RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 31, NUMBER 11 1 JUNE 1985

Electron-spin-resonance studies of spin-polarized hydrogen on the surface of liquid ^{4}He

M. W. Reynolds, I. Shinkoda, W. N. Hardy, and A. J. Berlinsky
Department of Physics, University of British Columbia, Vancouver, British Columbia, Canada V6T 2A6

F. Bridges
Physics Board of Studies, University of California, Santa Cruz, California 95064

B. W. Statt
Laboratory of Atomic and Solid State Physics, Cornell University, Ithaca, New York 14853

(Received 14 December 1984)

Electron-spin resonance at 114 GHz is used to measure the three-body recombination rate (L_3) and the two-body surface relaxation rate (G_{2s}) for doubly spin-polarized H between 100 and 200 mK. Our results for L_3 confirm and extend those of the MIT group (Hess *et al*). We find that G_{2s} is much smaller than previously reported and in reasonable agreement with theory. Around 100 mK, we also observe sidebands due to surface atoms whose frequency is shifted by dipole-dipole interactions.

In an earlier paper,[1] we demonstrated that electron-spin-resonance measurements of the density of H atoms in each of the two lower hyperfine states as a function of time could be used to obtain accurate values of the recombination-rate coefficients of a gas of spin-polarized atomic hydrogen. We also showed that the ubiquitous and troublesome one-body relaxation due to magnetic impurities in the substrate could be reduced by depositing a coating of solid H_2 on the surface. Here, we report an extension of our measurements to much lower temperature using a thick surface coating of solid H_2 which renders the one-body rate G_1 negligible. Under these conditions, nuclear polarization was essentially complete,[2] i.e., all but a negligible fraction of the atoms were in the state $|b\rangle = |\downarrow\uparrow\rangle$ where $\uparrow, \downarrow$ ($\uparrow, \uparrow$) represent the projection of the electron (proton) spin. The dominant recombination processes were then three-body recombination, with rate constant L_3, and two-body surface relaxation G_{2s} to the $|a\rangle$ state followed by effectively instantaneous recombination. Experiments under similar conditions have previously been reported by Hess *et al.*[3] and Bell *et al.*,[4] and we have confirmed and extended their measurements to lower temperature.

The very small value of G_1 in these measurements has allowed us to separate, for the first time, the smaller effect of two-body surface relaxation from the three-body recombination mechanism which is dominant at low T. We find that G_{2s} is about 10 times smaller than the value inferred from previous studies (Cline, Greytak, and Kleppner;[5] Sprik, Walraven, van Yperen, and Silvera;[6] and Yurke *et al.*[7]) and that it is now consistent with theoretical estimates[8–10] for a rough surface (i.e., the "muffin-tin" model of Refs. 6 and 9). It has been suggested by Hess *et al.*[3] that values of G_{2s} obtained in Refs. 5–7 are incorrect because the effect of three-body recombination was ignored in those analyses. Our results lend support to this suggestion.

An interesting and novel feature of our lowest temperature ($T \approx 100$ mK) spectra is the appearance of sidebands which we identify as arising from atoms on the end and side surfaces of the cylindrical electron-spin-resonance (ESR) cavity and which are shifted by the H-H dipole-dipole interaction. Figure 1 shows four of these sideband spectra for a single decay. To within the accuracy of the data, the shifts of these sidebands are proportional to the density of atoms on the surface. These data are discussed in more detail below.

The apparatus used for these measurements was essential-

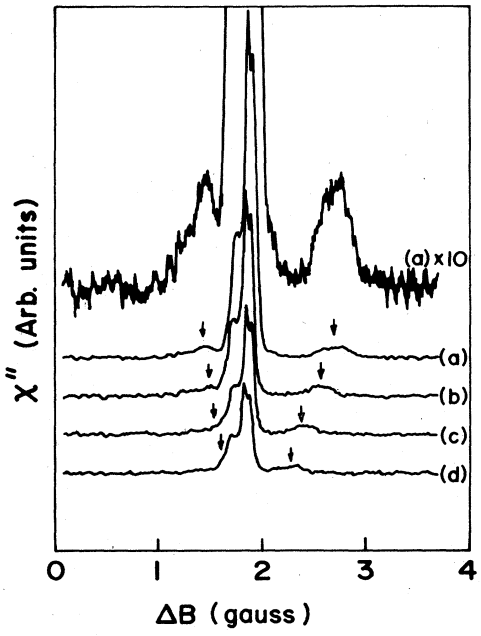

FIG. 1. Microwave absorption χ'' vs magnetic field for a sample of electron and nuclear spin-polarized atomic H gas at $T \approx 100$ mK. Curve (a) corresponds to a bulk density of 1.9×10^{14} cm^{-3}, and the integrated intensity of the right-hand peak implies a surface density of 5×10^{11} cm^{-2}. Curves (b), (c), and (d) show how the side peaks move inward with decreasing atom density.

ly as described in Ref. 1. The cell was coated with solid H_2 by running the hydrogen source at a high rate for about 12 h with simultaneous microwave irradiation so as to induce recombination in the cavity. By monitoring the shift of the cavity resonance, we were able to estimate that the average thickness of the H_2 layer was about a micron. The resulting one-body surface relaxation rate ($G_1 \approx 10^{-5}$ s^{-1} at $T = 0.4$ K) was at least a factor of 10 smaller than the "small G_1" data of Ref. 1.

The data for L_3 and G_{2s} presented in this paper were obtained by measuring the real part of the microwave reflection coefficient. In the same experimental run, but after the data on L_3 and G_{2s} had been taken, the apparatus was modified to allow simultaneous detection of both real and imaginary parts of the reflection coefficient. This allowed us to accurately determine and tabulate the nonlinear relationship between the integrated χ'' and the integrated real part of the reflection coefficient, under conditions of fixed Q, cavity coupling, and line shape. Using such a table, reliable densities were obtained even for traces where the nonlinear effects were large. A correction was also made for the fact that the fraction of atoms excited per sweep (typically $\langle 10^{-3}$) depends on the reflection coefficient and hence on the density of atoms in the cavity.

For a doubly spin-polarized sample (b atoms only), the rate equation for the density of atoms n may be written as

$$\dot{n} = -2G_1 n - 2G_2 n^2 - L_3 n^3 - 2n \sum_i F_i \delta(t - t_i) \ , \qquad (1)$$

where G_1 and G_2 are, respectively, the one- and two-body relaxation rate constants and L_3 is the three-body recombination-rate constant. G_2 and L_3 may contain both bulk and surface contributions, although for the temperature range of our measurements, 100 mK $< T <$ 200 mK, the bulk contributions are negligible. F_i is the fraction of atoms excited into the $|c\rangle$ state during the sweep at time t_i.

Each decay curve was fitted to Eq. (1) with four parameters: $n(0)$, G_1, G_2, and L_3. In order to interpret the parameters G_1, G_2, and L_3 as rate constants, the temperature rise due to recombination must be small. Estimates showed that this temperature rise should be negligible, mainly due to the low atom densities ($n \leq 5 \times 10^{14}$ cm^{-3}) and the small size of the cavity. This expectation was directly confirmed by the absence of any dependence of the binding energy E_B on the surface density in the measurement of E_B described below.

Results for the two-body relaxation rate are shown by the squares in Fig. 2. In the same figure, we have plotted values of G_2 from Refs. 5–7 which have been adjusted to correspond to our A/V ratio (30 cm^{-1}) and field (4.1 T). This allows a comparison of the various surface relaxation rates which is independent of any assumption about the value of the binding energy E_B. Our surface relaxation rates are seen to be an order of magnitude below the earlier results. The reason for this difference is simply that much of the effect attributed to G_2 in earlier work is accounted for by three-body recombination in our analysis. Our ability to simulataneously determine G_2 and L_3 results from the very small value of G_1 achieved by coating the cavity walls with solid H_2.

The solid curve in Fig. 2 corresponds to the theory of Ref. 10. G_2 was calculated from the expression

$$G_2 = G_2^{\text{bulk}} + (A/V)\Lambda^2 e^{2E_B/kT} \langle G_{2s} \rangle_\theta \ , \qquad (2)$$

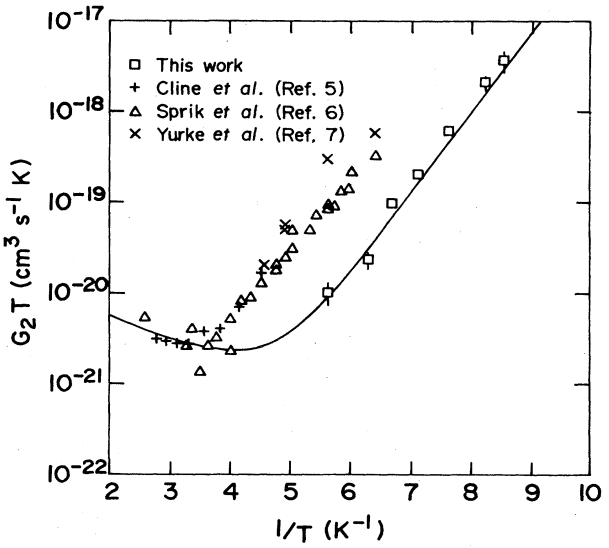

FIG. 2. Temperature dependence of the two-body relaxation rate. The data of Refs. 5–7 have been adjusted to correspond to the A/V ratio and field of our experiment. The solid curve is obtained from Eq. (2) using the theory of Ref. 10.

where

$$G_2^{\text{bulk}} T^{-1/2} = 1.68 \times 10^{-20} \text{ cm}^3\text{s}^{-1}\text{ K}^{-1/2} \ ,$$

$E_B = 1.00$ K, and $\langle G_{2s} \rangle_\theta \approx 1 \times 10^{-13}$ cm^{2}s^{-1} was obtained by averaging Statt's results for $G_{2s}(\theta)$ over the "muffin-tin" surface profile of Refs. 6 and 9, including in this average the ratio of microscopic to macroscopic surface area. The excellent agreement between this theoretical curve and our data demonstrates that at least most of the large discrepancy between theory and experiment for the surface relaxation rate has now been resolved.

Our data for the three-body recombination-rate L_3 vs $1/T$ are represented by the squares in Fig. 3. Also shown are the data of Hess *et al.*[3] and Bell *et al.*[4] which have been adjusted to correspond to our A/V ratio and magnetic field. For the field dependence, we have used the experimental observation of Ref. 4, $dL_3/dB = (-0.1\ \text{T}^{-1})L_3$. Figure 3 shows that there is good agreement between our measurements of L_3 and the extrapolated fit of Ref. 3.

At the lowest temperatures studied, $T \approx 100$ mK, the absorption line shapes exhibited an obvious density dependence as sidebands appeared asymmetrically above and below the main line at high densities ($n_{\max} \lesssim 10^{15}$ cm^{-3}) and moved inward with decreasing density (see arrows in Fig. 1). We have tentatively identified these sidebands as spectra of atoms on the walls of the cavity, the high-field line coming from atoms on the top and bottom surfaces which are normal to the magnetic field, and the low-field line from atoms on the side wall of the cylindrical cavity. The theory of this splitting is similar to that of the shift of the nuclear-magnetic-resonance (NMR) frequency which is treated briefly by Ruckenstein[11] and in somewhat more detail by Statt.[10] The shift in field of the ESR line may be written as

$$\delta B = \frac{3\pi\hbar\gamma_e n_s}{2r_0} P_2(\cos\theta) \ , \qquad (3)$$

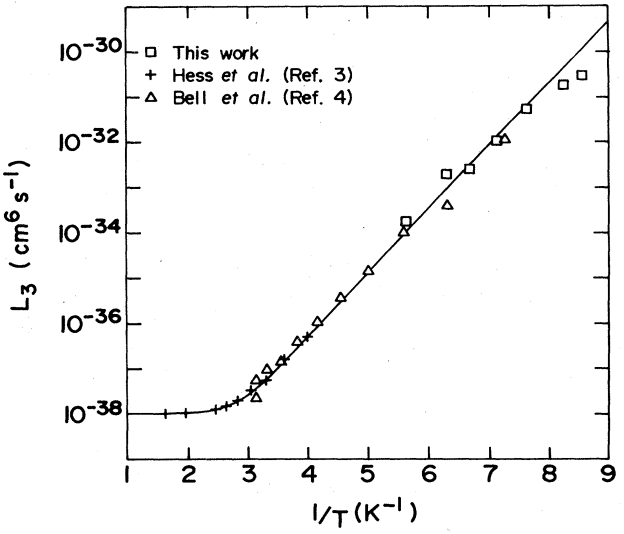

FIG. 3. Temperature dependence of the three-body recombination-rate constant L_3. The solid line is the fit from Ref. 3, adjusted in the same way as the data and extrapolated to lower temperature.

where n_s is the surface density of atoms, P_2 is a Legendre polynomial, and θ is the angle between the magnetic field and the normal to the surface. The factor $1/r_0$ is defined as

$$\frac{1}{r_0} = -\frac{A}{2\pi}\left\langle \frac{3\cos^2\theta' - 1}{r^3} \right\rangle , \tag{4}$$

where r is the distance between two H atoms on a surface of area A, θ' the angle between the vector joining these two atoms and the normal to the surface, and $\langle \ldots \rangle$ is a thermal average over triplet-scattering states. $1/r_0$ is related to the quantity $\langle R^t \rangle$, calculated in Statt's thesis, by $1/r_0 = -8(\pi/5)^{1/2}\langle R^t \rangle$. For $T = 100$ mK, $\langle R^t \rangle = -1.5 \times 10^{-2}$ Å^{-1}.

An important consequence of the shift of the surface-atom resonance is that the integrated intensity of a peak yields the surface-atom density n_s without requiring a knowledge of either the temperature or the binding energy E_B. If the temperature is known, then even a rough measurement of n_s gives a fairly accurate value of E_B. A dependence of E_B on n_s indicates the presence of heating due to recombination. The traces in Fig. 1 give $E_B = 1.00 \pm 0.05$ K with no observable heating effects. In addition, the *shift* of the sideband is related to the surface density n_s through Eq. (3). Using the results from Statt[10] for $\langle R^t \rangle$, the surface density of 5×10^{11} cm^{-2} in the top trace implies a splitting of 0.4 G, whereas the experimental value is about 0.8 G.

Experiments are presently underway to study the surface peaks in more detail, in particular by shimming the magnetic field to narrow the central peak due to atoms in the gas. Better measurements of the shape of the surface peaks will allow us to determine the surface profile through the angular factor $P_2(\cos\theta)$ in Eq. (3) and hence allow an improved comparison of theory and experiment for G_{2s}. Better measurements should also allow a much more accurate determination of E_B. At lower temperatures and higher coverages, this method could be used to study the equation of state of the surface gas as it starts to become degenerate.

The authors would like to thank R. W. Cline for many helpful discussions and suggestions. This work was supported by grants from the Natural Sciences and Engineering Research Council of Canada.

[1]B. W. Statt, A. J. Berlinsky, and W. N. Hardy, Phys. Rev. B **31**, 3169 (1985).

[2]For a recent review of spin-polarized hydrogen in general, and in particular of the T_1 bottleneck which gives rise to nuclear polarization, see T. J. Greytak and D. Kleppner, in *New Trends in Atomic Physics,* edited by G. Grynberg and R. Stora (North-Holland, New York, 1984), Vol. II, p. 1129.

[3]H. F. Hess, D. A. Bell, G. P. Kochanski, D. Kleppner, and T. J. Greytak, Phys. Rev. Lett. **52**, 1520 (1984).

[4]D. A. Bell, G. P. Kochanski, L. Pollack, H. F. Hess, D. Kleppner, and T. J. Greytak, in *Proceedings of the 17th International Conference on Low Temperature Physics,* edited by U. Eckern, A. Schmid, W. Weber, and H. Wühl (North-Holland, Amsterdam, 1984), pp. 449 and 450; D. A. Bell, G. P. Kochanski, T. J. Greytak, and D. Kleppner, *ibid.,* pp. 541 and 542.

[5]R. W. Cline, T. J. Greytak, and D. Kleppner, Phys. Rev. Lett. **47**, 1195 (1981).

[6]R. Sprik, J. T. M. Walraven, G. H. van Yperen, and I. F. Silvera, Phys. Rev. Lett. **49**, 153 (1982).

[7]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).

[8]A. Lagendijk, Phys. Rev. B **25**, 2054 (1982); A. E. Ruckenstein and E. D. Siggia, *ibid.* **25**, 6031 (1982); B. W. Statt, *ibid.* **25**, 6035 (1982).

[9]R. M. C. Ahn, J. P. H. W. van den Eijnde, C. J. Reuver, B. J. Verhaar, and I. F. Silvera, Phys. Rev. B **26**, 452 (1982).

[10]B. W. Statt, Ph.D. thesis, University of British Columbia, 1984 (unpublished). Copies may be obtained by writing to W. N. Hardy at the University of British Columbia, Vancouver.

[11]A. E. Ruckenstein, J. Low Temp. Phys. (to be published).