Spin Transport in Ferromagnetic Metals

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Ferromagnetism is a collective phenomenon, but some properties of ferromagnetic metals can be well understood in terms of a single-particle band picture. We investigate transport phenomena in ferromagnetic metals by means of a simple, solvable model in which the magnetic electrons are independent except for spin-spin correlations generated by an effective exchange field similar to a molecular field. The explicit solution shows that spin transport is reduced to negligible proportions, while charge transport remains the same as in the independent-particle case.

I. INTRODUCTION

IN the case where the relevant wavelengths are long compared to the lattice spacing, one can derive the equation of motion for the macroscopic magnetization $\mathbf{M}$ by equating $(d/dt)(-\mathbf{M}/\gamma)$ to the torque per unit volume and adding a relaxation term[1]:

$$d\mathbf{M}/dt = -\gamma\mathbf{M}\times\mathbf{H} - (2A\gamma/M_s^2)\mathbf{M}\times\nabla^2\mathbf{M}$$
$$- (\lambda/\gamma M_s^2)\mathbf{M}\times d\mathbf{M}/dt, \quad (1)$$

where $\gamma = g|e|/2mc$, A is the macroscopic exchange stiffness constant, M_s is the saturation magnetization, and λ specifies a spin-lattice torque responsible for magnetic relaxation. In at least some metals, the magnetic electrons are highly itinerant; so a spin-transport term must be added to the right side of the above equation. We propose to examine this term and to show, by explicit solution of a simplified model, that it is generally reduced to negligible proportions by exchange-induced spin correlations.

If one could neglect exchange correlations of the magnetic electrons and treat them as completely independent, then one would expect the spin-transport term to take the ordinary diffusion form $D_0\nabla^2\mathbf{M}$, with diffusion coefficient $D_0 \approx \frac{1}{3}v^2\tau$, where v is a mean Fermi velocity of the magnetic electrons and τ is a translational relaxation time which should be of the order of the conduction-electron relaxation time as calculated from the resistivity, since all electrons can scatter into the same final states. As will be noted below, the experimental evidence makes it clear that no diffusion term of such size is present.[2] This is something of a disparity, since the de Haas–van Alphen[3] and magnetoresistance[4] data for transition metals can be interpreted satisfactorily on a Fermi-surface picture, and in particular the Hall coefficient for nickel is in accurate quantitative agreement with such a picture with the magnetic electrons independent and fully itinerant.[4]

The reason for the disparity is exchange-induced spin correlation of the magnetic electrons, which reduces spin transport far below the independent-particle estimate while leaving charge-transport effects essentially unchanged. This has previously been recognized in a qualitative way,[5] but we believe it is of interest to see a little more explicitly how it comes about in a simple model. In this paper we represent the effects of exchange on an individual magnetic electron by a strong effective magnetic field, everywhere aligned parallel to the local macroscopic magnetization, which acts on the electron's spin but not on its orbital motion. This exchange field will cause the electron to precess at a rapid frequency ω_0 about the local $\mathbf{M}$, and to follow the relatively slow change in orientation of the local $\mathbf{M}$ along its path. Diffusive spin transport is thus quenched by the exchange field. A simplified kinetic-theoretic treatment given in Sec. III for the case of circularly polarized waves leads to a spin-transport term $[D_0/(1+i\omega_0\tau)]\nabla^2\mathbf{M}$, where D_0 is the diffusion coefficient which would be calculated in the absence of the exchange field. This transport term corresponds to a diffusion-like part plus a contribution to the exchange stiffness, both of which are found to be ordinarily small or negligible.

Our effective exchange field should be thought of as a self-consistent single-particle effective field in the Hartree-Fock sense. The exchange interaction conserves total angular momentum, so when one sums over the electrons in a macroscopic region the exchange torques cancel out except for a relatively small remnant, described by the macroscopic exchange stiffness, coming from interaction with electrons outside the given region. For this reason the "exchange precession" at frequency ω_0 of the individual electrons proceeds in such a way as to give zero total precessing magnetization. The precession at frequency ω_0 disappears from the macroscopic equations, and ω_0 does not correspond to an observable resonance. If the up-down splitting of the d electrons is 0.5 eV as for the case of nickel,[6] which we will have particularly in mind, the corresponding effective field is $H_{exch} = \alpha M_s \approx 5\times10^7$ Oe and the precession frequency is $\omega_0 = \gamma\alpha M_s \approx 10^{15}$ sec^{-1}.

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In this paper we consider only the transport of transverse magnetic moment in the weak-signal limit. The transport of the z component, which can become appreciable in the vicinity of the Curie temperature, has been considered by other authors.[7]

II. EXPERIMENTAL SIGNIFICANCE OF A DIFFUSION TERM

The classic theory of the effect of diffusion on electron spin resonance in nonmagnetic metals was given by Dyson.[8] In such metals the time for an electron to diffuse through the skin depth, $T_D \sim (D/\delta^2)^{-1}$, is typically much shorter than the electron spin relaxation time T_2. Thus, a resonating electron can diffuse into and out of the skin depth repeatedly, retaining spin "memory" from one visit to the next, with the exciting fields in the skin depth always in phase with the "remembered" spin. The resultant resonance line shape has a broad weak background of width corresponding to T_D, upon which is superposed a distorted but still narrow resonance of width corresponding to the spin relaxation time T_2. We remark that although Dyson's theory was given in terms of single-particle Green's functions, it could have been formulated as a macroscopic magnetic equation including diffusion, to be solved with Maxwell's equations and appropriate boundary conditions.[2]

The results of diffusion in ferromagnetic resonance are very different from those in Dyson's case. The reason is that the dynamic permeability, instead of being small, is now large compared with unity, and the magnetization is strongly coupled into the Maxwell equations. Whereas in the Dyson case the diffusion-determined penetration depth of the resonating electrons is large compared with the rf skin depth, in the ferromagnetic case the dynamic E, H, and M fields are all essentially characterized by a single penetration depth which is determined self-consistently and decreases at resonance.

Dynamic diffusion effects in a ferromagnetic metal are physically akin to those of exchange stiffness. Exchange stiffness, like diffusion, will cause a sharp local variation in $\mathbf{M}$ introduced at a given time to subsequently dissipate away; however, the exchange stiffness will support standing-wave oscillations, whereas the diffusion term will not. The mathematical relation between the two terms is especially simple in the case of circularly polarized fields, which is appropriate to the geometry with the dc field $H_0\hat{z}$ perpendicular to the sample surface:

$$\mathbf{M} = M_s\hat{z} + m(\hat{x} + i\hat{y})\exp i(qz - \omega t),$$
$$\mathbf{H} = H_0\hat{z} + h(\hat{x} + i\hat{y})\exp i(qz - \omega t). \tag{2}$$

Then the magnetic equation (1), with diffusion added and the dynamic fields assumed small, becomes

$$\omega m = -\gamma M_s h$$
$$+ (\gamma H_0 + (2A\gamma/M_s)q^2 - iDq^2 - (i\omega\lambda/\gamma M_s))m. \tag{3}$$

In this case the diffusion term is mathematically equivalent to a negative imaginary part of the exchange stiffness.

The problem of ferromagnetic resonance in a bulk sample with exchange stiffness and diffusion can be handled by the mathematical apparatus[9,10] which has been set up to handle the problem with exchange stiffness only. In general, the practical mathematics of the solution is complex, but for the perpendicular-field case it can be shown by the methods of Ref. 8 that the following result holds to good approximation, under the assumptions of zero surface-anisotropy energy and normal conductivity:

$$\frac{\sigma c^2}{4\pi i \omega}Z^2 = \mu_{equ} = \mu_1 + i\mu_2 = \frac{\eta - \Omega - iL\Omega + 2(1-i)\epsilon}{[\eta - \Omega - iL\Omega + (1-i)\epsilon]^2}, \tag{4}$$

where μ_{equ} is the "equivalent permeability," Z is the surface impedance, σ is the conductivity, $\eta = H_0/4\pi M_s$, $\Omega = \omega/4\pi M_s\gamma$, $L = \lambda/M_s\gamma$, and $\epsilon = [(2A/M_s - iD/\gamma) \times (2\pi\sigma\omega/c^2)(1/4\pi M_s)]^{1/2}$, $\mathrm{Im}\epsilon \le 0$. For the case of diffusion dominant over exchange stiffness, this solution gives a power-absorption resonance curve which is symmetric about its center and approximately of ordinary relaxation shape, its width being

$$\Delta H \sim \{[(D/4\pi M_s\gamma)(2\pi\omega\sigma/c^2)]^{1/2} + \omega\lambda/4\pi M_s^2\gamma^2\}4\pi M_s.$$

Results for other geometries will not differ greatly from this.

For nickel at room temperature we estimate the mean Fermi velocity as 10^7 cm/sec and $\tau \sim 10^{-14}$ sec. The independent-particle assumption would give a diffusion coefficient $D_0 = v^2\tau/3 \sim 0.3$ cm^2/sec at room temperature and increasing proportionately to the conductivity at lower temperatures. This would predict low-temperature ferromagnetic resonance linewidths many times larger than the observed values.[11] A diffusion term of this size would also require that in thin-film spin-wave resonance experiments the higher modes should be strongly damped, and is in conflict with experimental observations in which modes as high as the 17th have been observed.[12] We conclude that the independent-particle estimate of magnetic diffusion is incorrect.

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III. SOLUTION IN THE EFFECTIVE-FIELD MODEL

Let $\mathbf{u}(\mathbf{r},t; \mathbf{v},t')$ denote the statistically averaged moment of electrons at $(\mathbf{r},t)$ having velocity $\mathbf{v}$ whose last collisions occurred at $(\mathbf{r}-\mathbf{v}t', t-t')$. Assume the scattering to be isotropic and spin-independent and to occur with probability $1/\tau$ per electron per unit time. Then the dyadic magnetization current is

$$\mathbf{J}(\mathbf{r},t)=\frac{M_s}{\mu_B}\int dP_v \int_0^\infty \frac{dt'}{\tau} e^{-t'/\tau}\mathbf{v}\mathbf{u}(\mathbf{r},t; \mathbf{v},t'), \qquad (5)$$

where dP_v is a normalized velocity distribution and (M_s/μ_B) gives the number of magnetic electrons per unit volume. The rate of increase of the magnetization per unit volume at $(\mathbf{r},t)$ due to spin transport, i.e., the spin-transport term to be added to the right side of Eq. (1), is $-\nabla\cdot\mathbf{J}(\mathbf{r},t)$. Our problem is to calculate $\mathbf{u}(\mathbf{r},t; \mathbf{v},t')$.

Our scattering assumption means that each magnetic electron in a given volume has equal probability of being scattered; and since $\mathbf{M}=\langle\mathbf{u}\rangle M_s/\mu_B$, where the brackets denote averaging over a physically small volume, it follows that on the average the moment of an electron immediately after scattering at $(\mathbf{r}-\mathbf{v}t', t-t')$ is $(\mu_B/M_s)\mathbf{M}(\mathbf{r}-\mathbf{v}t', t-t')$. This initial condition plus the precession equation will determine $\mathbf{u}(\mathbf{r},t; \mathbf{v}t')$. We assume the transverse components of magnetization and spin moment to be small, so that their cross products can be ignored; this means the precession equation becomes linear and it is sufficient to consider a single sinusoidal component of transverse $\mathbf{M}$ in order to establish the form of the spin-transport term. We also assume fields of positive helicity, so $\mathbf{M}(\mathbf{r},t)$ has the form (2). The corresponding effective exchange field is $\mathbf{H}_{\mathrm{exch}}=\alpha\mathbf{M}(\mathbf{r},t)$.

Consider the precessional motion of an individual electron of moment $\mathbf{u}$ in a primed coordinate system traveling with the electron and rotating about the z-axis so as to keep the x' axis parallel to the local $\mathbf{m}$. The rate of rotation is $\omega_d=\omega-\mathbf{q}\cdot\mathbf{v}$, the Doppler-shifted frequency of the magnetic wave as seen by the electron. The precession equation is

$$(d\mathbf{u}/dt)'=-\omega_d\hat{z}\times\mathbf{u}+(d\mathbf{u}/dt)_{\mathrm{fixed}}$$
$$=(-\omega_d\hat{z}+\gamma\alpha(M_s\hat{z}+\mathbf{m}))\times\mathbf{u}.$$

The solution can be written as a "driven" term with $\mathbf{u}$ at rest parallel to the apparent field in the rotating system, $\mathbf{u}_{\mathrm{driven}}(t)=\mu_B[\hat{z}+\gamma\alpha\mathbf{m}(t)/(\omega_0-\omega_d)]$, plus a "homogeneous" transverse term which rotates in the primed system at frequency $\omega_0-\omega_d$ or in the fixed system at frequency ω_0.

The initial condition fixes the "homogeneous" term at time $t-t'$ as

$$\frac{\mu_B}{M_s}\mathbf{m}(\mathbf{r}-\mathbf{v}t', t-t')\left(\frac{-\omega_d}{\omega_0-\omega_d}\right)=\frac{\mu_B}{M_s}\mathbf{m}(\mathbf{r},t)\left(\frac{-\omega_d}{\omega_0-\omega_d}\right)e^{i\omega_d t'},$$

and so we find

$$\mathbf{u}(\mathbf{r},t; \mathbf{v},t')=\mu_B\left(\hat{z}+\frac{\gamma\alpha\mathbf{m}(\mathbf{r},t)}{\omega_0-\omega_d}\right)$$
$$+\frac{\mu_B}{M_s}\mathbf{m}(\mathbf{r},t)\left(\frac{-\omega_d}{\omega_0-\omega_d}\right)e^{-i(\omega_0-\omega_d)t'}$$
$$=\frac{\mu_B}{M_s}\mathbf{M}(\mathbf{r},t)+\frac{\mu_B}{M_s}\left(\frac{\omega_d}{\omega_0-\omega_d}\right)\mathbf{m}(\mathbf{r},t)$$
$$\times(1-e^{-i(\omega_0-\omega_d)t'}).$$

Substituting this into Eq. (5) with the approximation $\omega_0-\omega_d\approx\omega_0$, we obtain finally

$$-\nabla\cdot\mathbf{J}(\mathbf{r},t)=-\int dP_v(\mathbf{v}\cdot\nabla)(\mathbf{v}\cdot\nabla)$$
$$\times\int_0^\infty \frac{dt'}{\tau}e^{-t'/\tau}\frac{i}{\omega_0}(1-e^{-i\omega_0 t'})\mathbf{m}(\mathbf{r},t) \quad (6)$$
$$=(v^2\tau/3)[1/(1+i\omega_0\tau)]\nabla^2\mathbf{m}(\mathbf{r},t).$$

To get the final form we have assumed the velocity distribution to have cubic symmetry, and v^2 is the same mean-square velocity that would appear in an ordinary diffusion coefficient.

This term is to be added to the right of Eq. 1. To put it in the form of Eq. 3 we can regard the real part of $(v^2\tau/3)/(1+i\omega_0\tau)$ as a diffusion coefficient and $M_s/2\gamma$ times the imaginary part as a contribution to the exchange stiffness:

$$D=(v^2/3\omega_0)\omega_0\tau/[1+(\omega_0\tau)^2], \qquad (7a)$$

$$A=A_\infty+\frac{M_s}{2\gamma}\frac{v^2}{3\omega_0}\frac{1}{1+(\omega_0\tau)^2}. \qquad (7b)$$

Here A_∞ is the exchange stiffness for $\tau\to\infty$, such as might in principle be obtained from a band-theoretic calculation with correlation corrections. In Eq. (7b) the $\tau=\infty$ part of the transport term has been subtracted since it is to be thought of as already included in A_∞.

For nickel, we estimate $D=0.03\omega_0\tau/(1+(\omega,\tau)^2)$ which has a maximum at $\tau=1/\omega_0\sim10^{-15}$ sec occurring at roughly 600°K. The discussion in Sec. II indicates that the criterion for diffusion effects to be experimentally appreciable is $D\gtrsim2A\gamma/M_s$ and for nickel at low temperatures $2A\gamma/M_s\approx0.1$ cm²/sec. Diffusion effects thus should be negligible at ordinary temperatures. The τ-dependent variation in A reaches a maximum of about 30% of the low-temperature A value, but occurs only at high temperatures where it is masked by the fall-off of A_∞ near the Curie point. Numerical estimates for iron yield similar results.

IV. CONCLUSIONS

By taking account of exchange interactions between itinerant d electrons by means of an effective exchange field, we have found a magnetic diffusion coefficient $D=(v^2/3\omega_0)\{\omega_0\tau/[1+(\omega_0\tau)^2)\}$ which approaches the usual independent-particle form for small translational relaxation time τ, but instead of increasing monotonically with τ goes through a maximum and returns to zero. We also find a τ-dependent contribution to the exchange stiffness. The effect of these terms will be small, especially at low temperatures.

Obviously, independent d electrons plus an effective-field type of spin-spin correlation does not tell the whole story in a ferromagnetic metal, but we think it plausible that the real situation can be well described by a Fermi-liquid picture with spin splitting, with a varying axis of spin quantization corresponding to the varying direction of the macroscopic magnetization. Such a model would have transport properties similar to those of the present simplified model.

Remark on the Hopping Transition Probability of Polaron Holes in NiO

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For the hopping motion of polaron holes the frequency factor W_0 of the transition probability is determined by the observation that hole localization occurs in a molecular orbital of the irreducible set t_{2g} of a cation site group (with transfer integral b_π), whereas the superexchange mechanism is caused by virtual charge transfers between molecular orbitals of the irreducible set e_g (with transfer integral b_σ). The effect of short-range magnetic ordering on W_0 is determined by the time-dependent spin-pair correlation function $\gamma_{ij}(t)$ for neighbor cations i and j and for times $t \gg t_0 =$ relaxation time for the correlation.

I. INTRODUCTION

FOR nonmagnetic ionic crystals the theory of small polaron conduction has been developed by Yamashita and Kurosawa,[1] Holstein,[2] and Sewell.[3] If one wants to apply the results to a concrete case and calculate the dc conductivity of p-type material, one has four unknown parameters, namely, the acceptor concentration N_p and the activation energy E_p determining the concentration of mobile holes, the hopping activation energy E_0, and the frequency factor of the transition probability W_0. A theoretical discussion of W_0 is the object of this note. For bound holes hopping around acceptor sites, it can be determined from the frequency and temperature dependence of the ac conductivity or of the internal friction.[4,5] For mobile holes traveling through the lattice, an unambiguous experimental determination of W_0 is difficult, as can be

seen from the attempt of Y and K to determine it from Morin's[6] experimental data on conductivity and Seebeck coefficient. The magnitude of W_0, however, is of some interest because it can be quite different for the two types of hopping transitions: Adiabatic over-the-barrier transitions for which $W_0=\omega_0/2\pi$ ($\omega_0=$ frequency of longitudinal optical vibration), and non-adiabatic through-the-barrier tunneling transitions for which W_0 is determined by a transfer integral between ligand field orbitals. Therefore, we wish to discuss here:

(1) The relative magnitude of W_0 for the two types of transitions;

(2) the effect of magnetic ordering on W_0.

The first point has received the attention of Y and K and of Herring[7]; the authors come to opposite conclusions. The second point has been considered by Heikes.[8] His result, that W_0 calculated by Y and K and by H is multiplied by a factor depending on the sublattice magnetization $\mathbf{M}$ characterizing long-range order of the

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