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Citation for published version (APA):

de Goey, L. P. H., Driessen, J. P. J., Verhaar, B. J., & Walraven, J. T. M. (1984). Surface three-body recombination in spin-polarized atomic hydrogen. *Physical Review Letters*, 53(20), 1919-1922.
<https://doi.org/10.1103/PhysRevLett.53.1919>

DOI:

[10.1103/PhysRevLett.53.1919](https://doi.org/10.1103/PhysRevLett.53.1919)

Document status and date:

Published: 01/01/1984

Document Version:

Publisher's PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:

- A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
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- The final published version features the final layout of the paper including the volume, issue and page numbers.

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Surface Three-Body Recombination in Spin-Polarized Atomic Hydrogen

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 (Received 12 July 1984)

We calculate the surface dipolar recombination rate L_s for spin-polarized hydrogen adsorbed on ^4He surfaces at temperatures in the 0.2- to 0.6-K regime and for magnetic fields up to 30 T. For a magnetic field of 7.6 T normal to the surface and 0.4 K we find $L_s = 1.3(3) \times 10^{-25} \text{ cm}^4 \text{ s}^{-1}$ increasing by 10%/T in the range of experimental interest. The anisotropy with the direction of the magnetic field is considerably smaller than in the case of the surface dipolar relaxation.

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

The recent observation¹ of three-body phenomena in high-density spin-polarized hydrogen ($\text{H}\downarrow$) has focused considerable attention on a very interesting class of thresholdless recombination processes, first described by Kagan, Vartanyants, and Shlyapnikov.² Detailed understanding of these processes is of vital importance for $\text{H}\downarrow$ research as they appear to limit the highest densities that may be achieved experimentally. In a recent publication Hess and co-workers³ pointed out that effects previously attributed to an anomalously large surface two-body nuclear relaxation rate⁴ could be accounted for by a surface analog of the Kagan process. In their analysis the surface rate was estimated by a scaling argument taken from Ref. 2.

We took up this interesting suggestion and present here the first detailed calculation of the three-body surface recombination rate L_s . We analyze the nature of the Kagan dipole mechanism and discuss the differences between recombination on a ^4He surface and in the bulk. We find that the scaling argument, which results from a model in which the relative motion of the H atoms on the surface is assumed to be identical to that in the volume, is not supported by detailed theory. It leads to an overestimate of the surface rate by an order of magnitude. We calculate $L_s = 1.3(3) \times 10^{-25} \text{ cm}^4 \text{ s}^{-1}$ for a magnetic field $B = 7.6$ T normal to the surface and temperature $T = 0.4$ K, to be compared² with an experimental value $L_s = 2.0(6) \times 10^{-24} \text{ cm}^4 \text{ s}^{-1}$ obtained³ at the same field. In the range of experimental interest our results show an increase of the rate of recombination with growing field although this trend is weaker than theory predicts for the bulk process. Experiments show a decreasing behavior for growing fields.^{3,5,6} The anisotropy of the Kagan mechanism is found to be

less than that of the two-body surface dipolar relaxation.⁷ This feature is in common with a very recent experimental analysis of the surface rates by Bell *et al.*⁸ but seems to contradict earlier low-temperature results obtained by Sprik *et al.*⁶ using ^3He surfaces. We point out that in particular the large difference in absolute value indicates that the existing discrepancy⁴ between theory and experimentally observed decay rates remains unresolved. We also calculated the bulk dipolar recombination process and find a rate which at 10 T is in agreement with results obtained by Kagan, Vartanyants, and Shlyapnikov,² although our field dependence is slightly weaker. Our value is $L_g = 8.5 \times 10^{-39} \text{ cm}^6 \text{ s}^{-1}$ ($B = 10$ T and $T \rightarrow 0$).

At low temperatures ($T \leq 1$ K) the available number of recombination channels for a system of H atoms is vastly reduced. Resonance recombination, dominant at room temperature, may be excluded entirely as the energies of the resonances are too elevated to permit thermal population.⁹ The first description of a low-temperature recombination mechanism for H was given by Greben, Thomas, and Berlinsky.⁹ This exchange-recombination process requires a collision between a pair of H atoms with singlet character in their initial state. A third body is required to conserve energy and momentum in the process. Besides H other atoms or surfaces may be effective as third body. One of the most fascinating features of the $\text{H}\downarrow$ system is that the above mechanism implies (in combination with slow magnetic relaxation in high fields) preferential recombination and depletion of the "mixed" a state (a , b , c , and d are the hyperfine states in order of increasing energy). This process results in a gas of atoms in the "pure" b state, double-polarized hydrogen ($\text{H}\downarrow\uparrow$) in which both

electron and proton spins are polarized.

The Kagan process is the only recombination mechanism presented in the literature which may limit the stability of $H\downarrow\uparrow$. This process involves a combined relaxation-recombination mechanism which is thresholdless and in which the dipolar interaction between the electronic spins of the b -state atoms causes the spin flip required for recombination. We distinguish between single- and double-spin-flip processes and will show that the double-spin-flip process is dominant at low fields, whereas it may be suppressed entirely by application of a field $B \gtrsim 24$ T.

If we divide the triple of atoms in a bbb incoming state into a recombining pair (atoms 1 and 2) and a third body (atom 3), we note that one may neglect the electronic dipolar interaction between the atoms 1 and 2 as this interaction cannot cause triplet-singlet transitions. In principle the electronic-nuclear dipolar interaction may do so, but this process is believed to be much weaker. As a result only the difference in magnetic field experienced by the recombining atoms due to the third atom is effective in the recombination process. This causes the remarkable feature that even in the presence of an abundance of third bodies provided by the He surface a third H atom is required. In principle the interaction with a magnetic surface impurity may be present and may cause a similar process with a second-order character.

We write the transition amplitude f for recombination of atoms 1 and 2 as

$$f = \frac{2}{3} \frac{m_H}{2\pi\hbar^2} \langle \psi_f | V_{13}^d + V_{23}^d | \sum_P P \psi_i \rangle . \quad (1)$$

Here, m_H is the mass of the hydrogen atom, V_{ij}^d represents the dipolar interaction between atoms i and j , while the initial state $\sum_P P \psi_i$ is a symmetrized three-atom bbb state, P being a permutation operator. Following Kagan we approximate the initial state by only taking into account the spatial correlations between the atoms of the recombining pair and between the atoms interacting via the dipolar interaction. For instance, for the 13 term the initial state is written as

$$\psi_i = \phi_0(z_1)\phi_0(z_2)\phi_0(z_3)\psi_{k_{12}}^t(\vec{\rho}_{12})\psi_{k_{13}}^t(\vec{\rho}_{13})|bbb\rangle . \quad (2)$$

For each of the atoms we use a bound-state wave function⁴ $\phi_0(z) \sim z \exp(-\alpha z)$. For $\alpha = 0.2a_0^{-1}$, ϕ_0 resembles the bound-state wave function in a Stwalley-type potential reproducing the experimen-

tal adsorption energy,⁴ while for $\alpha = 0.15a_0^{-1}$ it resembles the Mantz and Edwards wave function.¹⁰ The error bar for our L_s value corresponds with these values for α . In Eq. (2) $\psi_{\vec{k}}^t$ describes the relative motion of a pair of H atoms along the surface distorted by the triplet interaction averaged over the z motion ("2½-dimensional" model⁴) and normalized with plane-wave part $\exp(i\vec{k} \cdot \vec{\rho})$. Here \vec{k} and $\vec{\rho}$ are two-dimensional momentum and position vectors. The final state ψ_f is assumed to be identical to that used by Kagan in the volume case, but expressed in cylindrical coordinates: the product of a final spin state $\sigma_f = +\frac{1}{2}$ or $-\frac{1}{2}$ of atom 3, a plane wave with three-dimensional momentum $\hbar\vec{q}_f(B, \nu, j, \sigma_f)$ for the motion of this atom relative to atoms 12, and a 12 molecular singlet state with vibrational and rotational quantum numbers νjm . In view of the rather high $H+H_2$ relative kinetic energy we neglect completely the influence of the helium surface on the final state, which at the same time reduces the expressions to a form manageable numerically. With this approximation we neglect a reduction of the available final-state phase space and a possible energy transfer to the center-of-mass motion or to the helium. These effects are estimated to be small.

We note that only ortho ($j = \text{odd}$) final states are allowed, as the proton spins are unaffected by the process. Furthermore, we note that in the matrix element of Eq. (1) the spatial intergration is over relative coordinates. The essential difference from the volume case is the lack of translational invariance in the z direction, which causes the result to depend on the center-of-mass coordinate Z in this direction. The recombination rate is obtained by summing $|f|^2$ over final states, integrating over Z , and thermal averaging over initial momenta along the surface:

$$L_s = \sum_{\nu jm \sigma_f} \left\langle \frac{\hbar q_f}{4m_H} \int dZ \int d\hat{q}_f \left| f_{\nu jm \sigma_f}(\vec{q}_f, Z) \right|^2 \right\rangle_{\text{thermal}} . \quad (3)$$

Notice that in two dimensions a $T \rightarrow 0$ approximation cannot be made. Instead we use a low-energy logarithmic k_{12}, k_{13} dependence of f following from two-dimensional effective-range theory,¹¹ using the value $2.3a_0$ for the scattering length. It is appropriate to point out here that the same logarithmic character of f probably contributes to the failure of the above-mentioned scaling argument.

To evaluate L_s we reexpress the spin wave functions using the surface normal as the new quantization axis. If we represent the transfer of angular

momentum from the spin system to the orbital system along this axis by $\mu\hbar$, we find an expression for L_s as an incoherent sum over μ :

$$L_s(\vec{B}) = \sum_{\sigma_f} \sum_{\mu=-2}^2 L_s^{|\mu|\sigma_f}(B) [d_{\sigma_f+3/2,\mu}^2(\theta)]^2 = \sum_{\sigma_f} \sum_{n=0,2,4} A_{n\sigma_f}(B) P_n(\cos\theta), \quad (4)$$

where the d functions are reduced Wigner functions and θ is the angle between \vec{B} and the surface normal. We note that the double-spin-flip contribution tends to dominate over the single-spin-flip one because of the relation

$$L_s^{|\mu|,+1/2}(B) = 4L_s^{|\mu|,-1/2}(2B).$$

For the dominant states $v=14$, $j=3$ and $j=1$ (all other molecular states contribute at a negligible level), and various $|\mu|$ and $|m|$ combinations, we calculated the behavior of $|f|^2$ as a function of Z and of the projection $q_{f\parallel}$ of \vec{q}_f along the surface. For $q_{f\parallel} \rightarrow q_f$ all $|f|^2$ surfaces show a strong decrease. Physically this is due to the absence of high relative momenta along the surface in the initial state. Essentially this is the same feature which gives rise to the strong B dependence of volume recombination. Because this argument applies in the surface case only for two coordinate directions instead of three, the B dependence of L_s is weaker

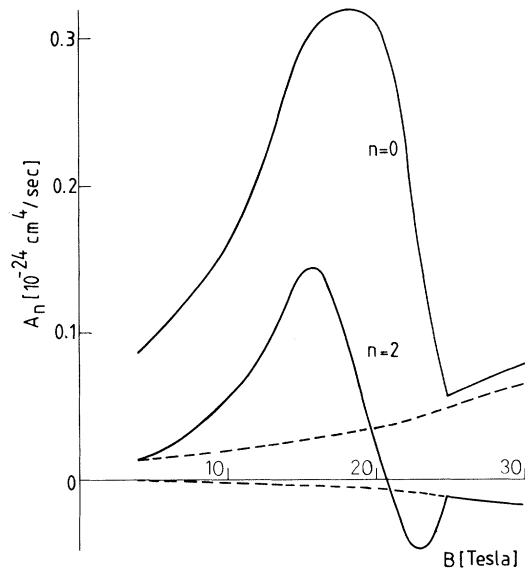


FIG. 1. Coefficients $A_n = \sum A_{n\sigma}$ of Legendre polynomials describing the surface recombination rate L_s for $\alpha=0.2a_0^{-1}$ at $T=0.4$ K as a function of magnitude and direction of magnetic field (solid curves). The coefficient A_4 is omitted (magnitude $< 0.014 \times 10^{-24} \text{ cm}^4 \text{ s}^{-1}$). The broken curves represent single-spin-flip contributions.

than that of L_g . In Fig. 1 we show the functions $A_{n\sigma}(B)$ for $T=0.4$ K. The coefficients $A_{0\sigma}$ represent the recombination rate averaged over the direction of \vec{B} . The coefficients $A_{2\sigma}$ and $A_{4\sigma}$ express the anisotropy as a function of the field direction. The A_4 coefficients are negligibly small, whereas the A_2 are small for the single-spin-flip contribution ($\sigma_f = -\frac{1}{2}$) and at most half the A_0 value for the double-spin-flip process ($\sigma_f = +\frac{1}{2}$).

Although the absence of a strong anisotropy is common with experimental indications,⁸ both the B dependence and the absolute magnitude of L_s seem to be at variance with the experimental data, although it would be desirable to extend the measurements to the double-spin-flip cutoff at 24 T. The extreme sharpness of the bends at this cutoff is due to the above-mentioned low-energy approximation and is similar to the behavior in the volume $T \rightarrow 0$ limit. We find a rate which is growing by 70% from $B=4$ to 9 T, whereas the experiments show a decrease by about the same amount. For $\theta=0$ we calculate $L_s = 1.3(3) \times 10^{-25} \text{ cm}^4 \text{ s}^{-1}$ at $B=7.6$ T. Experimentally a larger value will be observed because of the large probability for the c atom, originating from the double spin-flip process, to recombine in a subsequent collision.⁸ This implies that the experimental value given by Hess and co-workers³ has to be scaled down by approximately a factor 2×0.87 , where 0.87 is the double spin-flip fraction. This leads to $L_s = 1.1(4) \times 10^{-24} \text{ cm}^4 \text{ s}^{-1}$. An angular average reduces our theoretical L_s value by 25%. The calculated values show an increase by roughly a factor of 2 in the temperature range 0.2–0.6 K.

We stress that to evaluate the surface dipolar recombination process rather substantial approximations had to be imposed. Hence, our present results do not provide the same level of accuracy as the results for surface dipolar relaxation. However, we are convinced that refinements of the theory are unlikely to resolve the large discrepancy with experiment.

We would like to thank Joop van den Eijnde for his contributions to this work. One of the authors (J.T.M.W.) wishes to thank the University of Grenoble for hospitality during the preparation of the manuscript.

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