometer[4] is plated on the lower side of the diaphragm. Kapitza resistance between the gas and the thermometer is unimportant because little of the recombination heat from the hydrogen flows into the Kapton diaphragm. Thus, the thermometer can be expected to give a reliable measure of the gas temperature. No corrections to the hydrogen pressure due to surface-tension effects are needed because of the geometry of the cell, and the thermal analysis is simplified by the fact that the heat flow is essentially one dimensional. In this first trial of the new apparatus the compression ratio was conservatively set at 70; this can be increased by making the cell thinner. Most of the compression takes place in about 1 s.

Figure 2(a) shows the pressure P and the temperature T in the gas following compression at an ambient temperature of 0.355 K. Note that at high pressure the gas temperature is significantly above the ambient. It is in this region that three-body effects are most important so that direct knowledge of the temperature is vital for interpreting the data. (Falling temperature, by itself,

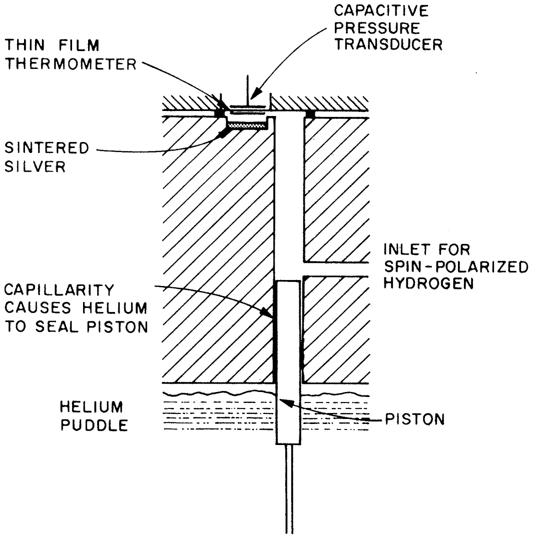

FIG. 1. Schematic drawing of the compression region of the apparatus. The compression cell was constructed from a 32-mm-diam copper rod and is suspended from a dilution refrigerator. Not shown are additional thermometers, heaters, bolometers, etc., and a second pressure transducer. The piston is driven by a computer-controlled stepping motor coupled through a bellows system.

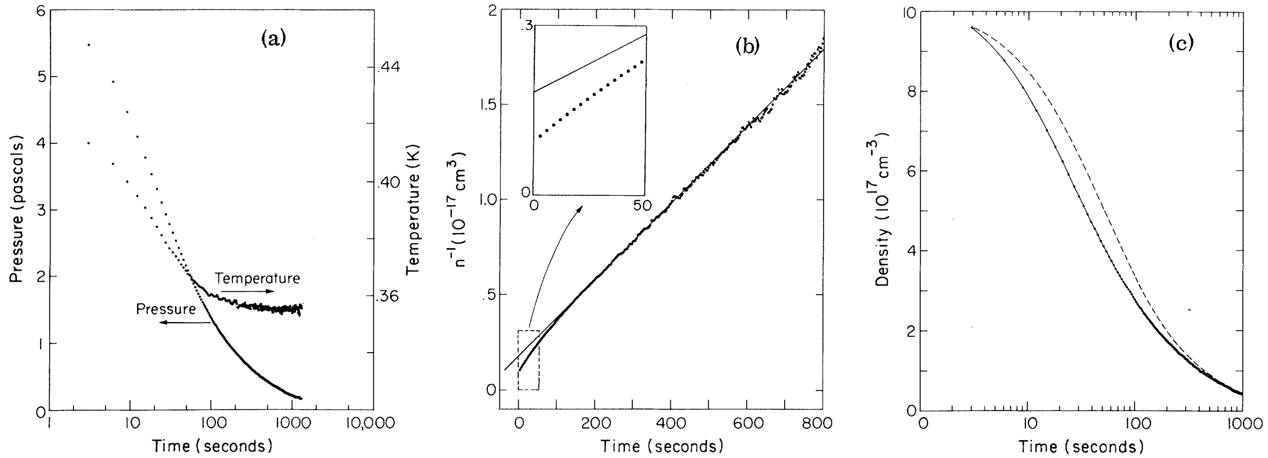

FIG. 2. Data from one typical compression; $B = 8$ T, T(ambient) $= 0.355$ K. (a) Measured pressure and temperature. (b) Reciprocal density computed from $n = PkT$. The straight line indicates the behavior expected for pure two-body decay; the deviation of the data for short times results from the three-body recombination. (c) Density. The points are data; the smooth line is a theoretical fit used to determine G and L; the dashed line indicates a pure two-body decay at the rate $2G$ determined from the straight-line fit to $1/n$ shown in (b).

can mimic three-body decay.) Because quantum corrections to the ideal gas law are not yet important, the density is computed with use of $n = P/kT$. If the gas were undergoing pure two-body decay at a rate limited by nuclear relaxation from the b to the a state (state a is the rapidly recombining "impure" state) with relaxation constant G,[2,3] then the inverse density would increase linearly with time:

$$1/n = 1/n_0 + 2Gt. \tag{1}$$

Figure 2(b) shows that over a large range $1/n$ displays the linear behavior of Eq. (1). The conspicuous curvature at short time indicates excess recombination, as would be expected from three-body effects. Before discussing possible fits to the curves, however, we introduce a construction which clearly demonstrates the three-body nature of the decay.

The presence of both nuclear relaxation and three-body recombination causes the density to decay according to $\dot{n} = -2Gn^2 - Ln^3$, where L is the three-body rate constant. Therefore,

$$-\dot{n}/n^2 = 2G + Ln. \tag{2}$$

A plot of $-\dot{n}/n^2$ is shown in Fig. 3. The cross at zero density indicates the value of $2G$ found from the slope of the data in Fig. 2(b), according to Eq. (1). Figure 3 shows that $-\dot{n}/n^2$ increases with the density in approximately the linear fashion given by Eq. (2). The dashed and solid curves are discussed below.

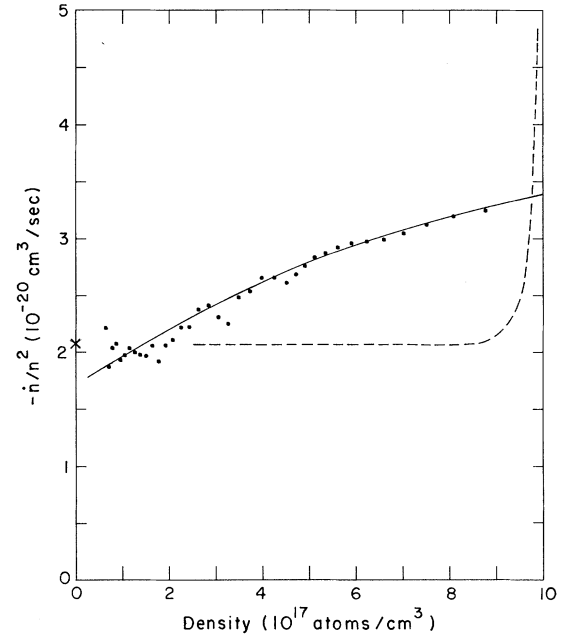

FIG. 3. A presentation of the data from Fig. 2 which illustrates the three-body nature of the decay. For pure two-body decay, $-\dot{n}/n^2$ would be constant, giving a horizontal line. [The cross shows where such a line would intercept $n = 0$ for the two-body rate determined from Fig. 2(b).] In the absence of thermal effects simultaneous two- and three-body decay would give a straight line with slope L and intercept $2G$ at $n = 0$. The solid line results from a fit to the data of $n(t)$ [Fig. 2(c)], including thermal effects on L and G. The dashed curve would result if $L = 0$ but the initial ratio of n_a/n_b were 0.04 instead of its equilibrium value for our geometry, about 4×10^{-3}.

An excess density of a-state atoms above the equilibrium value would also lead to increased recombination early in the decay. This might be mistaken for a three-body process. Such an excess could occur because the ratio n_a/n_b did not reach its low limiting value before compression, or because of a change in this limit due to the changing area to volume ratio during compression. Calculations shown by the dashed curve in Fig. 3, however, show that the evolution of $-\dot{n}/n^2$ would be entirely different from that governed by Eq. (2) and from the behavior displayed by the experimental data.

The physical situation is actually somewhat more complicated than Eq. (2) suggests as a result of the effect of the varying temperature on G and L. Some of the two-body relaxation and three-body recombination takes place while the atoms are adsorbed on the walls. The surface density σ is lowered early in the decay when the heating is greatest, decreasing the effective values of G and L. The thermometer is at the hottest part of the cell, and care must be taken to find the correct temperature of the lower surface where the density is highest. We have calculated the evolution of the system taking into account the following[5]: the theoretical thermal conductivity of the polarized gas,[6,7] our measured effective Kapitza resistance of the sintered silver surface, a value of the accommodation coefficient for heat transfer between the gas and the liquid helium deduced from our $T(t)$ data, deviations from the classical adsorption isotherms $\sigma(n, T)$ due to the quantum behavior of the adsorbed layer, and corrections to $\sigma(n, T)$ due to the gas being at a higher temperature than the adsorbed hydrogen. We find that at an ambient temperature of 0.355 K and above the sensitivity of the experiment to the choice of thermal parameters is weak. At the lower temperature of 0.300 K the thermal parameters have a significant effect, decreasing the reliability of the fitted value of L.

Figure 2(c) shows the calculated fit to the density at a temperature of 0.355 K. The fit is well within the experimental noise. It should be emphasized that *only* L and G were fitted; no thermal parameters were adjusted. A fit to the data in the absence of three-body recombination is shown for comparison. To confirm the consistency of the analysis, the calculated values of $-\dot{n}/n^2$ for this best-fit curve are plotted in Fig. 3, along with the experimental values. The curvature of the line is due to thermal effects.

Our measured values of the surface relaxation constant G_s fall within 50% of the value which we measured earlier.[3] Inspection with a microscope indicates that the Kapton (with its thermometer) and the sintered silver surface are smooth with no visible pits. Capillary condensation of the liquid helium would fill in any pits too small to see, resulting in a microscopically smooth surface. The surface is essentially perpendicular to the field, whereas in our earlier work[3] it was largely parallel. We conclude from this that the angular anisotropy of the surface relaxation on ^{4}He, if it exists, is small. In contrast, Sprik *et al.*[8] see a reduction of the relaxation rate on a perpendicular surface when ^{3}He is added.

Figure 4 shows our measured values of L. The work of Kagan, Vartanyants, and Shlyapnikov[1] leads to an expression for L of the form

$$L = L_g\left[1 + \frac{\Lambda^3(T)}{R_0^2}\,\frac{A}{V}\,\exp\left(+\frac{3E_b}{kT}\right)\right], \qquad (3)$$

where L_g is the three-body rate constant in the gas, R_0 is a characteristic interaction radius for atoms on the surface, $\Lambda(T)$ is the thermal de Broglie wavelength, and E_b is the binding energy of hydrogen atoms to the helium. The second term in the parentheses arises from the

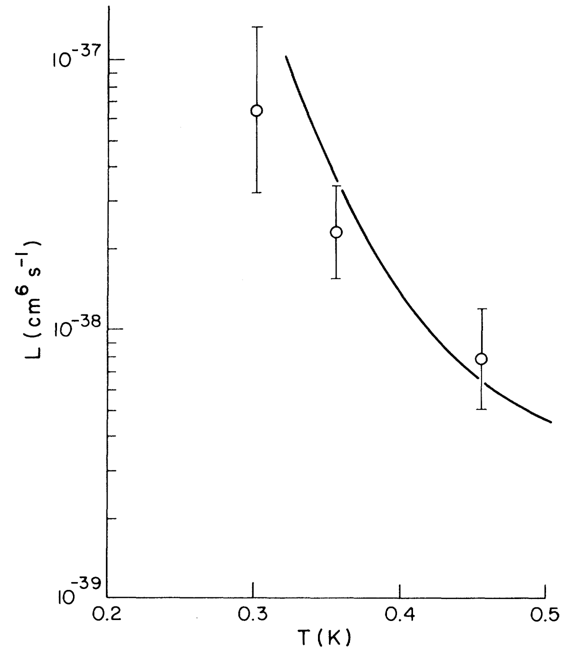

FIG. 4. Values of the three-body rate constant L determined from our measurements. The solid line is the prediction of Kagan, Vartanyants, and Shlyapnikov (Ref. 1). The error bars are based on the statistical error and our estimates of uncertainties in the fitting procedure.

atoms absorbed on the surface. The calculations of Kagan, Vartanyants, and Shlyapnikov yield $L_g = 3\times 10^{-39}$ cm^6 s^{-1} for the pure b state at a magnetic field of 8 T. They estimate R_0 to be 0.4 nm. We have plotted Eq. (3) in Fig. 4 using our area to volume ratio $A/V = 160$ cm^{-1}, and $E_b = 1.0$ K. At our highest temperature, 0.456 K, the ratio of the surface term in Eq. (3) to the bulk term is 1.2. The agreement with the measured values is satisfactory except, perhaps, at the lowest temperature. At this temperature, however, the interpretation is strongly affected by the values of the thermal parameters.

The implications of this three-body recombination for the study of the Bose-Einstein transition is easily understood by examining the characteristic decay time $\tau \equiv (\dot{n}/n)^{-1}$ for the density at which the transition should occur. Consider bulk processes only. With use of a value[3] of $G_g = 4 \times 10^{-21}$ cm^3 s^{-1}, the time for two-body relaxation is given by $\tau_2 = 0.25 T_c^{-3/2}$ s, where T_c is the transition temperature in kelvins. τ_2 is experimentally manageable, 2 s at 0.3 K. On the other hand, with use of $L_g = 3\times 10^{-39}$ cm^6 s^{-1}, the time for three-body relaxation is $\tau_3 = 1.4\times 10^{-3} T_c^{-3}$ s. At 0.3 K, $\tau_3 = 50$ ms. Heating due to such a rapid decay presents a major obstacle to achieving the Bose-Einstein transition in this temperature region. At much lower temperature three-body processes in the gas would not be a serious problem, but surface processes would be. Kagan *et al.*[9] have pointed out an interesting possibility. At very low temperature the probability of adsorption upon collision can be so small that it would take appreciable time for the surface atomic layer of hydrogen to reach equilibrium with the free gas. This time might be long enough to allow useful experiments before surface relaxation and recombination became appreciable.

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Note added.—During the preparation of this manuscript we received a preprint by Sprik, Walraven, and Silvera[10] also reporting observation of a three-body recombination process.

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[a] Present address: Department of Physics, Purdue University, West Lafayette, Ind. 47907.

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