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Surface magnetic relaxation rates in spin-polarized hydrogen

Andrei E. Ruckenstein* and Eric D. Siggia*
Institute for Theoretical Physics, University of California at Santa Barbara, Santa Barbara, California 93106
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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↓ 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 $T \lesssim 50$ mK, a large magnetic field $H_0 \sim 100$ kG, and superfluid ^{4}He-coated container walls), H↓ is unstable with respect to recombination of the hydrogen atoms into molecules.

At cryogenic temperatures only the lowest two hyperfine states of H↓ (denoted by $|a\rangle$ and $|b\rangle$, respectively) are thermally populated. In this regime a sample of H↓ decays predominantly through binary collisions on the ^{4}He surface that involve at least one atom in the lowest hyperfine state (state $|a\rangle$).[2] This process does not completely exhaust the $|b\rangle$ state population and thus the lifetime of a sample of H↓ is limited by the relaxation time between the two hyperfine states $|a\rangle$ and $|b\rangle$, denoted by T_1.[3]

In planning future experiments on H↓ 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_{1B}^{-1}, has already been discussed in Refs. 3 and 4. Here we emphasize the calculation of the surface contribution T_{1S}^{-1}, 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 $\delta h = \delta M_z/\chi_z$ generated by a slow homogeneous fluctuation δM_z in the total z magnetization M_z, where $\chi_z = (\partial M_z/\partial h)_T$ is the static magnetic susceptibility. This leads, after standard manipulations, to the Kubo-like formula

$$T_1^{-1} = -\left(\frac{\beta}{2\hbar^2\chi_z}\right)\int_{-\infty}^{\infty} dt\,\langle [H,M_z][H(t),M_z(t)]\rangle \quad . \tag{1}$$

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↓ atoms behaves like a gas of spin-$\frac{1}{2}$ bosons, i.e., bosons with two internal states. In this regime calculations are most conveniently done in terms of two bose operators $\psi_a^\dagger(r)$ and $\psi_b^\dagger(r)$ which create H↓ atoms in the $|a\rangle$ and $|b\rangle$ states, respec-

tively. The z component of the magnetization M_z is then written as $\mu_z[\rho_a(\vec{r}) - \rho_b(\vec{r})] - \mu_e\rho(\vec{r})$, where $\rho_{a,b}(\vec{r}) = \psi^\dagger_{a,b}(\vec{r})\psi_{a,b}(\vec{r})$ are the number densities of the two species; $\rho(\vec{r}) = \rho_a(\vec{r}) + \rho_b(\vec{r})$ is the total number density; finally, $\mu_z = \mu_p - \epsilon^2\mu_e$, where μ_p and μ_e are the magnetic moments of the proton and electron, respectively, and ϵ ($\sim 2\times 10^{-3}$ in a magnetic field of 100 kG) is the admixture of the "wrong" electronic spin (i.e., spin up) in the $|a\rangle$ 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.[4] In terms of ψ_a and ψ_b, the corresponding term in the Hamiltonian can be written as

$$H_{\rm dip} = 3\mu_e\mu_\perp \int d\vec{r}' \int d\vec{r}''\psi^\dagger_a(\vec{r}')\psi_b(\vec{r}') \times \rho(\vec{r}'')F(\vec{r}' - \vec{r}'') + \text{H.c.} \tag{2}$$

Here H.c. stands for "Hermitian conjugate," $\mu_\perp = \mu_p + \epsilon\mu_e$, and $F(\vec{r}) = \sqrt{8\pi/15}\,Y_{21}(\hat{r})/|\vec{r}|^3$, where Y_{21} 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 H↓ atom is of the order $H_z/2$ ($H_z \equiv$ Zeeman energy ~ 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, $\sigma \sim 3.76$ 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 [$\sim \hbar^2/(2m\sigma^2 k_B)$], where the thermal de Broglie wavelength λ_T becomes larger than σ, we expect our results to be independent of σ. The surface rate, on the other hand, depends only weakly on σ in a way which involves the details of the one-atom bound-state wave function.

To treat explicitly surface relaxation processes we write the many-body wave function as a linear superposition of product states of (the complete set of) one-particle wave functions $\phi_\alpha(\xi)e^{i\vec{k}\cdot\vec{\rho}}$, $\alpha = 0, 1, 2, \ldots$. Here ξ and $\vec{\rho}$ denote coordinates measured perpendicular and parallel to one of the surfaces of the container, respectively. Only $\phi_0(\xi)$ is localized on the surface with binding energy E_b of order 1 K. Consider an "ideal" two-dimensional gas of H↓ atoms of surface density n_S ($< 10^{13}$ atoms/cm^2), in which all atoms are restricted to $\alpha = 0$ states. In the classical regime (defined by $n_S\lambda_T^2 << 1$), the explicit evaluation of the correlation function in (1) to second order in $H_{\rm dip}$ gives

$$T_{1S}^{-1}(\Omega) = \tfrac{9}{2}(n_S\mu_e^2\mu_\perp^2 m\pi^2/\hbar^3) \times [\sin^2(2\Omega)F_1(\beta,H_z,\sigma;E_b) + \sin^2\Omega(1+\cos^2\Omega)F_2(\beta,H_z,\sigma;E_b)]\ , \tag{3}$$

where Ω is the angle between the direction of the stabilizing field and the normal to the surface. In general F_1 and F_2 are complicated functions of their arguments and depend in an intricate way on the details of the bound-state wave functions (symbolized by E_b). In the limit

$$(\beta\hbar^2/2m\sigma^2)^{1/2} > (\beta E_b)^{1/2} >> 1$$

the expressions for F_1 and F_2 simplify considerably. For arbitrary βH_z we find

$$F_1(\beta,H_z,\sigma;E_b) = F(\sigma)\cosh(\beta H_z/2)\exp(-\beta H_z/2) \tag{4}$$

and

$$F_2(\beta,H_z,\sigma;E_b) = \{\tfrac{1}{9}(mH_z/\hbar^2)\exp(-\beta H_z/2)(1+2/\beta H_z) + (\beta m H_z^2/36\hbar^2\pi)[3K_0^2(\beta H_z/4) - K_1^2(\beta H_z/4)]\}\cosh(\beta H_z/2)\ . \tag{5}$$

Here K_0 and K_1 are modified Bessel functions of zeroth and first order, respectively. The form factor $F(\sigma)$ is given by

$$F(\sigma) = \left\{\int d\xi \int d\xi' |\phi_0(\xi)|^2|\phi_0(\xi')|^2 \times \frac{\sigma^2}{[\sigma^2 + (\xi-\xi')^2]^{3/2}}\right\}^2\ , \tag{6}$$

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

In the high-temperature regime, 50 mK $< T <$ 1.7 K, $(\beta E_b)^{1/2}$ 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 intergrals remain to be evaluated numerically. To crudely estimate the rate, we assume that the integration over bound-state wave functions singles out a scale $\delta \sim (\hbar^2/2mE_b)^{1/2}$ for the relative coordinate $\xi_1 - \xi_2$ for two atoms. For $\beta H_z \ll 1$, Eq. (3) then becomes

$$T_{1S}^{-1}(\Omega, \beta H_z \ll 1) \sim \tfrac{9}{2}(n_S \mu_e^2 \mu_I^2 m \pi^2/\hbar^3)[\sin^2(2\Omega) F(\sigma) f_1(T) + \tfrac{2}{9}(m/\hbar^2\beta)\sin^2\Omega(1+\cos^2\Omega) f_2(T)] \quad . \quad (7)$$

In the temperature range $0.1 \leq T \leq 0.5$ K, f_1 and f_2 are only weakly temperature dependent; $f_1 \sim 0.2$ and $f_2 \sim 0.04$, and $f_1 \sim 0.7$ and $f_2 \sim 0.01$ for H$\downarrow$ adsorbed on ^{4}He and ^{3}He (Ref. 5), respectively.[6] The (inappropriate) high-temperature limit of (4) and (5) would give $f_1 = 1$ and $f_2 = (1 - 2/\pi)$.

Note that T_{1S}^{-1} in (3) vanishes when the stabilizing field is normal to the surface.[7] 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_{1S}^{-1} 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:

$$T_{1S}^{-1} \propto \sum_{n,m} e^{-\beta E_n} \delta(E_n - E_m) \int dr_1 \int dr_2 \int dr_{1'} \int dr_{2'} F(r_1 - r_2) F^*(r_{1'} - r_{2'}) \times \langle n|\psi_a^\dagger(r_1)\psi_b(r_1)\rho(r_2)|m\rangle\langle m|\rho(r_{2'})\psi_b^\dagger(r_{1'})\psi_a(r_{1'})|n\rangle \quad . \quad (8)$$

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 $F(\vec{r}_1 - \vec{r}_2)$ in (2) is odd with respect to interchanging ξ_1 and $\xi_{2'}$ and the over all ξ_1 and ξ_2 in (8) vanishes.

For a typical contribution to second order in H_T, we find approximately

$$T_{1S}^{-1}(\Omega = 0) \sim (10^4 \mu_e^2 \mu_I^2 n_S m^2/\hbar^5\beta)(n_S a^2)^2 \times (\hbar^2/ma^2E_b) G(\beta, H_z, \sigma; E_b) \quad , \quad (9)$$

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_1 and F_2 in (3) the function G in (9) can only be calculated analytically for $T \ll 50$ mK where we find $G \simeq \beta H_z/2$. At higher temperatures ($0.1 < T < 0.5$ K) our estimates give G of order unity for H$\downarrow$ adsorbed on both ^{4}He and ^{3}He. This suggests that even for surface densities as high as $n_S \sim 10^{13}$ atoms/cm^2, $T_{1S}(\Omega = 0)$ is 10–100 times longer than the relaxation times for arbitrarily oriented surfaces. Thus in order to study a two-dimensional H$\downarrow$ gas, it is essential to orient most of the available surface area perpendicular to the stabilizing field.

In the classical regime ($n_S\lambda_T^2 \ll 1$ and $n_B\lambda_T^3 \ll 1$), the total relaxation rate of M_z consists of two contributions: a bulk rate $T_{1B}^{-1}N_B/N$, and a surface rate $T_{1S}^{-1}N_S/N$. 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 $(N_S/N)T_{1S}^{-1}(\Omega = \pi/2)$. 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 exponetially 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), and 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 Larmour precession (i.e., $H_z\tau/\hbar \ll 1$ where τ is the collision time). At the temperatures of interest, this condition requires $n_S > 10^{13}$ atoms/cm^2, in qualitative agreement with the calculations in Ref. 11. A similar order-of-magnitude estimate gives 2×10^{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}^{-1} 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 $n_S < 10^{13}$ 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 ($\beta H_z \ll 1$) (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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*Premanent address: Laboratory of Solid State Physics, Cornell University, Ithaca, N.Y. 14853.

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