PHYSICAL REVIEW B VOLUME 35, NUMBER 10 1 APRIL 1987

Theory of nuclear spin-lattice relaxation of spin-polarized hydrogen on liquid-helium-coated surfaces due to magnetic particles in the substrate

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(Received 14 July 1986)

A theory is presented for the nuclear spin-lattice relaxation rate $1/T_1$ for electron-spin-polarized H atoms on or near a liquid-He-coated surface due to magnetic particles in the substrate below the He film. The H-atom motion is assumed to be free-particle-like for atoms on the surface, while atoms in the gas are assumed to be reflected elastically by the surface. $1/T_1$ is found to exhibit a minimum at low temperatures due to two competing tendencies. $1/T_1$ decreases with decreasing temperature because lower thermal velocities give a smaller density of fluctuating magnetic fields at the NMR frequency ω. However, at lower temperatures, the atoms spend more time on the surface so that eventually $1/T_1 \sim e^{\beta E_B}$, where $\beta = 1/k_B T$ and E_B is the binding energy of H to liquid He. For atoms on the surface, the dominant frequency dependence of $1/T_1$ is given approximately by $\exp[-3(\beta m \omega^2 d^2/2)^{1/3}]$ which predicts a very strong dependence of the relaxation on ω. For atoms reflected by the surface, the dependence is ω^{-4}, which agrees with the classical calculation of Purcell as reported by Kleppner, Goldenberg, and Ramsey.

I. INTRODUCTION

At low temperatures and high magnetic fields, a gas of hydrogen atoms becomes electron-spin polarized in the sense that all of the atoms are in the two lowest hyperfine states, $|a\rangle = [\,|\downarrow\downharpoonright\rangle - \epsilon\,|\uparrow\upharpoonright\rangle]/\sqrt{2}$ and $|b\rangle = |\downarrow\upharpoonright\rangle$, where $\downarrow, \uparrow$ ($\downharpoonright\upharpoonright$) refer to electron (proton) spin states and $\epsilon \approx (253\text{G})/H$ results from hyperfine mixing.[1] Since atoms in the $|a\rangle$ state preferentially recombine into molecules, then if T_1, the time for *a*-*b* relaxation, is sufficiently long, the gas will become nuclear-spin polarized, i.e., the ratio of the number of *b* atoms to the number of *a* atoms can become very large. This so-called "T_1 bottleneck" was predicted by us in 1980 (Ref. 2) and first confirmed experimentally by Cline, Greytak, and Kleppner.[3]

An earlier experimental attempt to study the T_1 bottleneck by van Yperen *et al.*[4] failed to observe this effect, apparently because of fast relaxation due to macroscopic impurities in the cell walls.[5] Surface relaxation due to substrate magnetic impurities has also been observed by Yurke *et al.*,[6] and in our own laboratory,[7] where we were able to measure, for the first time, the temperature dependence of this one-body relaxation mechanism. We also succeeded in suppressing the one-body relaxation by depositing several hundred Angstroms of solid molecular H_2 on the impure copper substrate. For sufficiently thick coatings, probably several thousands of angstroms, this mechanism could be made unobservably slow.

In this paper, we present a detailed theory of surface spin relaxation of H atoms due to substrate magnetic impurities. Parts of this theory were summarized in our earlier work.[7] The theory is a variation on and in some ways an extension of the work of Chapman and Bloom[8] on surface spin relaxation of ^{3}He. The main differences are that they treated the case of classical diffusive motion in the long- and small-jump limits while we concentrate on the quantum-mechanical free-particle limit. In the extreme free-particle limit for a perfectly smooth surface, atoms are only scattered by the inhomogeneous surface field which flips the spins. This type of motion is appropriate for a low-density gas of H atoms moving on a liquid ^{4}He-coated surface where the picture of atoms being bound to the surface with a binding energy E_B but otherwise free to move seems well established. The mean free path, which is mainly determined by H-ripplon scattering, is predicted theoretically to be very long.[9] The conditions under which these approximations are valid and the effects of a finite mean free path will be discussed later in the paper.

The most interesting results of our theory have to do with the dependence of $1/T_1$ on the thickness d of the nonmagnetic film and on the temperature T. These dependences are most easily understood if we think of $1/T_1$ as being a sum of contributions of spatial Fourier components, with wave vector $\mathbf{q}$ in the plane of the surface, of the inhomogeneous transverse field due to a magnetic particle a distance d below the surface (see Fig. 1). The magnitude of this field is proportional to e^{-qd}, and an atom moving with velocity $\mathbf{v}$ will see the field fluctuating at a frequency $\omega = \mathbf{q}\cdot\mathbf{v}$. If ω_{ab} is the frequency of the *a*-*b* splitting, then the probability that the atom will see a field oscillating at this frequency is proportional to $e^{-(1/2)\beta m(\omega_{ab}/q)^2}$, where $\beta = 1/k_B T$ and m is the mass of an H atom. Of course, the complete theory involves a configuration average and sum over the positions of the magnetic particle in the substrate.

Our main result can be expressed in terms of the configuration averaged rate of transitions from b to a. The result is

$$\langle R_{b\to a}\rangle_c = (3\pi^2\gamma_\perp^2/8)M_2 a(\theta)\int_0^\infty C_{b,a}(q,0)q^2\,dq \tag{1.1}$$

where $\hbar\gamma_\perp$ is the transition dipole moment for the *a*-*b*

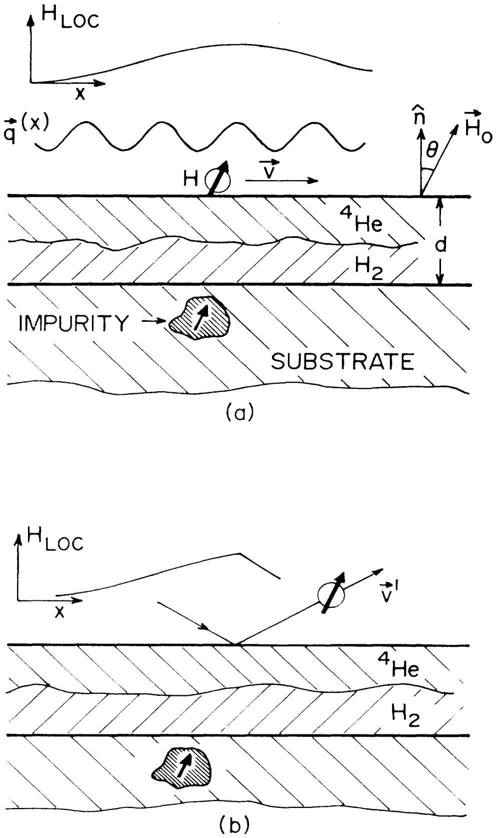

FIG. 1. (a) Situation where a hydrogen atom with velocity v bound to the liquid-helium surface, is moving close to a magnetic impurity in the substrate. The substrate has a coating of solid H_2 and liquid ^{4}He of combined thickness d. Both the magnetic moment of the atom and the impurity magnetic moment lie along the externally applied field $\mathbf{H}_0$. The dependence of the local field $H_{\text{loc}}(x)$ seen by the atom as it moves is shown schematically, together with the spatial dependence of a single Fourier component $H_{\mathbf{q}}(x)$. Transitions between the a and b states occur when $\mathbf{v}\cdot\mathbf{q}=\omega_{ab}$. (b) Same arrangement as in (a) except that the atom belongs to the gas phase and is scattered (reflected) from the surface. Here there is a discontinuity in the slope of H vs x (or t) and it is this feature that gives rise to the ω^{-4} dependence of this relaxation process for large ω. A scattering event for an atom on the surface (e.g., by a ripplon) produces an analogous discontinuity in slope. The corresponding ω^{-4} contribution may dominate the contributions from the collisionless encounters depicted in Fig. 1(a), which have a stronger frequency dependence, $\exp(-\omega^{2/3})$. (See discussion in Sec. IV.)

transition, $a(\theta)$ is a weak function of order unity of the angle θ between the external field and the normal to the surface [see Eq. (2.16)], and M_2 is the average of the squared magnetic moment of the magnetic particles in the substrate per unit volume of substrate, $M_2=(\sum_i m_i^2)/V$.

The correlation function $C_{b,a}(q,0)$ contains two contributions, one from atoms bound to the surface and the other from atoms in the gas which scatter elastically from the surface

$$C_{b,a}(q,0)=C_{b,a}^{(S)}(q,0)+C_{b,a}^{(G)}(q,0)\ . \tag{1.2}$$

Expressions for the two correlation functions on the right-hand side of Eq. (1.2) are given in Eqs. (3.17) and (3.20). For reasonably thick films, the correlation functions are described by the approximate expressions

$$C_{b,a}^{(S)}(q,0)\approx\frac{N_b m\Lambda^2}{\hbar q}\frac{A}{V}e^{\beta E_B}e^{-2qd}e^{-(\beta m/2)(\omega_{ab}/q)^2}\ , \tag{1.3a}$$

$$C_{b,a}^{(G)}(q,0)=\frac{2N_b}{\pi^2}\Lambda^3\frac{A}{V}\frac{q^2e^{-2qd}}{\beta^3\hbar^3\omega_{ab}^4}\ , \tag{1.3b}$$

where Λ is the thermal de Broglie wavelength, N_b is the total number of b atoms in the sample and E_B is the binding energy of a or b atoms to the surface.

Substituting the surface correlation function, Eq. (1.3a) into Eq. (1.1) and dividing by N_b, one obtains the rate of surface $b\rightarrow a$ transitions per b atom,

$$r_{b\rightarrow a}^{(S)}=\frac{3\pi^2\gamma_\perp^2}{8\hbar}M_2 m\Lambda^2\frac{A}{V}a(\theta)e^{\beta E_B}\times\int_0^\infty e^{-2qd}e^{-\beta m\omega_{ab}^2/2q^2}q\,dq\ . \tag{1.4}$$

The q integration in Eq. (1.4) is not analytically tractable. A very rough approximate evaluation may be done as follows. The exponential in Eq. (1.4) is peaked at $q=q_0$ where

$$q_0d=(\beta m\omega_{ab}^2d^2/2)^{1/3}\ . \tag{1.5}$$

If the exponential is replaced by a Gaussian of the appropriate width peaked at q_0, then the temperature dependence at the rate in Eq. (1.4) is roughly $\exp[\beta E_B-3(\beta m\omega_{ab}^2d^2/2)^{1/3}]$. For $\omega_{ab}\sim 2\pi\times10^9\ \text{s}^{-1}$ and $d=300$ Å, the quantity $m\omega_{ab}^2d^2/2$ is equal to 2.15 K whereas E_B is about 1 K, and so the relaxation rate has a minimum somewhat below 1 K. The thickness and frequency dependence are of the form $\exp[-A(\omega_{ab}d)^{2/3}]$, where $A\sim T^{-1/3}$. Thus the relaxation rate for surface atoms, which is almost but not quite exponentially dependent on d, can be greatly suppressed by increasing the thickness of the insulating layer.

The contribution from atoms in the gas which are reflected by the surface has the much weaker frequency dependence $r_{a\rightarrow b}^{(G)}\sim\omega_{ab}^{-4}$ which results from the discontinuous change in velocity of atoms striking the surface. This ω^{-4} behavior for the high-frequency limit of spin relaxation of atoms which are scattered in the presence of an inhomogeneous magnetic field was first considered by Purcell and reported by Kleppner, Goldenberg, and Ramsey[10] in their paper on the theory of the hydrogen maser. Although Purcell's calculation was purely classical, the physical content was identical to that of the quantum-mechanical result derived here. For large layer thickness, the transition rate per b atom of atoms scattered by the surface is approximately

$$r_{b\rightarrow a}^{(G)}=\frac{9\gamma_\perp^2M_2}{16\hbar^3\omega_{ab}^4d^5}\left[\frac{\Lambda}{\beta}\right]^3\frac{A}{V}a(\theta)\ . \tag{1.6}$$

The complete expression is given in Sec. III. The temperature dependence of this contribution is $r_{b\rightarrow a}^{(G)}\sim T^{3/2}$ and hence one would expect this contribution to dominate for large T.

The remainder of this paper consists of a more formal and detailed presentation of the theory whose qualitative features are discussed above. In Sec. II, a general formalism is presented which relates the relaxation rate to the spin-spin correlation function of atoms near the surface. Specific expressions for the spin-spin correlation functions for atoms on or near the liquid He surface are derived in Sec. III in a way which allows for the possibility that the density of a- and b-state atoms might be out of equilibrium. The results are specialized to simple limiting cases, and finally, numerical evaluations are given and compared to the approximate expressions discussed above. Finally, Sec. IV contains a brief discussion of the effects of scattering of surface atoms on the surface spin-relaxation rate.

II. GENERAL FORMALISM

In this section, we derive the expressions for the configuration averaged transition rates $\langle R_{b\to a}\rangle_c$ and $\langle R_{a\to b}\rangle_c$ for H atoms in the two lowest hyperfine states a and b moving on or near a liquid-He-coated surface and interacting with magnetic particles in the substrate. The magnetic particles are assumed to be fully saturated by the external field and are randomly distributed in a half-space which extends to within a distance d below the liquid-He surface. The inert region of thickness d corresponds to the combined thickness of liquid-He film and solid H_2 on the metal substrate.

We consider H atoms moving on or near a liquid-He surface with unperturbed, single-particle energies

$$E_{\alpha,\nu}(\mathbf{k})=-E_B\delta_{\nu,s}+\hbar^2k^2/2m+\alpha\hbar\omega_{ab}/2\ , \tag{2.1}$$

where $\nu=s$ for atoms on the surface or g for atoms in the gas and $\alpha=+1\ (-1)$ for atoms in the b (a) state. $\mathbf{k}$ is a two- or three-dimensional wave vector, as appropriate, for atoms on the surface or in the gas.

The atomic spin states are assumed to be weakly perturbed by their interaction with a dipolar field $[\mathbf{H}_\perp(\mathbf{x}),H_z(\mathbf{x})]$ due to magnetic particles in the substrate. This interaction has the form

$$V=-\int d^3x\,[\hbar\gamma_\perp\mathbf{S}_\perp(\mathbf{x})\cdot\mathbf{H}_\perp(\mathbf{x})+\hbar\gamma_zS_z(\mathbf{x})H_z(\mathbf{x})]\ , \tag{2.2}$$

where $S_z(\mathbf{x})$ and $\mathbf{S}_\perp(\mathbf{x})$ are longitudinal and transverse spin-density operators in the spin space defined by the a and b states,

$$S_z(\mathbf{x})=\tfrac{1}{2}[\psi_b^\dagger(\mathbf{x})\psi_b(\mathbf{x})-\psi_a^\dagger(\mathbf{x})\psi_a(\mathbf{x})]\ , \tag{2.3a}$$

$$S_+(\mathbf{x})=\psi_b^\dagger(\mathbf{x})\psi_a(\mathbf{x})\ , \tag{2.3b}$$

$$S_-(\mathbf{x})=\psi_a^\dagger(\mathbf{x})\psi_b(\mathbf{x})\ , \tag{2.3c}$$

where, as usual, $S_\pm(\mathbf{x})=S_x(\mathbf{x})\pm iS_y(\mathbf{x})$ and $\mathbf{S}_\perp(\mathbf{x})=[S_x(\mathbf{x}),S_y(\mathbf{x}),0]$ and $\psi_\alpha^\dagger(x)$ and $\psi_\alpha(x)$ are field operators which create and destroy atoms in spin state α at position $\mathbf{x}$. Then for a particular configuration of dipolar fields, the rate of transitions from spin state α to state $-\alpha$ is given by

$$R_{\alpha\to-\alpha}=2\pi\hbar(\gamma_\perp^2/4)\int d^3\mathbf{x}d^3\mathbf{x}'\sum_{i,f}P(i)\langle i\,|\,S_\alpha(\mathbf{x})H_{-\alpha}(\mathbf{x})\,|\,f\rangle\langle f\,|\,S_{-\alpha}(\mathbf{x}')H_\alpha(\mathbf{x}')\,|\,i\rangle\delta(E_i-E_f)\ , \tag{2.4}$$

where $H_\alpha(\mathbf{x})=H_x(\mathbf{x})+i\alpha H_y(\mathbf{x})$ and $P(i)$ is the probability that the system is in initial state i. Note that $P(i)$ need not correspond to an equilibrium ratio of a and b atom populations. In particular, we will assume separate chemical potentials μ_a and μ_b for the two species in order to describe the effects of the T_1 bottleneck.

In the usual way, Eq. (2.4) can be rewritten as the temporal Fourier transform of a two time correlation function

$$R_{\alpha\to-\alpha}=(\gamma_\perp^2/4)\int dt\int d^3\mathbf{x}\int d^3\mathbf{x}'H_{-\alpha}(\mathbf{x})H_\alpha(\mathbf{x}')\times\langle S_\alpha(\mathbf{x},t)S_{-\alpha}(\mathbf{x}',0)\rangle\ , \tag{2.5}$$

where $\langle\cdots\rangle$ is a thermal average over the distribution $P(i)$. The inhomogeneous field due to a density distribution $M_I(\mathbf{y})$ of fully saturated magnetic particles in the substrate is

$$H_\alpha(\mathbf{x})=-(24\pi/5)^{1/2}\int d^3yM_I(\mathbf{y})Y_2^\alpha(\mathbf{x}-\mathbf{y})/\,|\,\mathbf{x}-\mathbf{y}\,|^3\ , \tag{2.6}$$

where

$$M_I(\mathbf{y})=\sum_i m_i\delta(\mathbf{y}-\mathbf{R}_i)\ , \tag{2.7}$$

and m_i is the moment of the particle at $\mathbf{R}_i$, a random position in the substrate. The transition rate, Eq. (2.5), thus involves the quantity $M_I(\mathbf{y})M_I(\mathbf{y}')$, and the configuration averaged rate $\langle R_{\alpha\to-\alpha}\rangle_c$, for uncorrelated impurity positions, depends on

$$\langle M_I(\mathbf{y})M_I(\mathbf{y}')\rangle_c=V^{-2}\left[\left[\sum_i m_i\right]^2-\sum_i m_i^2\right]+\delta(\mathbf{y}-\mathbf{y}')V^{-1}\sum_i m_i^2\ , \tag{2.8}$$

where V is the volume of the substrate. The first term in Eq. (2.8) has no spatial dependence and hence will not contribute to the relaxation rate. The second term involves the quantity

$$M_2\equiv V^{-1}\sum_i m_i^2\ , \tag{2.9}$$

the average squared moment per unit volume of substrate. The configuration averaged relaxation rate is then

$$\langle R_{\alpha\to-\alpha}\rangle_c=(6\pi/5)\gamma_1^2M_2\int dt\int d^3xd^3x'd^3y\frac{Y_2^\alpha(\mathbf{y}-\mathbf{x})^*}{|\mathbf{y}-\mathbf{x}|^3}\frac{Y_2^\alpha(\mathbf{y}-\mathbf{x}')}{|\mathbf{y}-\mathbf{x}'|^3}\langle S_\alpha(\mathbf{x},t)S_{-\alpha}(\mathbf{x}',0)\rangle\ . \tag{2.10}$$

The **y** integral is most easily done in a coordinate system with the origin at the surface and with the y_3 axis normal to the surface. Then

$$\frac{Y_2^\alpha(\mathbf{y}-\mathbf{x})^*Y_2^\alpha(\mathbf{y}-\mathbf{x}')}{|\mathbf{y}-\mathbf{x}|^3|\mathbf{y}-\mathbf{x}'|^3}\to\sum_{m,n}D^{(2)}_{\alpha,m}(0,\theta,0)^*D^{(2)}_{-\alpha,n}(0,\theta,0)\frac{Y_2^m(\mathbf{y}-\mathbf{x})^*Y_2^n(\mathbf{y}-\mathbf{x}')}{|\mathbf{y}-\mathbf{x}|^3|\mathbf{y}-\mathbf{x}'|^3}\ , \tag{2.11}$$

where θ is the angle between the magnetic field and the normal to the surface, and vectors on the right-hand side are measured in the new coordinate system. Defining

$$I_{m,n}(\mathbf{x},\mathbf{x}')=\int d^3y\frac{Y_2^m(\mathbf{y}-\mathbf{x})^*}{|\mathbf{y}-\mathbf{x}|^3}\frac{Y_2^n(\mathbf{y}-\mathbf{x}')}{|\mathbf{y}-\mathbf{x}'|^3}\ , \tag{2.12}$$

where the integral is over the half space $y_3<0$, we find that if x_3 and x'_3 are positive,

$$I_{m,n}(\mathbf{x},\mathbf{x}')=\tfrac{5}{32}A_m A_n\int q_\perp d^2q_\perp e^{i(\mathbf{x}-\mathbf{x}')\cdot\mathbf{q}_\perp}\times e^{i(m-n)\theta_q}e^{-q_\perp(x_3+x'_3)}\ , \tag{2.13}$$

where $A_0=1$, $A_{\pm1}=\sqrt{2/3}$, $A_{\pm2}=1/\sqrt{6}$, $\mathbf{q}_\perp$ is a vector in the plane of the surface and θ_q is the angle between $\mathbf{q}_\perp$ and the y_1 direction which is perpendicular to the external magnetic field and to the normal to the surface. Then

$$\langle R_{\alpha\to-\alpha}\rangle_c=(3\pi/16)\gamma_1^2M_2\sum_{m,n}A_m A_nD^{(2)}_{\alpha,m}(0,\theta,0)^*D^{(2)}_{\alpha,n}(0,\theta,0)\times\int dt\int q_\perp d^2q_\perp e^{i(m-n)\theta_q}\left[\int d^3xd^3x'e^{-q_\perp(x_3+x'_3)}e^{i(\mathbf{x}-\mathbf{x}')\cdot\mathbf{q}_\perp}\langle S_\alpha(\mathbf{x},t)S_{-\alpha}(\mathbf{x}',0)\rangle\right]\ . \tag{2.14}$$

An important simplification results from the fact that the factor in square brackets, the unperturbed spin-spin correlation function, does not depend on the direction of $\mathbf{q}_\perp$. Therefore the angular part of the $\mathbf{q}_\perp$ integral can be performed, yielding

$$\langle R_{\alpha\to-\alpha}\rangle_c=(3\pi^2/8)\gamma_1^2M_2a(\theta)\int q_\perp^2dq_\perp\int dt\int d^3xd^3x'e^{i\mathbf{q}_\perp\cdot(\mathbf{x}-\mathbf{x}')}e^{-q_\perp(x_3+x'_3)}\langle S_\alpha(\mathbf{x},t)S_{-\alpha}(\mathbf{x}',0)\rangle\ , \tag{2.15}$$

where

$$a(\theta)=\sum_m A_m^2\,|D^{(2)}_{1,m}(0,\theta,0)|^2 \tag{2.16}$$

is the weakly angular-dependent function

$$a(\theta)=\tfrac{3}{8}(\sin2\theta)^2+\tfrac{1}{3}(4\cos^4\theta-3\cos^2\theta+1)+\tfrac{1}{12}(1+\cos^2\theta)\sin^2\theta\ . \tag{2.17}$$

A plot of $a(\theta)$ is given in Fig. 2.

III. SPIN-SPIN CORRELATION FUNCTIONS

A. Relationship between the correlation function and the susceptibility

We would like to relate the correlation function $\langle S_\alpha(\mathbf{x},t)S_{-\alpha}(\mathbf{x}',0)\rangle$ to the time and space dependent susceptibility which describes the linear response of the magnetization to a weak perturbing magnetic field. Our approach is based on that of Kadanoff and Baym,[11] but is modified to describe the "nonequilibrium" situation in which the a and b atoms have different chemical potentials. We begin by defining the imaginary time Green's function

$$L_{\alpha,-\alpha}(1,2)=-i\langle T(S_\alpha(1)S_{-\alpha}(2))\rangle\ , \tag{3.1}$$

where $(1)=(\mathbf{x},t)$, $(2)=(\mathbf{x}',t')$, and T is a time-ordering operator:

$$L_{\alpha,-\alpha}(1,2)=L^>_{\alpha,-\alpha}(1,2)=-i\langle S_\alpha(1)S_{-\alpha}(2)\rangle \quad \text{for } t_1>t_2\ , \tag{3.2a}$$

$$L_{\alpha,-\alpha}(1,2)=L^<_{\alpha,-\alpha}(1,2)=-i\langle S_{-\alpha}(2)S_\alpha(1)\rangle \quad \text{for } t_1<t_2\ . \tag{3.2b}$$

It is then straightforward to show that $L_{\alpha,-\alpha}$ satisfies the

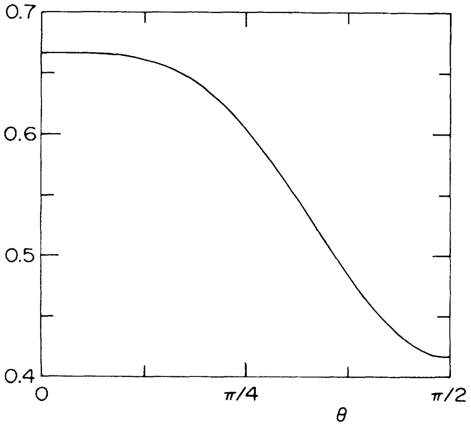

FIG. 2. Plot of the function $a(\theta)$ defined in Eq. (2.17), which describes the dependence of the impurity relaxation on the angle between the substrate normal and the applied magnetic field. The magnetization of the impurity has been assumed to be saturated and always along **H**.

boundary condition

$$L_{\alpha,-\alpha}(1,2)\,|_{t_1=0}=e^{\beta(\mu_{-\alpha}-\mu_\alpha)}L_{\alpha,-\alpha}(1,2)\,|_{t_1=-i\beta}\ . \tag{3.3}$$

To check this, note that $L_{\alpha,-\alpha}(1,2)$ has the same form as $G_{-\alpha}(1,2)=-i\langle T(\psi_{-\alpha}(1)\psi^{+}_{-\alpha}(2))\rangle$, with respect to the field operators $\psi_{-\alpha}$.

Fourier transforming in the time variable, Eq. (3.3) implies that

$$\begin{aligned}L^{<}_{\alpha,-\alpha}(\mathbf{x},\mathbf{x}';\omega)&=i\int_{-\infty}^{\infty}dte^{i\omega t}L^{<}_{\alpha,-\alpha}(\mathbf{x},t;\mathbf{x}',0)\\&=e^{-\beta(\hbar\omega-\mu_{-\alpha}+\mu_\alpha)}L^{>}_{\alpha,-\alpha}(x,x';\omega)\ .\end{aligned} \tag{3.4}$$

The Fourier transform of the Green's function has the spectral representation

$$L_{\alpha,-\alpha}(\mathbf{x},\mathbf{x}';\Omega)=\int\frac{d\omega}{2\pi}\frac{L^{>}_{\alpha,-\alpha}(\mathbf{x},\mathbf{x}';\omega)-L^{<}_{\alpha,-\alpha}(\mathbf{x},\mathbf{x}';\omega)}{\Omega-\omega} \tag{3.5}$$

for arbitrary complex Ω. Then for real Ω,

$$\begin{aligned}L^{>}_{\alpha,-\alpha}(\mathbf{x},\mathbf{x}';\Omega)&=\frac{i}{1-e^{-\beta(\hbar\Omega-\mu_{-\alpha}+\mu_\alpha)}}\\&\quad\times[L_{\alpha,-\alpha}(\mathbf{x},\mathbf{x}';\Omega+i\epsilon)\\&\qquad-L_{\alpha,-\alpha}(x,x';\Omega-i\epsilon)]\\&=\frac{-2\,\mathrm{Im}L_{\alpha,-\alpha}(\mathbf{x},\mathbf{x}';\Omega+i\epsilon)}{1-e^{-\beta(\hbar\Omega-\mu_{-\alpha}+\mu_\alpha)}}\ .\end{aligned} \tag{3.6}$$

Kadanoff and Baym also show that the Green's function $L_{\alpha,-\alpha}(\mathbf{x},\mathbf{x}';\Omega)$ for Ω in the upper half of the complex plane describes the linear response of the system to a weak external field. That is, $L_{\alpha,-\alpha}$ is the susceptibility $\chi_{\alpha,-\alpha}$. Combining Eqs. (3.3), (3.4), and (3.6) yields

$$\int_{-\infty}^{\infty}dte^{i\omega t}\langle S_\alpha(\mathbf{x},t)S_{-\alpha}(\mathbf{x}',0)\rangle=-\frac{\mathrm{Im}\chi_{\alpha,-\alpha}(\mathbf{x},\mathbf{x}';\omega)}{1-e^{-\beta(\hbar\omega-\mu_{-\alpha}+\mu_\alpha)}}\ . \tag{3.7}$$

Thus the correlation function on the left-hand side of Eq. (3.7) can be calculated from the Green's function, Eq. (3.1), or it can be obtained by calculating the susceptibility from a Boltzmann equation for the spin density.[12] Alternatively, the left-hand side of (3.7) may be calculated directly from

$$\begin{aligned}\int_{-\infty}^{\infty}&dte^{i\omega t}\langle S_\alpha(\mathbf{x},t)S_{-\alpha}(\mathbf{x}',0)\rangle\\&=2\pi\hbar\sum_{i,f}P(i)\langle i\,|S_\alpha(\mathbf{x})|\,f\rangle\\&\qquad\times\langle f\,|S_{-\alpha}(\mathbf{x}')|\,i\rangle\delta(\hbar\omega+E_i-E_f)\ ,\end{aligned} \tag{3.8}$$

which is analogous to Eq. (2.4).

In the following subsections, we evaluate the spin-spin correlation function of noninteracting atoms for the two cases of interest: (i) atoms in the surface state and (ii) atoms which are scattered quasielastically by the surface. In each case, we calculate the correlation function

$$\begin{aligned}C_{\alpha,-\alpha}(\mathbf{q},\omega)=\int_{-\infty}^{\infty}&dt\,e^{i\omega t}\int d^3xd^3x'\\&\times e^{i\mathbf{q}\cdot(\mathbf{x}-\mathbf{x}')}e^{-q(x_3+x_3')}\\&\times\langle S_\alpha(\mathbf{x},t)S_{-\alpha}(\mathbf{x}',0)\rangle\ ,\end{aligned} \tag{3.9}$$

where $\mathbf{q}$ is a two-dimensional wave vector. In terms of this quantity,

$$\langle R_{\alpha\to-\alpha}\rangle_c=(3\pi^2/8)\gamma_1^2M_2a(\theta)\int_0^\infty q^2C_{\alpha,-\alpha}(q,0)dq\ . \tag{3.10}$$

B. Free surface atoms

Surface atoms are described by a product wave function which is a plane wave for motion along the surface and a bound-state wave function $\Theta_s(x_3-d)$ for motion normal to the surface. The form of the argument of Θ_s, x_3-d, is chosen to make Θ_s independent of the thickness d of the helium film. (Recall that the origin of x_3 is the surface of the substrate. Thus d represents the thickness of an insulating layer which is free of magnetic impurities.) In this basis, the field operator may be written as

$$\psi_\alpha(\mathbf{x},t)=A^{1/2}\int\frac{d^2k}{(2\pi)^2}e^{i\mathbf{k}\cdot\mathbf{x}}e^{-iE_{\alpha,s}(\mathbf{k})t/\hbar}\Theta_s(x_3-d)c_{\alpha,s}(\mathbf{k}) \tag{3.11}$$

where $c_{\alpha,s}(\mathbf{k})$ is a Bose annihilation operator. The correlation function [Eq. (3.9)] may now be calculated directly from Eqs. (2.3), (3.8), and (3.11). The result is

$$\begin{aligned}C^{(S)}_{\alpha,-\alpha}(\mathbf{q},\omega)=2\pi\hbar Ae^{-2qd}\left[\int_{-\infty}^{\infty}dz\,|\Theta_s(z)|^2e^{-qz}\right]^2\int\frac{d^2k}{(2\pi)^2}n_\alpha[E_{\alpha,s}(\mathbf{k})]\{1+n_{-\alpha}[E_{-\alpha,s}(\mathbf{k}-\mathbf{q})]\}\\\times\delta[\hbar\omega+E_{\alpha,s}(\mathbf{k})-E_{-\alpha,s}(\mathbf{k}-\mathbf{q})]\ ,\end{aligned} \tag{3.12}$$

where A is the area of the surface, and

$$n_\alpha[E_{\alpha,\nu}(\mathbf{k})]=(e^{\beta[E_{\alpha,\nu}(k)-\mu_\alpha]}-1)^{-1}\ . \tag{3.13}$$

In this paper, we will ignore the q dependence of the form factor $\int_{-\infty}^{\infty}dz\,|\Theta_s(z)|^2e^{-qz}$ by setting $|\Theta_s(z)|^2=\delta(z)$ so that the integral is equal to unity.

It is useful and instructive also to derive the correlation function from the spin density response function $\chi^{(0)}_{\alpha,-\alpha}(\mathbf{q},\omega)$, which is a straightforward generalization of the density response function $L_0(\mathbf{q},\omega)$ derived in the appendix of Kadanoff and Baym.[11]

$$\chi^{(0)}_{\alpha,-\alpha}(\mathbf{q},\omega)=\hbar A e^{-2qd}\int\frac{d^2k}{(2\pi)^2}\frac{n_\alpha[E_{\alpha,s}(\mathbf{k})]-n_{-\alpha}[E_{-\alpha,s}(\mathbf{k}-\mathbf{q})]}{\hbar\omega+E_{\alpha,s}(\mathbf{k})-E_{-\alpha,s}(\mathbf{k}-\mathbf{q})}\ . \tag{3.14}$$

Then, following Eq. (3.6),

$$\begin{aligned}C^{(S)}_{\alpha,-\alpha}(\mathbf{q},\omega)&=\frac{-2\,\mathrm{Im}\chi^{(0)}_{-\alpha,\alpha}(\mathbf{q},\omega+i\epsilon)}{1-e^{-\beta(\hbar\omega-\mu_{-\alpha}+\mu_\alpha)}}\\&=2\pi\hbar A e^{-2qd}\int\frac{d^2k}{(2\pi)^2}\frac{n_\alpha[E_{\alpha,s}(\mathbf{k})]-n_{-\alpha}[E_{-\alpha,s}(\mathbf{k}-\mathbf{q})]}{1-e^{-\beta(\hbar\omega-\mu_{-\alpha}+\mu_\alpha)}}\delta[\hbar\omega+E_{\alpha,s}(\mathbf{k})-E_{-\alpha,s}(\mathbf{k}-\mathbf{q})]\ ,\end{aligned} \tag{3.15}$$

and Eq. (3.12) follows directly by using the δ function to eliminate $\hbar\omega$ from the denominator on the right-hand side of (3.15).

Equation (3.15) is particularly convenient for performing the d^2k integral. For the rest of this paper, we will assume that the surface density of atoms is low, $e^{\beta\mu_\alpha}\ll 1$, and hence that

$$n_\alpha(E_{\alpha,\nu}(\mathbf{k}))\approx e^{\beta\mu_\alpha}e^{-\beta E_{\alpha,\nu}(\mathbf{k})}\ . \tag{3.16}$$

Then

$$C^{(S)}_{\alpha,-\alpha}(\mathbf{q},\omega)=\frac{mA}{\Lambda\hbar q}e^{-2qd}e^{\beta[\mu_\alpha+E_B-(\alpha/2)\hbar\omega_{ab}]}\times e^{-(\beta m/2q^2)(\omega+\alpha\omega_{ab}-\hbar q^2/2m)^2}\ . \tag{3.17}$$

Simple limiting cases of this expression will be discussed in Sec. III D below.

C. Gas phase atoms which are scattered by the surface

To simplify this part of the calculation, we treat the surface as a flat hard wall. The field operator is then

$$\psi_\alpha(\mathbf{x},t)=\sqrt{2AL}\int\frac{d^2k}{(2\pi)^2}\int_0^\infty\frac{dk_z}{\pi}e^{i\mathbf{k}\cdot\mathbf{x}}\sin(zk_z)\times e^{-iE_\alpha(\mathbf{k},k_z)t/\hbar}c_\alpha(\mathbf{k},k_z)\ , \tag{3.18}$$

where L is the size of the sample in the direction perpendicular to the surface. The correlation function for the gas phase atoms is then

$$\begin{aligned}C^{(G)}_{\alpha,-\alpha}(q,\omega)=\frac{2\pi\hbar A e^{-2qd}}{1-e^{-\beta(\hbar\omega-\mu_{-\alpha}+\mu_\alpha)}}&\int_0^\infty\frac{dk_z}{\pi}\int_0^\infty\frac{dp_z}{\pi}\left[\frac{q}{(k_z-p_z)^2+q^2}-\frac{q}{(k_z+p_z)^2+q^2}\right]^2\\&\times\int\frac{d^2k}{(2\pi)^2}\{n_\alpha[E_\alpha(\mathbf{k},k_z)]-n_{-\alpha}[E_{-\alpha}(\mathbf{k}-\mathbf{q},p_z)]\}\\&\times\delta[\hbar\omega+E_\alpha(\mathbf{k},k_z)-E_{-\alpha}(\mathbf{k}-\mathbf{q},p_z)]\ .\end{aligned} \tag{3.19}$$

Using the result, Eq. (3.17), in the low-density limit, we find

$$\begin{aligned}C^{(G)}_{\alpha,-\alpha}(\mathbf{q},\omega)=\frac{mA}{\Lambda\hbar q}e^{-2qd}e^{\beta[\mu_\alpha-(\alpha/2)\hbar\omega_{ab}]}&\int_0^\infty\frac{dk_z}{\pi}\int_0^\infty\frac{dp_z}{\pi}\left[\frac{q}{(k_z-p_z)^2+q^2}-\frac{q}{(k_z+p_z)^2+q^2}\right]^2\\&\times e^{-\beta\hbar^2k_z^2/2m}e^{(-\beta m/2q^2)[\omega+\alpha\omega_{ab}+(\hbar/2m)(k_z^2-p_z^2-q^2)]^2}\ .\end{aligned} \tag{3.20}$$

The integrals in Eq. (3.20) are not particularly tractable. The limiting cases of interest will be treated below.

D. Simple limiting cases

To simplify the discussion, we specialize to the case of large d, so that the factor e^{-2qd} restricts q to be small. Then we can neglect $\hbar q^2/2m$ compared to ω_{ab}. The rate of transitions from b to a depends on the two correlation functions

$$C^{(S)}_{b,a}(q,0)=\frac{mA}{\Lambda\hbar q}e^{-2qd}e^{\beta[E_B-(1/2)\hbar\omega_{ab}+\mu_b]}e^{-(\beta m/2)(\omega_{ab}/q)^2} \tag{3.21}$$

and

$$C_{b,a}^{(G)}(q,0)=\frac{mA}{\Lambda\hbar q}e^{-2qd}e^{\beta[\mu_b-(1/2)\hbar\omega_{ab}]}\int_0^\infty\frac{dk_z}{\pi}\int_0^\infty\frac{dp_z}{\pi}\left[\frac{q}{(k_z-p_z)^2+q^2}-\frac{q}{(k_z+p_z)^2+q^2}\right]^2$$
$$\times e^{-\beta\hbar^2k_z^2/2m}e^{(-\beta m/2q^2)[\omega_{ab}+(\hbar/2m)(k_z^2-p_z^2)]^2}\ . \tag{3.22}$$

We evaluate the integrals in Eq. (3.22) approximately as follows. If q is sufficiently small, then the Gaussian factor on the right can be approximated by a δ function:

$$e^{(-\beta m/2q^2)[\omega_{ab}+(\hbar/2m)(k_z^2-p_z^2)]^2}\rightarrow\left[\frac{2\pi q^2}{\beta m}\right]^{1/2}\delta[\omega_{ab}+(\hbar/2m)(k_z^2-p_z^2)]\ . \tag{3.23}$$

Then q^2 can be neglected compared to $k_z^2\pm p_z^2$ and Eq. (3.22) is approximately given by

$$C_{b,a}^{(G)}(q,0)\approx\frac{mAq^2}{\hbar^2\beta}e^{-2qd}e^{\beta[\mu_b-(1/2)\hbar\omega_{ab}]}\int_0^\infty\frac{dk_z}{\pi}\int_0^\infty\frac{dp_z}{\pi}\frac{(4k_zp_z)^2}{(k_z^2-p_z^2)^4}e^{-\beta\hbar^2k_z^2/2m}\delta[\omega_{ab}+(\hbar/2m)(k_z^2-p_z^2)]$$
$$=\frac{2}{\pi^2}\frac{Aq^2e^{-2qd}e^{\beta(\mu_b-\frac{1}{2}\hbar\omega_{ab})}}{\beta^3\hbar^3\omega_{ab}^4}\left[\frac{\beta\hbar\omega_{ab}}{2}K_1\left[\frac{\beta\hbar\omega_{ab}}{2}\right]\right]\ , \tag{3.24}$$

where K_1 is a modified Bessel function. The factor $[\cdots]$ on the right-hand side of (3.24) is equal to 1 for $\beta\hbar\omega_{ab}/2\ll 1$.

The factor $e^{\beta(\mu_b-\hbar\omega_{ab}/2)}$ is determined by

$$N_b=(e^{\beta E_B}A/\Lambda^2+V/\Lambda^3)e^{\beta(\mu_b-\hbar\omega_{ab}/2)} \tag{3.25}$$

or

$$e^{\beta(\mu_b-\hbar\omega_{ab}/2)}=\frac{N_b\Lambda^3/V}{(\Lambda A/V)e^{\beta E_B}+1}\ , \tag{3.26}$$

where N_b is the total number of b atoms in the sample.

At moderately high temperatures, when most of the atoms are in the gas

$$e^{\beta(\mu_b-\hbar\omega_{ab}/2)}\approx N_b\Lambda^3/V\ . \tag{3.27}$$

In this limit, the surface and gas correlation functions for $\omega=0$ are given by Eqs. (1.3a) and (1.3b).

E. Numerical evaluation of the relaxation rates

The complete result for the surface and gas relaxation rates is obtained by substituting Eqs. (3.17) and (3.20) for the correlation functions into the integral of Eq. (3.10). Except for the assumed form of the wave functions, Eqs. (3.11) and (3.18), the only approximation which has been made is that the H gas is nondegenerate. Then the b-to-a transition rate is

$$R_{b\rightarrow a}=(3\pi^2/8)\gamma_1^2M_2a(\theta)(mA/\Lambda\hbar)e^{\beta(\mu_b-\hbar\omega_{ab}/2)}$$
$$\times\Bigg[\left[e^{\beta E_B}\int_0^\infty qdqe^{-2qd}e^{-\beta m(\omega_{ab}-\hbar q^2/2m)^2/2q^2}\right]$$
$$+\int_0^\infty q\,dq\,e^{-2qd}\int_0^\infty\frac{dk}{\pi}\int_0^\infty\frac{dq}{\pi}\left[\frac{q}{(k-p)^2+q^2}-\frac{q}{(k+p)^2+q^2}\right]^2$$
$$\times e^{-\beta\hbar^2k^2/2m}e^{-\beta m[\omega_{ab}+\hbar(k^2-p^2-q^2)/2m]^2/2q^2}\Bigg]\ . \tag{3.28}$$

If the fraction of atoms on the surface is small, one can make the further approximation, Eq. (3.27). It is also useful to change integration variables so that the various wave vectors are dimensionless, $\beta\hbar^2q^2/2m\rightarrow q^2$, etc., and to define

$$\Omega=\beta\hbar\omega_{ab}\ , \tag{3.29a}$$

$$d_{\text{eff}}=(2md^2/\beta\hbar^2)^{1/2}\ . \tag{3.29b}$$

Then

$$R_{b\rightarrow a}/N_b=r_0G(\beta E_B,d_{\text{eff}},\Omega)\ , \tag{3.30}$$

where

$$r_0=(3\pi^3/2)\gamma_1^2M_2a(\theta)mA/\hbar V\ , \tag{3.31}$$

$$G(\beta E_B,d_{\text{eff}},\Omega)=e^{\beta E_B}\int_0^\infty q\,dq e^{-2d_{\text{eff}}q}e^{-(\Omega-q^2)^2/4q^2}$$
$$+\int_0^\infty q\,dq\,e^{-2d_{\text{eff}}q}\int_0^\infty \frac{dk}{\pi}\int_0^\infty \frac{dp}{\pi}\left[\frac{q}{(k-p)^2+q^2}-\frac{q}{(k+p)^2+q^2}\right]^2 e^{-k^2}$$
$$\times e^{-[\Omega+k^2-p^2-q^2]^2/4q^2}\,. \qquad (3.32)$$

Note that all of the temperature dependence is contained in the dimensionless function $G(\beta E_B,d_{\text{eff}},\Omega)$. The prefactor r_0 is sensitive to the experimental parameters such as the size, shape, and magnetic inhomogeneity of the substrate and also, through $\gamma_\perp^2 a(\theta)$, to the magnitude and direction of the applied magnetic field. The ESR experiments of Ref. 7 were performed in a field of 40 kG, and hence $\gamma_\perp=\gamma_p+\epsilon\gamma_e\approx 5.2\gamma_p$. The A/V ratio was 30 cm^{-1}.

In order to fit the one-body relaxation data, it was necessary to assume an iron impurity of 0.1 at. % in the form of 75-Å-diameter particles. The mean-square moment per unit volume may be written as

$$M_2=(\mu_{\text{Fe}}n)^2/Nv_a\ , \qquad (3.33)$$

where $\mu_{\text{Fe}}=2.22\mu_B$ is the magnetic moment of an iron atom,[13] n is the number of iron atoms in a cluster, N is the number of copper atoms in a region containing one iron cluster, so that $X=n/N$ is the fraction of iron atoms in the substrate, and v_a is the atomic volume of copper which is essentially identical to that of iron,[13] $1/v_a=8.5\times10^{22}$ cm^{-3}. For 0.1% of iron atoms in the form of 75-Å particles, the dimensionless factor $nX=18.8$, and $M_2=\mu_{\text{Fe}}^2 nX/v_A$. Thus for the parameters of Ref. 7, the rate $r_0=29a(\theta)$ s^{-1}. From Fig. 2, we see that an angular average of $a(\theta)$ will yield a value of 0.3–0.4 and hence the average relaxation time $1/\langle r_0\rangle$ is about 0.1 s for these parameters.

Next, we consider the dimensionless function $G(\beta E_B,d_{\text{eff}},\Omega)$. The qualitative behavior of this function is as follows. For fixed d and ω_{ab}, G increases rapidly as a function of increasing β. We argued in the Introduction that this increase goes roughly as $\exp[\beta E_B-3(\beta m\omega_{ab}^2d^2/2)^{1/3}]$ at low T where the relaxation is predominantly due to atoms on the surface. At higher T, the contribution of atoms in the gas dominates and G increases with increasing T. A numerical evaluation of Eq. (3.32) is shown in Fig. 3 for $d=300$ Å, $\omega_{ab}=2\pi\times10^9$ s^{-1} and $E_B=1$ K. We note that the effective activation energy, which is about 0.88 K at $T=0.1$ K, is not nearly as small as that of the approximate formula given above. This means that the Gaussian approximation to Eq. (1.4) is not very accurate for these parameters.

The thickness dependence of the relaxation rate is shown in Fig. 4 plotted as $\ln G$ versus d, for $T=0.3$ K where the surface contribution is dominant, and $\omega_{ab}=2\pi\times10^9$ s^{-1}. The curve fits very well to a function of the form $A\exp(-Bd^x)$ with $A=17.6$, $B=0.838$ for d in Å, and $x=0.45$. The exponent x is significantly smaller than the value $\frac{2}{3}$ mentioned above.

IV. DISCUSSION

In this section, we discuss the relevance of the T_1 mechanism derived above to the physical system of H atoms in the presence of liquid-He surfaces. Consider first the contribution corresponding to gas atoms which scatter elastically from the surface. The main approximations made in deriving Eq. (3.20) were that the surface acts like a hard wall and also that the motion is otherwise free, i.e., that the atom does not collide with other H atoms or He atoms in the gas. This second assumption corresponds to moderately low temperatures and densities of H atoms. The hard wall approximation should be valid as long as the region of the actual potential well [e.g., the region in which $V(z)<-kT$] is very small compared to Λ.

The distinctive frequency dependence of the relaxation rate for atoms reflected by the surface, $1/T_1\sim\omega_{ab}^{-4}$, is a much more slowly varying function of ω_{ab} than the rate

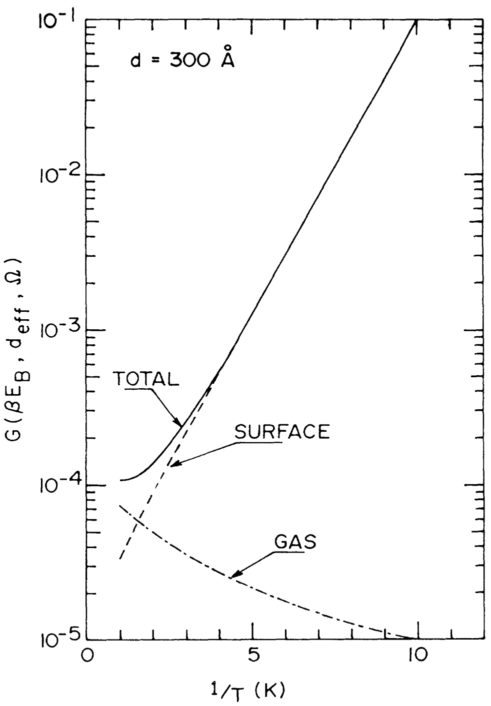

FIG. 3. Dimensionless relaxation rate $G(\beta E_B,d_{\text{eff}},\Omega)$, defined in Eq. (3.32), plotted vs inverse temperature, for $E_B=1$ K, $d=300$ Å and $\omega_{Ab}=2\pi\times10^9$ s^{-1}. The solid line is the total rate. The dashed and dash-dot curves are the surface and gas contributions, respectively.

for atoms moving freely on the surface ($1/T_1 \sim \exp[-A(\omega_{ab}d)^{2/3}]$). This suggests that the assumption of free-particle motion on the surface must be looked at carefully, particularly for thick films. If an atom is scattered, for example, by emitting or absorbing a ripplon while moving through the inhomogeneous field of a magnetic impurity, then the time derivative of the transverse field will change more or less discontinuously, giving rise to an ω_{ab}^{-4} frequency dependence for the relaxation. Thus the assumption of free particle motion might be inappropriate unless the mean free path of the atom is much larger than $1/q_0$, where q_0 is the most effective wave vector defined in Eq. (1.5). Typical experimental values of q_0 are of order $10^{-2} A^{-1}$. Zimmerman and Berlinsky[9] have calculated the mean free path l for ripplon emission by H on liquid He. They find $l > 10^3$ Å for $T < 0.3$ K and $l > 10^4$ Å for $T < 0.1$ K. Therefore the free-particle picture is probably valid at least for $T \lesssim 0.3$ K.

The effect of scattering by ripplons may be calculated as follows. Equation (3.7) relates the spin-spin correlation function to the imaginary part of the transverse spin susceptibility. The effect of scattering on the spin susceptibility may be calculated using the Boltzmann equation. In particular, if the scattering is treated as a perturbation, then, to first order, the effect of scattering is to add a term proportional to $(\omega - \omega_{ab})^{-4}$ to $\mathcal{X}(\omega)$ and hence a contribution which goes like ω_{ab}^{-4} to the spin relaxation rate.[14] The physical interpretation is the same as for scattering from the surface.

A less important effect than ripplon scattering is the finite time that an H atom spends on the surface. Since atoms presumably leave the surface after absorbing one or more high-energy ripplons, such events are relatively rare compared to those which simply change the momentum of an atom bound to the surface. The mean time between adsorption and desorption for an atom is given by

$$\tau_s = (4\Lambda / S\bar{v}) e^{\beta E_B} ,$$

where S, the sticking probability, is of order 2–4 % and $\bar{v}$ is the average speed of an atom in the gas. For $T < 0.3$ K, $\tau_s > 10^{-7}$ s and $\omega_{ab}\tau_s > 200\pi$. Hence τ_s can safely be treated as infinite in this temperature range.

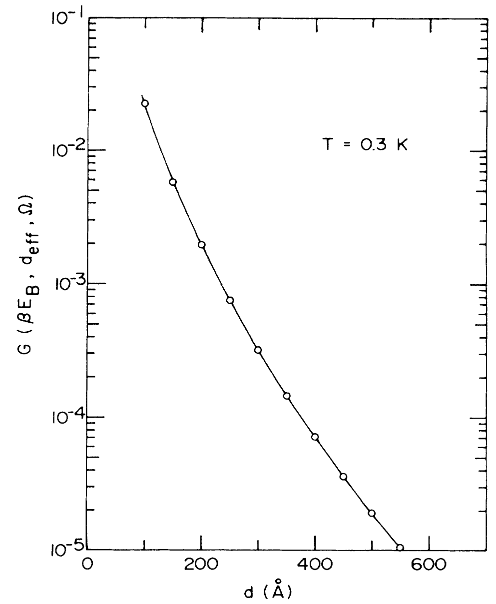

FIG. 4. Dimensionless relaxation rate $G(\beta E_B, d_{\rm eff}, \Omega)$ for $E_B = 1$ K, $\omega_{ab} = 2\pi \times 10^9$ s^{-1} and $T = 0.3$ K vs the thickness d of the nonmagnetic layer.

ACKNOWLEDGMENTS

The authors would like to acknowledge several useful conversations with Myer Bloom. We also thank André Ruckenstein for sending us a copy of his work on a closely related subject[15] before publication and for his helpful comments and suggestions. This work has been supported by grants from the Natural Science and Engineering Research Council (NSERC) of Canada.

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