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Strong anisotropy of the nuclear relaxation of spin-polarized atomic hydrogen ($H\downarrow$) in two dimensions

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A two-dimensional model is used to describe spin relaxation of $H\downarrow$ on the surface which takes into account both binary collisions as well as hydrodynamical modes. The relaxation rates show a strong anisotropy with respect to the orientation of the magnetic field. This anisotropy might be very important for the stabilization of high-density $H\downarrow$.

It has recently been shown that atomic hydrogen can be stabilized against recombination up to densities of $\sim 5 \times 10^{16} \text{ cm}^{-3}$.^{1,2} The necessary experimental conditions are a high magnetic field ($\sim 10 \text{ T}$) at low temperatures ($\sim 0.3 \text{ K}$) to polarize the electronic spins, and the coating of the walls with superfluid ^4He , or a mixture of ^4He - ^3He , to suppress condensation and recombination on the surface. One of the most interesting features of this new quantum gas is the possibility of Bose-Einstein condensation (BEC) which is expected to occur at densities $\sim 10^{18} \text{ cm}^{-3}$ for currently available temperatures. Considerable progress has recently been made in identifying and understanding the various decay channels of atomic hydrogen.³⁻⁶ From these works and from the calculation of adsorption isotherms^{7,8} it is clear that both the low-temperature thermodynamics and kinetics of spin-polarized atomic hydrogen ($H\downarrow$) are affected crucially by the presence of container walls. At present the creation of densities of $H\downarrow$ required for BEC seems rather difficult to achieve.^{5,6}

An interesting suggestion for achieving high densities was made by Statt and Berlinsky,⁹ who proposed that $H\downarrow$ might be stabilized in one of its hyperfine states: the pure electronic spin "down" and nuclear spin "down" ($|\downarrow\downarrow\rangle$) the "b" state. Long nuclear relaxation times T_1 (of the order of hours) would prevent rapid thermalization of the "b" atoms (which are relatively stable against recombination) with the ground hyperfine "a" state ($-\downarrow\uparrow + \epsilon\uparrow\downarrow$), which has a small admixture ϵ of reversed electronic spin and recombines at a much higher rate. Similar results were also found by Siggia and Ruckenstein.¹⁰ However, in the latest experiments⁶ a surprising value of $T_1 < 10 \text{ s}$ was found under conditions where theory predicted $T_1 \sim 20\,000 \text{ s}$ in the gas phase, rendering the suggestion of a nuclear-spin-relaxation bottleneck useless.

It is the purpose of this Communication to investigate whether the observed relaxation times can be explained on the basis of a two-dimensional (2D)

model in which adsorbed $H\downarrow$ atoms interact through a dipolar interaction and relax on the surface. It will be shown that the relaxation times on the surface are much shorter than those in the bulk, and indeed can be as short as 0.1 s. However, the relevant kinetic quantity for the instability of b atoms in the bulk due to surface relaxation and recombination is the surface nuclear relaxation rate times the probability that a b atom sits on the surface, and this number turns out to be much smaller than the experimental found lower bound of $\sim 0.1 \text{ s}^{-1}$.⁶ It will be demonstrated that the geometry of the hydrogen-stabilization cell, and its orientation with respect to the confining magnetic field will be very important in reducing the surface nuclear relaxation rates. The results might have important consequences for achieving high densities of $H\downarrow$.

In a simple weak-coupling approach the relaxation rate is the product of a second moment and a narrowing factor: $(T_1)^{-1} = M_2 \tau(\Omega)$, in which M_2 is the second moment (the "average" dipolar interaction squared), and $\tau(\Omega)$, the narrowing factor, is the frequency Fourier transform of a correlation function of a bath variable.¹¹ The translational degrees of freedom of the $H\downarrow$ atoms form the bath, and Ω is a characteristic angular frequency associated with the magnetic degrees of freedom (typically a Larmor or hyperfine frequency).

The importance of the relaxation processes on the surface can be understood on general grounds. In the first place the second moment is much larger in 2D than in 3D because, on the average, the atoms are much closer to each other, and in the second place the translational motion is much "slower" in 2D than in 3D. Both these effects reduce T_1 . Only the two lowest hyperfine levels will be considered: the a and b state. The part of the dipolar interaction (nuclear-electronic and electronic-electronic) having orientational dependence $Y_2^j(\hat{r}_{12})$ (\hat{r}_{12} is the internuclear vector of two interacting atoms) contributes to the relaxation of $H\downarrow$. The second moment of the

longitudinal magnetization, M_2^z , in 3D is calculated to be

$$M_2^z(3D) = \frac{N}{V} \frac{Q(\epsilon)}{3d^3}, \quad (1a)$$

in which d is the hard-core diameter, N the number of particles, V the volume, and $Q(\epsilon) \equiv 0.6\gamma_e^2\gamma_p^2 h^2 \pi \times (1 + \epsilon\gamma_e/\gamma_p)^2$. Here $\epsilon \approx a/\gamma_e hB$ is the mixing parameter, a is the isotropic hyperfine interaction constant, and B is the magnetic field. The second moment of the longitudinal nuclear magnetization on the surface is given by

$$M_2^z(2D) = \frac{15}{256} \frac{N}{A} \frac{Q(\epsilon)}{d^4} [2(1 - \cos^4\theta) + \sin^2 2\theta] \quad (1b)$$

in which A is the area of the surface and θ is the angle between the magnetic field and the normal to the plane. Two important conclusions can already be drawn from Eqs. (1): (i) for a general orientation of the magnetic field, and for practical values of N/V and N/A , $M_2^z(2D) \gg M_2^z(3D)$, and (ii) if the magnetic field B is perpendicular to the surface ($\theta = 0^\circ$) $M_2^z(2D)$ vanishes, and there is no relaxation within this model.

The translational motion of the atoms provide a narrowing mechanism, and in a simple weak-coupling approach the actual relaxation rates are obtained by multiplying the second moments by the narrowing factors. One must be careful, however, because breakdown of simple motional-narrowing theory in reduced dimensionality is possible. This breakdown will result in strong frequency-dependent contributions to T_1 , and diverging contributions to T_2 , which can be renormalized with simple mode-coupling theory.¹²

The bath correlation functions relevant for dipolar relaxation are¹¹

$$f_m(t) = \frac{1}{2} \sum_{i(\neq j)} \left\langle \left\langle \frac{Y_2^m(\hat{r}_{ij}(0))^*}{r_{ij}^3(0)} \frac{Y_2^m(\hat{r}_{ij}(t))}{r_{ij}^3(t)} \right\rangle \right\rangle, \quad (2)$$

in which the spherical harmonics are described in a reference frame in which the magnetic field defines the z direction, and the double brackets indicate thermal averaging. For the description of surface relaxation¹³ it is more useful to transform the spherical harmonics to a frame in which the z direction is normal to the plane. The correlation functions in this frame will be denoted by $F_m(t)$, and the relevant relationship for H \downarrow is, neglecting small imaginary parts,

$$f_1(t) = \frac{1}{2} (1 - \cos^4\theta) F_2(t) + \frac{3}{8} \sin^2 2\theta F_0(t). \quad (3)$$

Equation (3) shows an important feature of the present model. On general grounds one expects that $F_2(t)$ decays much faster than $F_0(t)$, because in the latter there can be no decay due to loss of correlation in the *direction* of the internuclear vectors, but only due to loss of correlation in the *magnitude* of the in-

ternuclear vectors. So if a surface cannot be parallel to the magnetic field it should be oriented as well as possible perpendicular to the magnetic field to suppress the important $m=0$ component of the relaxation.

I now come to the actual calculation of T_1 . Two model contributions have been taken into account: (i) the effect of binary collisions (BCM), using a hard core, and (ii) the relaxation due to collective, hydrodynamic modes using a diffusion model (DIF), so $(T_1)^{-1} = (T_1^{\text{BCM}})^{-1} + (T_1^{\text{DIF}})^{-1}$. In both models the contributions can be partitioned in m -dependent components, $(T_{1,m})^{-1}$, in the following way:

$$\frac{1}{T_1} = \frac{1}{2} (1 - \cos^4\theta) \frac{1}{T_{1,2}} + \frac{3}{8} \sin^2 2\theta \frac{1}{T_{1,0}}, \quad (4a)$$

in which

$$\frac{1}{T_{1,m}} = \frac{1}{T_{1,m}^{\text{BCM}}} + \frac{1}{T_{1,m}^{\text{DIF}}}. \quad (4b)$$

$(T_{1,m})^{-1}$ denotes the contribution to the relaxation rate of the Y_2^m component, and is given by

$$\frac{1}{T_{1,m}} = Q(\epsilon) F_m(\Omega), \quad (5)$$

in which $F_m(\omega)$ is the Fourier transform of $F_m(t)$: $F_m(\omega) = \int e^{i\omega t} F_m(t) dt$, and Ω is the energy difference between the a and the b state in angular frequency units. Note that $F_m(t=0)$ is the m component of the second moment. Usually $F_m(\Omega)$ can be replaced by $F_m(\omega=0)$ (extreme motional narrowing), unless there is a zero-frequency divergence. Taking the $\omega=0$ limit in the BCM in 2D does not give rise to problems, and the results in the low-temperature region are

$$F_0^{\text{BCM}}(\omega=0) = \frac{N}{A} \hbar d^{-4} (k_B T)^{-1} (0.98\alpha^2 - 2.2\alpha^3) \quad (6a)$$

and

$$F_2^{\text{BCM}}(\omega=0) = \frac{N}{A} \hbar d^{-4} (k_B T)^{-1} (0.33\alpha^4 - 0.55\alpha^5), \quad (6b)$$

where the expansion parameter α is given by

$$\alpha = (mk_B T d^2 \hbar^{-2})^{1/2}. \quad (6c)$$

More terms in the expansion in α are easily obtained.

One cannot overlook the possibility that collective modes make a contribution to the relaxation rates. In the non-Bose-condensed phase these can only be of hydrodynamic nature, and I will assume a simple diffusion model. The concept of self-diffusion in dense 2D fluids is still a controversial issue, and is far from being settled.¹⁴ However, for our purpose it is more than sufficient to use the first Enskog approximation in 2D, because the logarithmic correc-

tions will be very small for the actual surface coverages of H \downarrow . The result is

$$F_2^{\text{DIF}}(\omega=0) = 0.30 \frac{N}{A} \frac{1}{Dd^2}, \quad (7a)$$

in which D is the diffusion coefficient in the first Enskog approximation in 2D.¹³ As $F_0^{\text{DIF}}(\omega=0)$ diverges, the frequency dependence has to be retained, giving

$$\lim_{\omega \rightarrow 0} F_0^{\text{DIF}}(\omega) = 0.31 \frac{N}{A} \frac{1}{Dd^2} \left(\ln \frac{D}{\omega d^2} - 2.2 \right). \quad (7b)$$

All my results were obtained using a correlation-function formalism, and agreed exactly with the ap-

proaches using the Boltzmann equation for the three-dimensional case. The 3D results were in exact agreement with the work of Siggia and Ruckenstein, who also used a hard-core potential, and differed by approximately a factor of 2 from the work of Statt and Berlinsky, who used a more realistic potential. For the 2D calculations suitable ultraviolet cutoffs had to be introduced. The advantage of the correlation-function approach is the ease with which the frequency dependence of the relaxation rates can be calculated.

The relaxation rates can now be evaluated and the results are

$$\frac{1}{T_{1,0}^{\text{BCM}}} = \left(0.53 \times 10^{-12} \frac{N}{A} - 0.63 \times 10^{-12} \frac{N}{A} T^{1/2} \right) \left(1 + \frac{16.68}{B} \right)^2, \quad (8a)$$

$$\frac{1}{T_{1,2}^{\text{BCM}}} = \left(0.51 \times 10^{-13} \frac{N}{A} T - 0.44 \times 10^{-13} \frac{N}{A} T^{3/2} \right) \left(1 + \frac{16.68}{B} \right)^2, \quad (8b)$$

$$\frac{1}{T_{1,0}^{\text{DIF}}} = 0.15 \times 10^{-26} \left\{ \ln \left[0.11 \times 10^{15} \left(\frac{N}{A} \right)^{-1} T^{1/2} \right] - 2.2 \right\} \left(\frac{N}{A} \right)^2 T^{-1/2} \left(1 + \frac{16.68}{B} \right)^2, \quad (8c)$$

$$\frac{1}{T_{1,2}^{\text{DIF}}} = 0.15 \times 10^{-26} \left(\frac{N}{A} \right)^2 T^{-1/2} \left(1 + \frac{16.68}{B} \right)^2, \quad (8d)$$

In Eqs. (8) N/A should be in cm^{-2} and T in kelvin. The magnetic field B should be in tesla. Let us calculate the various contributions for experimental conditions similar to those used in Ref. 6: $N/A = 10^{12} \text{ cm}^{-2}$, $T = 0.1 \text{ K}$, and $B = 10 \text{ T}$. One obtains $T_{1,0}^{\text{BCM}} = 0.4 \text{ s}$, $T_{1,2}^{\text{BCM}} = 38 \text{ s}$, $T_{1,0}^{\text{DIF}} = 5 \text{ s}$, and $T_{1,2}^{\text{DIF}} = 30 \text{ s}$. It is clear that these T_1 's are much shorter than the bulk (3D) relaxation times. However, if an effective T_1 of bulk atoms would be calculated due to surface relaxation, Eqs. (8) should be multiplied by the probability for an atom to be on the surface. This would increase the relaxation times by several orders of magnitude. It is clear that these effective relaxation times will be much longer than the experimental bound of Van Yperen *et al.*⁶: $T_1 < 10 \text{ s}$. This means that either a different, much more effective, relaxation mechanism is operative, or that the inter-

pretation of the experiments should be reconsidered. Another point is that the adsorbed atoms will have some freedom to move perpendicular to the surface. It is extremely difficult to take this effect into account in a satisfactory way since one needs anisotropic two-particle correlation functions. For this reason the T_1 's calculated in this paper should be considered as lower bounds for adsorbed H \downarrow .

Note added in proof. Very recently Cline, Greytak, and Kleppner¹⁵ observed nuclear spin relaxation rates of the same order of magnitude as those being calculated in this paper.

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