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Surface spin relaxation of stabilized atomic hydrogen

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We present a distorted-wave calculation of the spin relaxation of hydrogen atoms adsorbed at a superfluid helium surface, due to H-H collisions. Both the interatomic triplet potential and the dipole-dipole interactions are averaged over the bound state at the surface. The result is compared with experiment and with some alternate methods.

In recent years important progress has been made in stabilizing atomic hydrogen against molecular recombination by polarizing the electron spins.[1,2] Several papers[3,4] have been published, which throw further light on the problems to be overcome on the way to the higher densities necessary for Bose-Einstein condensation. One of these problems is a relaxation mechanism,[5] consisting of transitions between the lowest two hyperfine levels $b=\downarrow\downharpoonright$ and $a=\downarrow\upharpoonright-\epsilon\uparrow\downharpoonright$ ($\downarrow$ electron and $\downharpoonright$ proton spins) for a magnetic field $B\neq 0$, taking place in H-H collisions both in the gas phase and at the helium surface. Both the volume and surface relaxation rates, T_1^{-1}, have recently been measured by Cline *et al.*[6] The rather large value of T_1 makes the achievement of higher densities, and possibly the Bose condensed state, feasible. Due to the importance of this problem, in this paper we reexamine the theory, relaxing some of the restrictions of earlier models.

The gas-phase relaxation rate was calculated using the distorted-wave and several other approximations by Statt and Berlinsky,[5] and in the plane-wave approximation by Siggia and Ruckenstein.[7] In a forthcoming publication[8] we shall present a more accurate calculation of the differential cross section for transitions between a and b, relaxing various approximations in Ref. 5. This calculation confirms the validity of the approximations up to the 10^{-3} level of accuracy. In addition we shall go into the discrepancy of almost a factor of 2 between the relaxation times T_1 of Refs. 5 and 7.

The relaxation of H atoms at the helium surface due to binary collisions and diffusion has recently been studied by Lagendijk[9] in the plane-wave approximation, excluding in the Born integral a volume corresponding to a hard-core radius $\sigma=3.67$ Å. The collisions of the H atoms are treated as a purely two-dimensional problem, the interatomic radius vector in the magnetic dipole-dipole interactions being taken parallel to the surface. For the binary collisions the same excluded volume plane-wave approximation has been considered by Ruckenstein and Siggia.[10] The dipole-dipole interactions, however, are averaged over the uncoupled probability distributions in the z direction perpendicular to the surface. In this paper we present a more satisfactory calculation of the surface relaxation rate due to binary collisions, based on the distorted-wave approximation.

The three-dimensional nature of the H-H collision process is taken into account in a rudimentary way by z averaging both the central and the dipole-dipole interactions. This procedure can be considered as a first step in a more systematic approach in which more than one state in the z direction is included. We thus restrict ourselves to the open-channel part of the collision problem. The spin states are treated three dimensionally with the homogeneous magnetic field as a quantization axis. The choice of the coordinate frame is illustrated in Fig. 1.

As in the gas phase the relevant inelastic processes[5]

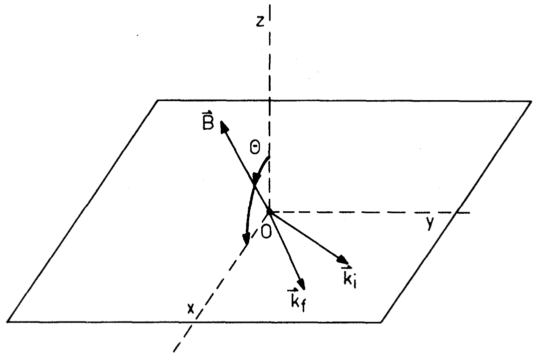

FIG. 1. Choice of coordinate frame. Initial and final relative H-H momenta $\vec{k}_i$ and $\vec{k}_f$ are indicated. Magnitudes $|\vec{k}_i|\simeq|\vec{k}_f|=k$.

are $\alpha \rightarrow \beta = bb \rightarrow ab$ and $aa \rightarrow ab$. The differential cross length λ (analog of the three-dimensional cross section) for each of these is

$$\lambda_{\alpha\rightarrow\beta}(\vec{k}_f, \vec{k}_i) = \frac{\mu^2}{2\pi\hbar^4 k_i} |T_{\alpha\rightarrow\beta}(\vec{k}_f, \vec{k}_i)|^2 \ , \tag{1}$$

where μ is the reduced mass. Taking antisymmetrization into account,[11] we have in the distorted-wave approximation

$$T_{\alpha\rightarrow\beta}(\vec{k}_f, \vec{k}_i) = \langle \chi^{(-)}_{\beta \vec{k}_f} \Psi_\beta (V - U)(1 - P_{12})(1 - P_{AB}) | \chi^{(+)}_{\alpha k_i} \Psi_\alpha \rangle \ . \tag{2}$$

The functions Ψ indicate normalized spin functions aa, etc. The partition of electrons 1,2 and protons A,B into two H atoms is identical for bra and ket vectors, say $1A + 2B$, with two-dimensional relative vector $\vec{\rho}_{1A,2B}$. Electron and proton permutation operators are indicated by P_{12} and P_{AB}, respectively. The distorted waves $\chi^{(+)}$ and $\chi^{(-)}$, in customary notation, contain plane waves $\exp(i \vec{k}_i \cdot \vec{\rho}_{1A,2B})$ and $\exp(i \vec{k}_f \cdot \vec{\rho}_{1A,2B})$, respectively, distorted by the triplet potential and the triplet spin-down Zeeman energy:

$$U = V_1 - g_S \mu_B B \ . \tag{3}$$

The potential V is the interatomic potential for the same partition:

$$V = P_0 V_0 + P_1 V_1 + H_{SS} + H_{SI} + g_S \mu_B \hbar^{-1} (\vec{S}_1 + \vec{S}_2) \cdot \vec{B} \ , \tag{4}$$

V_0 denoting the singlet potential and $P_0(P_1)$ standing for the projection operators on electron singlet (triplet) states.

The only term in Eq. (4) contributing to the relevant inelastic processes are the spin-dependent electron-electron and electron-proton dipole-dipole interactions H_{SS} and H_{SI}. Taking into account the large distance of closest approach at the collision energies involved,[5,8] their combination takes effectively the form

$$H_{SS} + H_{SI} \rightarrow \hbar^2 \gamma_e \gamma_p \left(\frac{12\pi}{5} \right)^{1/2} \times \sum_{M=\pm 1} \left[T^M_{ep} - \left(\frac{\gamma_e}{\gamma_p} \right) T^M_{ee} \right] \left\langle \frac{Y^*_{2M}(\hat{R})}{R^3} \right\rangle_Z \tag{5}$$

in the notation of Ref. 5, $\vec{R}$ standing for the three-dimensional relative vector $\vec{R}_{1A,2B}$ and M being the angular momentum transfer along the direction of $\vec{B}$. Brackets $\langle \ \rangle_Z$ indicate an average over the probability distribution $F(Z)$ for the relative distance Z of the atoms in the z direction. We approximate $F(Z)$ to be of Gaussian form. The averaging procedure is of special importance for the $m = 0 \rightarrow m' = 0$ transition between partial waves m and m' relative to the z axis. We take the value $F(0) = 0.095$ Å^{-1} from Edwards and Mantz.[12] Furthermore, we note that in the above-mentioned large-distance approximation only the $P_{12} P_{AB}$ exchange term has to be taken into account. The initial aa and bb states having $I = S$, antisymmetrization for electrons and protons requires m values to be even. We thus find

$$T_{\alpha\rightarrow\beta}(\vec{k}_f, \vec{k}_i) = \pm 6\hbar^2 \gamma_e \gamma_p \left(1 + \epsilon \frac{\gamma_e}{\gamma_p} \right) k^{-1} \sum_{m'm} \exp(i m' \phi_{\vec{k}_f} - i m \phi_{\vec{k}_i}) \times \sin\theta \left[\mp \delta_{m-m',0} \tfrac{1}{2} r_{m'm} \cos\theta + \delta_{m-m',2} \tfrac{1}{4} \bar{r}_{m'm} (1 \pm \cos\theta) + \delta_{m-m',-2} \tfrac{1}{4} \bar{r}_{m'm} (-1 \pm \cos\theta) \right] \ . \tag{6}$$

In this expression the radial integrals are defined by

$$r_{m'm} = \int_0^\infty u_{m'}(\rho) \langle 3\rho^2 R^{-5} - 2R^{-3} \rangle_Z u_m(\rho) \, d\rho \ , \quad \bar{r}_{m'm} = \int_0^\infty u_{m'}(\rho) \langle \rho^2 R^{-5} \rangle_Z u_m(\rho) \, d\rho \ , \tag{7}$$

where the u functions are normalized as cosine functions asymptotically. The upper (lower) signs in Eq. (6) refer to the $bb \rightarrow ab$ ($aa \rightarrow ab$) process, whereas m' and m run through even values only. The integrated cross lengths, averaged over initial directions of relative motion, turn out to be equal and are

$$\lambda_{\text{eff}} = 9\mu^2 \gamma_e^2 \gamma_p^2 \left(1 + \epsilon \frac{\gamma_e}{\gamma_p} \right)^2 k^{-3} \sin^2\theta \sum_{m'm} \left[\delta_{m-m',0} |r_{m'm}|^2 \cos^2\theta + \tfrac{1}{4} (\delta_{m-m',2} + \delta_{m-m',-2}) |\bar{r}_{m'm}|^2 (1 + \cos^2\theta) \right] \ . \tag{8}$$

The surface relaxation rate $T_1^{-1} = 2 n_s G_s$, where n_s is the surface coverage, is characterized by the intrinsic factor

$$G_s = (2\mu)^{-1/2} \beta \int_0^\infty dE \, \lambda_{\text{eff}}(E) \, E^{1/2} \exp(-\beta E) \ , \tag{9}$$

where $\beta = 1/k_B T$. Using Eq. (8), we find the final expression

$$G_s = G_{s,0}(T)\sin^2 2\theta + G_{s,2}(T)\sin^2\theta(1+\cos^2\theta) \quad , \tag{10}$$

in which for high fields we have approximately (in 10^{-18} $m^2 s^{-1}$)

$$G_{s,0}(T) = (0.96 - 0.82T + 0.74T^2)\left(1 + \frac{16.68}{B}\right)^2 \quad , \tag{11}$$

$$G_{s,2}(T) = (0.019 + 0.139T)\left(1 + \frac{16.68}{B}\right)^2 \quad ,$$

in the temperature range $0.2 < T < 0.4$ K (B in tesla). To derive Eq. (11) we have described the behavior of the $|r|^2$ and $|\bar{r}|^2$ quantities in a limited energy range by a quadratic and linear function of energy, respectively. Note, furthermore, that the temperature dependence of G_s, on which the analysis of Cline *et al.*[6] is based, does not seem to be consistent with our theoretical results: Instead of being dominantly proportional to T, the theoretical value of G_s shows a weak temperature dependence. Taking into account the strong temperature dependence due to the wall-binding Boltzmann factor, the experimental data in Ref. 6, however, are not inconsistent with a different dependence on temperature. We have reanalyzed the data of Cline *et al.* assuming G_s to be temperature independent and find a value of $(0.4 \pm 0.1) \times 10^{-16}$ $m^2 s^{-1}$.

Now let us compare with theory, taking $B = 11$ T and $T = 0.3$ K. The experimental geometry essentially has $\bar{\theta} = 90°$ for all surfaces, where $\bar{\theta}$ characterizes the macroscopic surface normal. In this case G_s equals $G_{s,2} = 0.38 \times 10^{-18}$ $m^2 s^{-1}$, in very poor agreement with experiment. However, the surfaces of the copper cell probably have a roughness of order 0.1–1 μm. This roughness is not leveled by the saturated helium film with a thickness of order 200 Å. Since the microscopic angle θ does not significantly vary over many atomic wavelengths the H-atom motions do not motionally average the surface normal to give $\bar{\theta}$. Thus the angular factors in Eq. (11) must be averaged over the rough surface. We assume a simple model in which the surface normal has a spherical probability for $\theta < 45°$, so that the surface looks like a muffin tin. The surface area becomes a factor $\alpha = 1.17$ times the projected area; the experimental value of G_s is reduced by α. By averaging about any angle $\bar{\theta}$ we find that $G_{s,0}$ dominates $G_{s,2}$. For $\bar{\theta} = 90°$ we find $G_s = 0.23 \times 10^{-17}$ $m^2 s^{-1}$, which is an order of magnitude smaller than experiment. We note that for an atomically flat surface the maximum value of G_s is 0.52×10^{-17} $m^2 s^{-1}$ at $\theta = 45°$ and the minimum value is $G_s = 0$ at $\theta = 0°$. Compare the latter case with a rough surface with $\bar{\theta} = 0°$. Averaging as above we then find $G_s = 0.36 \times 10^{-17}$ $m^2 s^{-1}$. We conclude that, unless surfaces are atomically flat, the geometry of an experimental cell is unimportant with respect to T_1.

It is interesting to compare also with other theoretical methods. We restrict ourselves to the $G_{s,0}$ term, which is apparently the dominant term. Starting from the approach of the present paper (see Table I: $2\frac{1}{2}$D model), let us first study the effect of leaving out the z averaging of the triplet potential. It turns out that this effect is negligible ($2\frac{1}{4}$D model). A significant effect is probably restricted to higher energies where the relative wavelength becomes comparable to the radial displacement of the classical turning point from σ to smaller radii due to z averaging. Next, let us also leave out the z average of the dipole-dipole interactions. This does have an appreciable effect: an increase by a factor of about 3.5 (2D model), due to the increase of the dipole-dipole coupling at small distances. In order to compare with previous work based on hard-sphere scattering, it is also of interest to study the three above-mentioned averaging options in combination with a hard-sphere potential with radius $\sigma = 3.67$ Å. We have used the exact hard-sphere radial wave functions, consisting of Bessel and Neumann functions. In this case the $2\frac{1}{2}$D and $2\frac{1}{4}$D models coincide and lead to a $G_{s,0}$ value lower than our most exact result by a factor of approximately 2.7. Subsequently, leaving out the z averaging leads to a net decrease by about 1.25. Lagendijk's approach[9] can be considered as a modification of this last method. He uses only the Bessel part of the radial wave function which has a finite value at the hard-core radius and he excludes the hard-core volume from the Born integral. One should expect an increase of $G_{s,0}$ from this modification. From our own calculations we conclude that this approximation is bad: It modifies the final $G_{s,0}$

TABLE I. Comparison of relative magnitudes of $G_{s,0}$ for two-dimensional (2D) approach (no z average, i.e., $Z = 0$), $2\frac{1}{4}$D approach (only dipole-dipole interactions averaged), $2\frac{1}{2}$D approach (all interactions z averaged), for both triplet and hard-sphere potentials.

Averaging option	Triplet potential	Hard sphere
$2\frac{1}{2}$D	1	0.37
$2\frac{1}{4}$D	1	0.37
2D	3.5	0.80

value by a factor of 18 to 21 in the temperature range from 0.2 to 0.4 K. We note, furthermore, that the omission of the Neumann function part enables Lagendijk to express his final binary collision results as expansions in a dimensionless parameter α proportional to $T^{1/2}$. Such an expansion is not possible for the exact hard-sphere case. Our formulas (11) should not be considered as low-energy expansions, but as best-fit polynomials in the above-mentioned temperature interval. If we use Lagendijk's approximation and compare to the triplet potential $2\frac{1}{2}$D model we thus find a factor of approximately 16, compared to a value of 5 found if we extrapolate Lagendijk's formula to our temperature range. Ruckenstein and Siggia also use the hard-sphere model and apparently introduce the same approximation as Lagendijk. Although this approximation tends to lead to better agreement with experiment, our more exact calculation indicates that a major discrepancy still exists.

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