Surface magnetic relaxation rates in spin-polarized hydrogen

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The short-wavelength surface contributions to the magnetic relaxation times \(T_1\) and \(T_2\) are calculated for a dilute H\(_1\) gas, in the classical, low-temperature regime. Both surface rates are highly anisotropic with respect to the direction of the stabilizing field. Of experimental interest is the potentially long \(T_1\) in samples with most of the surface area oriented perpendicular to the stabilizing field.

Ever since the recent stabilization of spin-polarized hydrogen (H\(_1\)), much effort has been directed towards achieving the large densities and low temperatures required to observe Bose-Einstein condensation. The major obstacle facing the experimentalist is that even under the most ideal, yet realizable conditions (temperatures \(T \leq 50\) mK, a large magnetic field \(H_0 \sim 100\) kG, and superfluid \(^4\)He-coated container walls), H\(_1\) is unstable with respect to recombination of the hydrogen atoms into molecules.

At cryogenic temperatures only the lowest two hyperfine states of H\(_1\) (denoted by \(|a\rangle\) and \(|b\rangle\), respectively) are thermally populated. In this regime a sample of H\(_1\) decays predominantly through binary collisions on the \(^4\)He surface that involve at least one atom in the lowest hyperfine state (state \(|a\rangle\)). This process does not completely exhaust the \(|b\rangle\) state population and thus the lifetime of a sample of H\(_1\) is limited by the relaxation time between the two hyperfine states \(|a\rangle\) and \(|b\rangle\), denoted by \(T_1\).

In planning future experiments on H\(_1\) it then becomes important to understand in some detail the mechanisms responsible for \(T_1\) relaxation. In this Communication we identify these mechanisms and present analytical calculations which should clarify the dependence of \(T_1^{-1}\) on the relevant experimental parameters. The rate for a bulk sample, denoted by \(T_1^B\), has already been discussed in Refs. 3 and 4. Here we emphasize the calculation of the surface contribution \(T_1^S\), which we expect to be important at the lowest temperatures. In what follows, recombination is ignored, and for the present the system is assumed to be close to thermal equilibrium.

The \(T_1\) relaxation time is the decay time for a small departure of the component of the magnetization along the stabilizing field (or equivalently of the relative population of \(|a\rangle\) and \(|b\rangle\)) from its equilibrium value. (Hereafter, the stabilizing field is taken to point in the \(z\) direction.) To arrive at a quantitative expression for \(T_1\), we consider the linear response of the system to a field \(\delta h = \delta M_z / x_z\) generated by a slow homogeneous fluctuation \(\delta M_z\) in the total \(z\) magnetization \(M_z\), where \(x_z = (\partial M_z / \partial h)_T\) is the static magnetic susceptibility. This leads, after standard manipulations, to the Kubo-like formula

\[
T_1^{-1} = - \frac{\beta}{2k_x^2 x_z} \int_{-\infty}^{\infty} dt \langle [H, M_z] [H(t), M_z(t)] \rangle
\]

The thermal average indicated by \(\langle \rangle\) is performed in the macrocanonical ensemble, \(H\) is the Hamiltonian of the system, and \(\beta = 1 / k_B T\).

The derivation of (1) assumes that the torque-torque correlation function decays on a time scale whose magnitude is set by the thermalization time of the translational degrees of freedom, which is fast compared to \(T_1\). Subject to the restrictions noted above, formula (1) applies at all temperatures, in the normal as well as superfluid regimes, and includes both kinetic (short-wavelength) and hydrodynamic (long-wavelength) contributions to the rate. Finally, (1) takes into account both bulk and surface relaxation processes.

At the temperatures and densities of interest a collection of H\(_1\) atoms behaves like a gas of spin-\(1/2\) bosons, i.e., bosons with two internal states. In this regime calculations are most conveniently done in terms of two boson operators \(\psi_a^\dagger (r)\) and \(\psi_b^\dagger (r)\) which create H\(_1\) atoms in the \(|a\rangle\) and \(|b\rangle\) states, respec-
respectively. The $z$ component of the magnetization $M_z$ is then written as
\[ \mu_z [\rho_a(\mathbf{F}) - \rho_b(\mathbf{F})] - \mu_\rho(\mathbf{F}), \]
where $\rho_a(\mathbf{F}) = \psi_a(\mathbf{F})\psi_a(\mathbf{F})$ are the number densities of the two species; $\rho(\mathbf{F}) = \rho_a(\mathbf{F}) + \rho_b(\mathbf{F})$ is the total number density; finally, $\mu_z = \mu_\rho - e^2\mu_s$, where $\mu_s$ and $\mu_\rho$ are the magnetic moments of the proton and electron, respectively, and $e\approx2\times10^{-3}$ in a magnetic field of 100 kG) is the admixture of the “wrong” electronic spin (i.e., spin up) in the $|\alpha\rangle$ state. The largest contribution to the correlation function in (1) comes from the dipolar interaction between the electron on one atom and the proton on another. In terms of $\psi_a$ and $\psi_b$, the corresponding term in the Hamiltonian can be written as
\[ H_{\text{dip}} = 3\mu_\rho \mu_s \int d\mathbf{r} \int d\mathbf{r}' \psi_a^*(\mathbf{r}') \psi_b(\mathbf{r}') \]
\[ \times \rho(\mathbf{r}') F(\mathbf{r}' - \mathbf{r}'') + \text{H.c.} \]
(2)

Here H.c. stands for “Hermitian conjugate,” $\mu_\rho = \mu_\rho + e\mu_s$, and $F(\mathbf{r}) = \sqrt{A\pi}Y_{11}(\mathbf{r})/|\mathbf{r}|^3$, where $Y_{11}$ is the $l = 2$, $m = 1$ spherical harmonic.

At low densities the correlation function in (1) can be evaluated analytically by standard techniques. In a $T_1$ process the typical energy of a $\text{H}_1$ atom is of the order $H_2/2$ ($H_2$ = Zeeman energy $\approx 50$ mK) which is appreciably higher than the triplet potential energy ($<5$ mK/particle). Thus the atoms can be regarded as free until the density becomes so high that hydrodynamic and mode-coupling arguments are required to evaluate (1). The main effect of the triplet interaction (with one exception to be discussed below) is to cut off the pair correlation function whenever any two particles come closer together than a hard-core diameter, $\sigma \approx 3.76$ Å. This we put in by hand in our calculation by introducing suitable short-distance cutoffs in the spacial integrations. However, for the bulk contribution at temperatures well below 1.7 K ($\sim h^2/(2m^2\lambda_k)$), where the thermal de Broglie wavelength $\lambda_T$ becomes larger than $\sigma$, we expect

\[ F_1(\beta H_x, \sigma; E_k) = \left[ \frac{1}{2} (m H_x/\hbar^2) \exp(-\beta H_x/2) (1 + 2/\beta H_x) \right. \]
\[ + \left. (\beta m H_x^2/36 \hbar^2 \pi) [3 K_0^2 (\beta H_x/4) - K_1^2 (\beta H_x/4)] \cosh(\beta H_x/2) \right. \]
(5)

Here $K_0$ and $K_1$ are modified Bessel functions of zeroth and first order, respectively. The form factor $F(\sigma)$ is given by
\[ F(\sigma) = \left[ \int d\mathbf{r} \int d\mathbf{r}' |\phi_0(\mathbf{r})|^2 |\phi(\mathbf{r}')|^2 \right] \]
\[ \times \frac{\sigma^2}{[\sigma^2 + (\xi - \xi')^2]^{1/2}} , \]
(6)

and comes from averaging the three-dimensional dipolar interaction over $\phi_0(\mathbf{r})$. For $H_1$, (4) and (5) are valid in the low-temperature regime ($T < 50$ mK for $\text{H}_1$ on $^4$He) where the inequality $(\beta E_k)^{1/2} >> 1$ is well satisfied. In particular, in the limit $\beta H_x >> 1$, $F_1 = F(\sigma)/2$ and $F_2 = m H_x/9\hbar^2$.

In the high-temperature regime, $50$ mK < $T < 1.7$ K, $(\beta E_k)^{1/2}$ is sufficiently close to unity so that an
analytic or perturbative treatment of $F_1$ and $F_2$ is not feasible and some five-dimensional integrals remain to be evaluated numerically. To crudely estimate the rate, we assume that the integration over bound-state wave functions singles out a scale $\delta \sim (k/2mE_0)^{1/2}$ for the relative coordinate $\xi_1 - \xi_2$ for two atoms. For $\beta H_z << 1$, Eq. (3) then becomes
\begin{equation}
T_{13}^z(\Omega, \beta H_z << 1) \sim \left( \frac{3}{2} (n_5 \mu_1^2 \mu_3^2 n m^2 / \hbar^2) \right) \sin^2(2\Omega) \int F(\sigma) f_1(t) + \frac{2}{9} (m / \hbar^2 \beta) \sin^2 \Omega (1 + \cos^2 \Omega) f_2(t) \right) .
\end{equation}

In the temperature range $0.1 \leq T \leq 0.5 \text{ K}$, $f_1$ and $f_2$ are only weakly temperature dependent; $f_1 \sim 0.2$ and $f_2 \sim 0.04$, and $f_1 \sim 0.07$ and $f_2 \sim 0.01$ for $\text{H}_1$ adsorbed on $^4\text{He}$ and $^3\text{He}$ (Ref. 5), respectively. The (inappropriate) high-temperature limit of (4) and (5) would give $f_1 = 1$ and $f_2 = (1 - 2/\pi)$.

Note that $T_{13}^z$ in (3) vanishes when the stabilizing field is normal to the surface. Under these conditions the electronic moments are polarized perpendicular and are restricted to move parallel to the surface. This generates a fluctuating dipolar field perpendicular to the surface which cannot flip the spin of a neighboring atom. In the real system, however, the triplet interaction $H_T$ induces virtual transitions between $\phi_0(\xi)$ and bulk states which break the symmetry of the surface state. This gives rise to the transverse local-field fluctuations necessary for $T_1$ relaxation on the surface.

The largest contribution to $T_{13}^z$ with the field perpendicular to the surface ($\Omega = 0$) is second order in the triplet interaction $H_T$. This can be seen by writing the correlation function in (1) explicitly as a trace over many-body eigenstates $|m\rangle$ of the Hamiltonian with energies $E_m$:

\begin{equation}
T_{13}^z = \sum \langle \psi_a^r | (E_n - E_m) \int dr_1 \int dr_2 \int dr_3 \int dr_4' \int F(r_1 - r_2) F^*(r_1' - r_2') \times \langle n | \psi_a^r \psi_s(r_1) \rho(r_2) | m \rangle \langle m | \rho(r_3) \psi_s^* (r_4') \psi_a (r_4) | n \rangle .
\end{equation}

Imagine now doing perturbation theory in $H_T$ so as to mix the many-body surface states with bulk states. Then to first order in $H_T$ one of the matrix elements in (8) (without loss of generality the first) involves only surface atoms and must be proportional to $|\phi_0(\xi_1)|^2 |\phi_0(\xi_2)|^2$. For $\Omega = 0$ the corresponding $F(r_3 - r_4)$ in (2) is odd with respect to interchanging $\xi_1$ and $\xi_2$ and the over all $\xi_1$ and $\xi_2$ in (8) vanishes.

For a typical contribution to second order in $H_T$, we find approximately
\begin{equation}
T_{13}^z(\Omega = 0) \sim \left( 10^4 \mu_1^2 \mu_3^2 n m^2 / \hbar^2 \beta \right) \left( n_5 a^2 \right)^2 \times (k/ma^2 \hbar) G(\beta, H_z, \sigma; E_b) ,
\end{equation}
where the $s$-wave scattering length, $a = 0.73$ Å (Ref. 8), and all integrals over bound-state wave functions, were crudely estimated in the way explained in the paragraph preceding Eq. (7). Like $F_3$ and $F_2$ in (3) the function $G$ in (9) can only be calculated analytically for $T << 50$ mK where we find $G \approx \beta H_z / 2$. At higher temperatures ($0.1 < T < 0.5 \text{ K}$) our estimates give $G$ of order unity for $\text{H}_1$ adsorbed on both $^4\text{He}$ and $^3\text{He}$. This suggests that even for surface densities as high as $n_s \sim 10^5$ atoms/cm$^2$, $T_{13}^z(\Omega = 0)$ is $10$–$100$ times longer than the relaxation times for arbitrarily oriented surfaces. Thus in order to study a two-dimensional $\text{H}_1$ gas, it is essential to orient most of the available surface area perpendicular to the stabilizing field.

In the classical regime $(n_5 a^2 \sigma/\hbar \beta << 1$ and $n_5 \sigma a^2 \lambda \beta << 1$), the total relaxation rate of $M_z$ consists of two contributions: a bulk rate $T_{13}^{-1} N_B / N$, and a surface rate $T_{13}^{-1} N_S / N$. The total number of atoms is $N = N_B + N_S$, where $N_B$, $N_S$ denote the total number of bulk and surface atoms, respectively, and the ratio $N_S / N_B$ is given by thermodynamics. The calculated bulk rate $^{3,4}$ agrees within a factor of 2 with the experimental value reported recently by Cline et al.$^9,10$ However, the deviation from the bulk rate, attributed by the MIT group to pure surface relaxation, appears to be two orders of magnitude larger than our estimate for $(N_S / N) T_{13}^{-1} (\Omega = \pi / 2)$. A more detailed account of the experimental situation is required before this discrepancy can be resolved.

Direct comparison with experiment is meaningful since it appears that our results are not restricted to the linear-response regime. We have considered the problem of the decay rate of a highly nonthermal state with an inverted population. When heating effects were ignored we found that, to second order in the dipolar interaction (2), $M_z$ relaxes exponentially on a time scale identical with the one obtained near thermal equilibrium. This somewhat surprising result can be traced back to the fact that (2) generates local fluctuating torques, dependent only on the total density (and not on the z magnetization of the state), which can flip only one spin at a time. Thus after a time, long compared to the collision time, the environment of any one spin is identical to that near thermal equilibrium. The details of the calculation will be presented elsewhere.

In the above discussion, the short-wavelength nonhydrodynamic modes were found to dominate the
relaxation process. Relaxation through collective modes becomes important only when an atom undergoes many collisions during one Larmor precession (i.e., $H_T/\hbar < 1$ where $\tau$ is the collision time). At the temperatures of interest, this condition requires $n_T > 10^{13}$ atoms/cm$^3$, in qualitative agreement with the calculations in Ref. 11. A similar order-of-magnitude estimate gives $2 \times 10^{20}$ atoms/cm$^3$ as the corresponding crossover density for a bulk sample.

Using the approach developed for $T_1$, we have also calculated the transverse relaxation time $T_2$. In all cases discussed above but one, we find that $T_2$ is of the same order of magnitude as $T_1$. The one exception is the surface contribution with the field perpendicular to the surface in which case $T_{2S}$ is of the same order of magnitude as the first term in (3) (with, for example, $\Omega = \pi/4$). Thus at low temperatures and for $n_T < 10^{13}$ atoms/cm$^2$, $T_{1S}(\Omega = 0)$ will be much longer than $T_{2S}(\Omega = 0)$. This configuration therefore suggests one means by which to realize the novel magnetic resonance effects proposed in Refs. 12 and 4, that are unique to Bose condensed spin-polarized hydrogen.

Statt has independently calculated the surface $T_1$ in the high-temperature regime ($\beta H_T < 1$) (Ref. 7). His rates are identical with ours in the limit of an infinite binding energy but are faster (by, at most, an order of magnitude) for $E_b \sim 1$ K. We expect that the disagreement is due to the different approximations used in evaluating the integrals over bound-state wave functions.

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6The factor $f_2$ involves the difference of two small numbers. Even though these are, individually, well approximated by the first term in a perturbation expansion in $(2m^2/\hbar^2 \beta)$, $f_2$ itself may be sensitive to the value of $\sigma$.
7The same observation was made, independently, by B. Statt, Phys. Rev. B 25, 6035 (1982) (this issue), and A. Lagendijk, ibid. 25, 2454 (1982); the latter author considers a model which ignores the finite extent of the bound-state wave function perpendicular to the surface of the film. See also P. Brütel and H. M. McConnell, Proc. Nat. Acad. Sci. U.S.A. 72, 1451 (1975).
10In terms of $T_{1B}$, $G_B$ in Ref. 9 is given by $G_B = 1/2T_{1B} n_B$, and similarly, $G_S = 1/2T_{2S} n_S$ for the surface contribution.