

## Temperature and Magnetic Field Dependence of Three-Body Recombination in Spin-Polarized Hydrogen

Harald F. Hess, David A. Bell, Gregory P. Kochanski, Daniel Kleppner, and Thomas J. Greytak

*Department of Physics and Center for Material Science and Engineering,  
Massachusetts Institute of Technology, Cambridge, Massachusetts 02139*

(Received 7 February 1984)

The three-body recombination rate in spin-polarized hydrogen has been measured at temperatures from 0.25 to 0.60 K in magnetic fields from 5 to 9 T. In contrast to theoretical expectation, the three-body rate is a weakly decreasing function of field in this region. At 7.6 T the gas and surface three-body rate constants are  $L_g = 7.5(3) \times 10^{-39} \text{ cm}^6 \text{ s}^{-1}$  and  $L_s = 2.0(6) \times 10^{-24} \text{ cm}^4 \text{ s}^{-1}$ . The large value of  $L_s$  can account for effects previously attributed to an anomalously large surface two-body nuclear relaxation rate.

PACS numbers: 67.40.Fd, 67.70.+n, 68.10.Jy

The three-body dipole recombination process in spin-polarized hydrogen predicted by Kagan, Vartanyantz, and Shlyapnikov<sup>1</sup> and recently observed experimentally<sup>2,3</sup> places severe limits on the densities which can be achieved in this quantum gas. We have measured how this process varies with temperature and magnetic field. The temperature dependence permits the gas and surface three-body rate constants to be determined separately. The magnetic field dependence of these constants is found to differ from that predicted theoretically. A troublesome problem to the entire study of spin-polarized hydrogen is that the two-body nuclear relaxation rate on a pure <sup>4</sup>He surface reported by several groups, including our own, is about a factor of 50 larger than theories predict. We find that the surface three-body recombination can explain effects which previous work may have erroneously attributed to surface two-body relaxation.

The experimental technique is identical to that described earlier<sup>2</sup>; however, we have achieved an improved signal-to-noise ratio which allows more precise data to be taken over a wider range of temperatures and densities. Electron- and nuclear-polarized atomic hydrogen is compressed into a pancake-shaped cell by a piston. The flat <sup>4</sup>He-coated walls of the cell are perpendicular to the magnetic field. For these experiments the separation was reduced to  $7.5(10) \times 10^{-3} \text{ cm}$ . After the compression, the pressure and temperature of the gas are recorded as the hydrogen sample decays at constant volume. The ideal gas law is then used to compute the density  $n(t)$  and its time derivative  $\dot{n}(t)$ . Measurements were carried out between 0.25 and 0.60 K. Thermal explosions<sup>3,4</sup> limit the compressions at temperatures above this region; heating problems complicate the interpretation of results below it. The highest density achieved in these experiments was  $4.5 \times 10^{18} \text{ atoms cm}^{-3}$  at a

temperature of 0.57 K.

Under our experimental conditions the observed three-body recombination rate constant  $L$  is related to the gas and surface rate constants  $L_g$  and  $L_s$  by

$$L = L_g + (A/V)L_s(\sigma/n)^3, \quad (1)$$

where  $A/V$  is the area to volume ratio for the cell and  $\sigma$  is the surface density of adsorbed hydrogen atoms. A similar expression relates the two-body relaxation rate constants

$$G = G_g + (A/V)G_s(\sigma/n)^2. \quad (2)$$

For an isothermal decay under ideal experimental conditions

$$-\dot{n}/n^2 = 2G + nL. \quad (3)$$

The intercept at  $n = 0$  on such a plot gives  $2G$  and the slope gives  $L$ . Figure 1 shows experimental measurements of  $-\dot{n}/n^2$  as a function of  $n$  for two different temperatures. Our data differ from Eq. (3) in three respects, none of which significantly impairs the ability to extract precise values of  $G$  and  $L$ . At our higher temperatures, the gas does not achieve a high nuclear polarization by the time the compression takes place. This causes  $-\dot{n}/n^2$  to lie above Eq. (3) early in the decay (i.e., at large values of  $n$ ) as the steady-state value of the polarization is established by the recombination of excess  $\alpha$ -state atoms (Fig. 1,  $T = 0.60 \text{ K}$ ). At lower temperatures and high density, the heating of the helium film relative to the copper cell decreases the density of adsorbed hydrogen atoms. As a result the apparent  $G$  and  $L$  vary during a decay, rising to their equilibrium values as  $n$  decreases (Fig. 1,  $T = 0.30 \text{ K}$ ). Finally, early in the experimental run there was a noticeable one-body term in the decay. We attribute it to a one-body surface relaxation due to impurities on the cell walls since it diminished in magnitude after many decays, presumably because

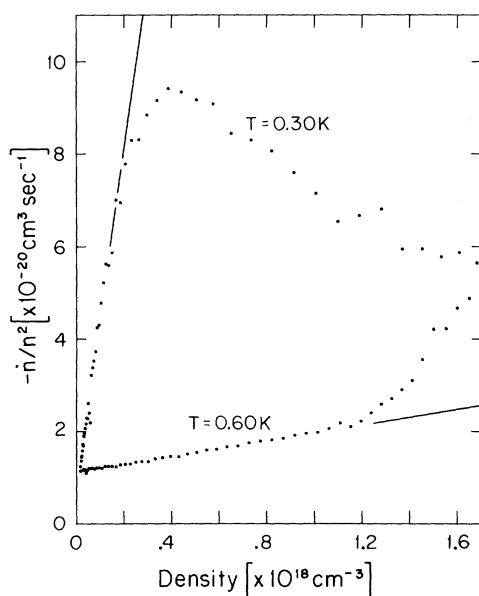


FIG. 1. Data showing the three-body nature of the density decay rates at 7.6 T. The intercept at  $n = 0$  is  $2G$ ; the slope near  $n = 0$  is  $L$ . The solid lines show fits by Eq. (3) with  $L$  and  $G$  held constant.

of the buildup of a microscopic layer of  $H_2$  in the cell. We have modeled these three effects and have been able to achieve excellent fits to the experimental decays. In all cases where the one-body term is small, the values of  $G$  and  $L$  thus obtained are identical to those that would be obtained by fitting Eq. (3) with constant  $L$  and  $G$  (Fig. 1, solid lines) to the low-density part of the  $-\dot{n}/n^2$  data.<sup>5</sup>

Figure 2 shows  $L$  as a function of temperature at a magnetic field of 7.6 T. For the conditions under which  $L$  was determined the classical adsorption isotherm  $\sigma = n\Lambda(T)\exp(E_b/kT)$  pertains. Here  $\Lambda(T)$  is the thermal de Broglie wavelength and  $E_b$  is the binding energy of atomic hydrogen to liquid  $^4\text{He}$ . Assuming that  $L_g$  and  $L_s$  are independent of temperature, we have used this expression for  $\sigma$  to do a least-mean-squares fit of Eq. (1) to the data in Fig. 2 and obtain  $E_b$ ,  $L_g$ , and  $L_s$ . We find that  $E_b = 0.99(2)$  K. This should be compared to the values found previously: 1.15(5) from the temperature dependence of the zero-field hyperfine resonance,<sup>6</sup> and 0.89(7),<sup>7</sup> 1.01(6),<sup>8</sup> and 1.06(4)<sup>9</sup> from the two-body surface recombination.

From the fit to the 7.6-T data in Fig. 2 we find  $L_g = 7.5(3) \times 10^{-39} \text{ cm}^6 \text{ s}^{-1}$ . This is about half the value  $16 \times 10^{-39}$  estimated by Kagan, Vartanyantz, and Shlyapnikov<sup>1</sup> at  $B = 10$  T.<sup>10</sup> It is twice the value  $4(1) \times 10^{-39}$  measured by Sprik, Walraven, and Silvera<sup>3</sup> in compressed bubbles at  $T = 0.75$  K

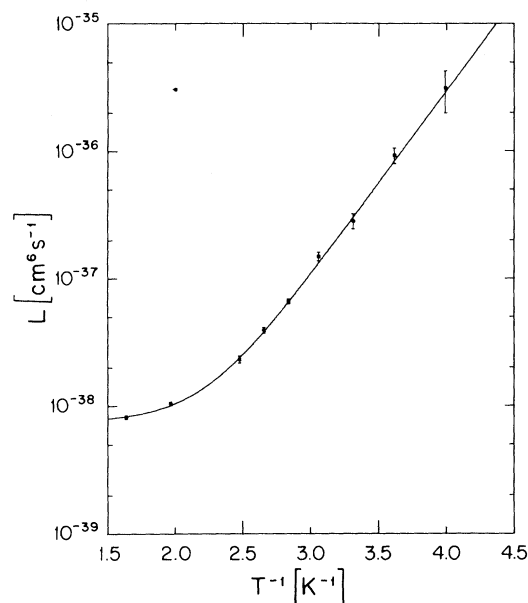


FIG. 2. Temperature dependence of the three-body recombination rate constant  $L$  at a magnetic field of 7.6 T. The solid line is the result of fitting the data by Eq. (1).

and  $B = 9.8$  T. We also find  $L_s = 2.0(6) \times 10^{-24} \text{ cm}^4 \text{ s}^{-1}$  where the principal uncertainty arises from the tradeoff between  $E_b$  and  $L_s$  in the fitting procedure. Using these values we find  $(L_g/L_s)^{1/2} = 6(1) \times 10^{-8} \text{ cm}$ . Kagan *et al.*<sup>11</sup> predict a value of about  $4 \times 10^{-8}$  for this ratio.

Figure 3 shows the magnetic field dependence of  $L$  at 0.40 K.  $L$  decreases weakly with field between 5 and 9 T. At this temperature  $L$  is dominated by the surface contribution. Data taken at 0.5 K, where the surface contribution is reduced to about 29%, show a similar insensitivity to field in this region. This is at variance with the prediction of Kagan, Vartanyantz, and Shlyapnikov<sup>1</sup> who calculated that the three-body dipole contribution to  $L$  should increase by a factor of 3 between 5 and 10 T. They also considered a three-body recombination process arising from the exchange interaction. We have evaluated this contribution to  $L$  in our experiments and found that it is negligible as a result of the high degree of nuclear polarization.

To our knowledge, the only other three-body process which could contribute to the decay of double-polarized hydrogen is a third-order contribution to the nuclear relaxation discussed by Lagedijk.<sup>12</sup> The decay rate for such a process depends on the effective perpendicular moment of the atoms and would therefore vary with field as  $(1 + 16.7/B)^2$ , where  $B$  is the magnetic field in tes-

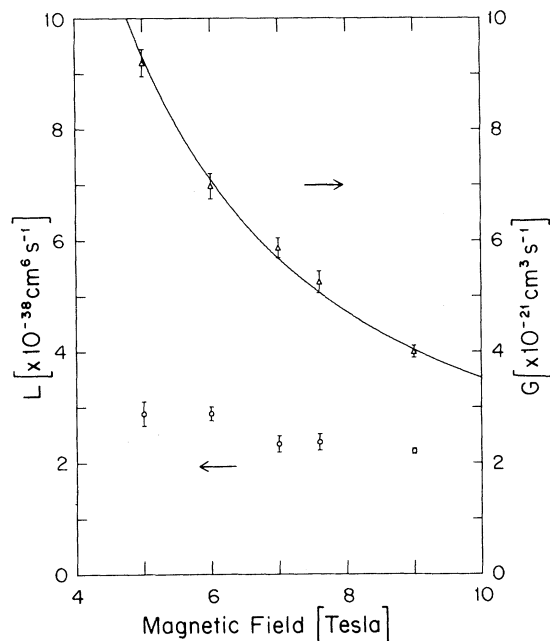


FIG. 3. Magnetic field dependence of the three-body recombination rate constant  $L$  (circles) and the two-body relaxation rate constant  $G$  (triangles) at a temperature of 0.40 K. The solid curve is the field dependence of the square of the effective perpendicular moment fitted to the data for  $G$ .

la. The data for  $G$  in Fig. 3 exhibit this dependence; the data for  $L$  do not. Furthermore, the rate calculated from the theory is far too small to account for our observations. Not only is the third-order contribution much smaller in magnitude than the second-order part at our densities, but its effect is further diminished by a dependence on the orientation of the magnetic field which drives it to zero for surfaces oriented perpendicular to the field.

Our measurements are consistent with a  $G$  which is a weakly increasing function of temperature throughout the region we studied. At 0.5 K and 7.6 T we find  $G = 5.7(3) \times 10^{-21} \text{ cm}^3 \text{ s}^{-1}$ . This is consistent with previously measured values of the bulk two-body relaxation constant<sup>8,9,13</sup> and in good agreement with the theoretical calculations of Ahn, Eijnde, and Verhaar.<sup>14</sup> Below 0.4 K our ability to determine  $G$  precisely is impaired by the dominance of the three-body surface recombination, a possible one-body term, and the finite signal-to-noise ratio. We feel that a combination of these effects led us to the erroneous conclusion that we were seeing a surface contribution to  $G$  in our first experiments with this apparatus.<sup>2</sup> Our present measurements allow us to put an upper limit on the surface two-body relaxation constant:  $G_s < 2 \times 10^{-13} \text{ cm}^2 \text{ s}^{-1}$ . This is

about a factor of 5 smaller than values of  $G_s$  reported in geometries where a significant fraction of the wall area is oriented parallel to the magnetic field.<sup>8,9,13</sup> We do not believe this is evidence for an angular anisotropy in the surface two-body relaxation rate; rather, as explained below we now believe that the previous measurements of  $G_s$  may all be in error.

We have used the values of  $L$  measured here to reexamine our earlier results on the decay of double-polarized hydrogen.<sup>8</sup> In those experiments we attribute the relatively temperature-independent decay rate at high temperatures to nuclear relaxation in the gas ( $G_g$ ) and the strongly temperature-dependent decay rate at low temperatures to relaxation on the surface ( $G_s$ ). The value of  $G_g$  agreed well with theoretical predictions,<sup>14</sup> yet  $G_s$  was about fifty times larger than the theoretical estimates.<sup>14</sup> Subsequent experiments in other laboratories<sup>9,13</sup> arrived at similar conclusions. Computations now show that surface three-body recombination alone can completely account for the low-temperature decay rates in our earlier experiments.<sup>15</sup> For a given trace, deviations from the two-body decay attributed to incomplete nuclear polarization could in fact have been the three-body form for the decay. When studying the temperature dependence, the quantity  $-\dot{n}/n^2$  was computed (but not plotted over a wide range of  $n$  as we have done here). At the low temperatures it was assumed to have a contribution  $G_s(\sigma/n)^2$  which agreed with the observed temperature dependence  $\exp(2E_b/kT)$ . If the effect was caused by three-body recombination, the contribution to  $-\dot{n}/n^2$  would be  $nL_s(\sigma/n)^3 = \sigma L_s(\sigma/n)^2$ . The gas densities at which " $G$ " was measured decreased with decreasing temperature. We have determined that our reported decay rates were analyzed at roughly constant surface density  $\sigma$ ,<sup>16</sup> so that this contribution to  $-\dot{n}/n^2$  would also be proportional to  $\exp(2E_b/kT)$ , and thus appear to display the signature of two-body surface relaxation. We conclude that the large experimental values formerly attributed to  $G_s$  are in error and that the effect was due instead to surface three-body recombination.

We would like to thank Lois Pollack and Dr. Saps Buchman for assistance in carrying out these experiments. We have benefited from correspondence with Dr. Yu. Kagan and Dr. G. V. Shlyapnikov. One of us (G.P.K.) would like to acknowledge the support of the AT&T Bell Laboratories Ph.D. Scholarship Program. This work was supported by the National Science Foundation through Grants No. DMR-8304888 and No. DMR-8119295.

<sup>1</sup>Yu. Kagan, I. A. Vartanyantz, and G. V. Shlyapnikov, *Zh. Eksp. Teor. Fiz.* **81**, 1113 (1981) [*Sov. Phys. JETP* **54**, 590 (1981)].

<sup>2</sup>H. F. Hess, D. A. Bell, G. P. Kochanski, R. W. Cline, D. Kleppner, and T. J. Greytak, *Phys. Rev. Lett.* **51**, 483 (1983).

<sup>3</sup>R. Sprik, J. T. M. Walraven, and I. F. Silvera, *Phys. Rev. Lett.* **51**, 479, 942 (1983).

<sup>4</sup>Yu. Kagan, G. V. Shlyapnikov, and I. A. Vartanyantz, to be published.

<sup>5</sup>When the steady-state nuclear polarization is less than 100%, the  $n = 0$  intercept of  $-\dot{n}/n^2$  is less than  $2G$ . In our experiments this correction amounts to 12% at  $T = 0.60$  K, 5% at  $T = 0.50$  K, and is negligible at lower temperatures.

<sup>6</sup>M. Morrow, R. Jochemsen, A. J. Berlinsky, and W. N. Hardy, *Phys. Rev. Lett.* **46**, 195 (1981), and **47**, 455 (1981).

<sup>7</sup>A. P. M. Matthey, J. T. M. Walraven, and I. F. Silvera, *Phys. Rev. Lett.* **47**, 668 (1981).

<sup>8</sup>R. W. Cline, T. J. Greytak, and D. Kleppner, *Phys. Rev. Lett.* **47**, 1195 (1981).

<sup>9</sup>B. Yurke, J. S. Denker, B. R. Johnson, N. Bigelow,

L. P. Levy, D. M. Lee, and J. H. Freed, *Phys. Rev. Lett.* **50**, 1137 (1983).

<sup>10</sup>This value is obtained by multiplying the number of three-body events per atom per second [expression (8.11) of Ref. 1 corrected for 100% nuclear polarization] by the mean number of atoms eventually eliminated per event ( $\approx 3.9$  atoms).

<sup>11</sup>Yu. Kagan, G. V. Shlyapnikov, I. A. Vartanyantz, and N. A. Glukhov, *Pis'ma Zh. Eksp. Teor. Fiz.* **35**, 386 (1982) [*JETP Lett.* **35**, 477 (1982)]. Equation (11) contains a misprint:  $l$  should be replaced by  $l^2$ .

<sup>12</sup>A. Legendijk, *Phys. Rev. B* **25**, 2054 (1982).

<sup>13</sup>R. Sprik, J. T. M. Walraven, G. H. van Yperen, and I. F. Silvera, *Phys. Rev. Lett.* **49**, 153 (1982).

<sup>14</sup>R. M. C. Ahn, J. P. H. W. v. d. Eijnde, and B. J. Verhaar, *Phys. Rev. B* **27**, 5424 (1983), and references therein.

<sup>15</sup>Such a possibility was raised independently by A. E. Ruckenstein, *J. Low Temp. Phys.* (to be published).

<sup>16</sup>This probably arose because the initial density for a trace was determined by a balance between a constant filling flux and a decay rate governed by the surface density.