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DIRECT RELAXATION OF A LOCAL MOMENT SPIN TO THE LATTICE

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ABSTRACT

Two new relaxation processes of a localized spin moment in a metal are considered: A one-step process in which the local spin is flipped while a conduction electron is scattered without flipping its spin, and a two-step process involving first a change in a local moment by two \hbar units, and subsequently an exchange scattering which flips another local moment by \hbar in the opposite direction. Both processes occur in third order of perturbation theory under the combined action of s - d mixing and spin-orbit interaction. The calculated values of the relaxation time for Mn and Cr in Cu are in good agreement with the measured values.

Measurements [1, 2] of the resonance linewidth in solutions of $3d$ local moments in the noble metals have provided experimental values for the relaxation time T_2 in the bottlenecked regime. Given the fact that the g values of these local moments are very close to the g value of the host conduction electrons, Hasegawa's equations [3] predict that at low concentration the spin-lattice relaxation time T_1 , which determines the rate of loss of Zeeman energy by the spin system, is very nearly equal to the linewidth T_2 .

Previous discussions of the spin-lattice relaxation have been confined to a two-step process in which, first, the local moment and a conduction electron mutually flip their spins under the effect of exchange (T_{sd} and T_{ds} being the corresponding spin-flip times) and second, the conduction electron spin relaxes with a characteristic time T_{sl} to the energy reservoir provided by the kinetic energy of the conduction electrons. This second step occurs under the influence of the spin-orbit interaction. When the system is bottlenecked ($T_{sl} \gg T_{sd}$) and when the local moment susceptibility dominates over the Pauli susceptibility, the effective spin-lattice relaxation time resulting from this process is

$$\frac{1}{T_{eff}} = \frac{T_{sd}}{T_{ds}} \frac{1}{T_{sl}} \quad (1)$$

In this note [4] we point out the existence of two alternative relaxation processes which do not involve the time T_{sl} , namely process 1: The local moment spin flips

by one \hbar unit, the Zeeman energy being transferred in a single process to the kinetic energy of a conduction electron *without* involving a change in the latter's spin, and process 2: Which is a two-step process, first the local moment spin flips by two \hbar units while a conduction electron is scattered with a change in its own spin in the opposite direction, and second, the conduction electron restores its spin to the original direction by exchange-scattering against another local moment. The net change in the local moment spin system is thus one \hbar unit.

Both processes 1 and 2 occur in third order of perturbation theory under the combined action of the s - d mixing and the spin-orbit interaction. Denoting by $T_{dl}^{(1)}$ and $T_{dl}^{(2)}$ the corresponding spin-lattice times, the total relaxation rate is

$$\frac{1}{T_1} = \frac{1}{T_{eff}} + \frac{1}{T_{dl}^{(1)}} + \frac{1}{T_{dl}^{(2)}} \quad (2)$$

and is to be compared with the experimental linewidth. The orbital moment is assumed quenched here so that all the relaxation proceeds via the spin-orbit coupling.

Experimentally [1], a temperature independent contribution to the linewidth has been observed. The times $T_{dl}^{(1)}$ and $T_{dl}^{(2)}$ depend on temperature as T^{-1} so that they are not related to any temperature-independent broadening and the interpretation of the latter is still open. An outline of the calculation of $T_{dl}^{(1)}$ and $T_{dl}^{(2)}$ follows.

The system is described by the Anderson hamiltonian including the conduction band energy H_s , the full energy of the $3d$ ion, H_d (including the electron-electron interaction), the s - d mixing interaction H_{sd} , and the spin-orbit interaction H_{so} . The unperturbed hamiltonian is $\mathcal{H}_0 = H_s + H_d$ and the perturbation is $\mathcal{H}' = H_{sd} + H_{so}$. Since we require orbital quenching of the localized state, our calculation will be valid for an S state of H_d ; if we were to include crystal field effects we could also treat orbital singlets [5]. Here we deal only with the S state case so that our calculation is applicable to Mn in the noble metals in the limit where the level width is small compared to the Coulomb energy.

a. *Calculation of $T_{dl}^{(1)}$.* Let $|{}^6S_m\rangle$, where $m = -5/2$ to $+5/2$, denote the state of the local moment and let $|k\sigma\rangle$ denote the conduction electrons states.

We are interested in transitions that flip the local spin alone, i.e. transitions from the initial state $|{}^6S_m\rangle$ to the final state $|{}^6S_{m\pm 1}\rangle$.

These transitions occur in third order of H' , with H_{sd} acting twice and H_{so} acting once. For instance, a conduction electron $|k\sigma\rangle$ jumps (by H_{sd}) onto the impurity, then one of the electrons on the impurity flips its spin (H_{so}) and finally one of the electrons on the impurity comes off (H_{sd}) with the same spin as the incident electron and goes into the conduction band state $|k'\sigma\rangle$. The matrix element for this transition is:

$$M(m-1, k'\sigma; m, k\sigma) = \sum_{r,s} \frac{\langle m-1, k'\sigma | H' | r \rangle \langle r | H' | s \rangle \langle s | H' | m, k\sigma \rangle}{(E(m, k\sigma) - E_r)(E(m, k\sigma) - E_s)} \quad (3)$$

where the sum is over all possible intermediate states. It is sufficient to compute the $m \rightarrow m-1$ transitions since the $m \rightarrow m+1$ transitions can then be obtained by time reversal.

There are three types of terms in Eq. (3) corresponding to the three possible positions of H_{so} in the product of factors. Note that if H_{so} is at the right or left positions it acts on the ${}^6S, j=5/2$ state and gives [6] a ${}^4P, j=5/2$ state which, having the same number of electrons as the 6S state, is separated from the latter by an exchange energy \mathcal{J} . In terms of the Slater integrals F^2 and F^4 for the free ion, \mathcal{J} has the value [6]

$$\mathcal{J} = \frac{1}{7}F^2 + \frac{10}{21}F^4 \quad (4)$$

On the other hand if H_{so} is in the middle position, H_{sd} acting on the 6S state changes the number of d electrons by ± 1 so that the corresponding excitation energies are U_+ , the energy needed to promote an electron from the Fermi level to the local moment, and U_- , the energy needed to promote an electron from the local moment to the Fermi level.

The transition rate from $|m\rangle$ to $|m-1\rangle$ is given by

$$W_{m-1,m} = \frac{2\pi}{\hbar} \sum_{\sigma} \langle |M(m-1, k'\sigma; m, k\sigma)|^2 \rangle_{k,k'} (N\rho_{at}(\epsilon_F))^2 k_B T \quad (5)$$

where $\langle \rangle_{k,k'}$ denotes the average over k and k' at the Fermi sphere (free electron conduction band), $\rho_{at}(\epsilon_F)$ is the density of conduction band states at the Fermi level per atom, for one spin direction, and N is the number of atoms in the crystal.

The spin-lattice relaxation rate is [7]

$$\frac{1}{T_{dl}^{(1)}} = 2 \frac{\sum_m W_{m-1,m} (E_m - E_{m-1})^2}{2 \sum_m E_m^2} \quad (6)$$

where the factor 2 takes into account the transitions $m \rightarrow m+1$. We omit the details of the calculation of $W_{m-1,m}$, which will be published in a more comprehensive paper, and give only the final result. Defining the quantity C ,

$$C = \frac{2}{\pi \hbar} \lambda^2 \Delta^2 k_B T \quad (7)$$

where λ is the spin-orbit constant of the $3d$ state, and Δ the virtual level width, which is proportional to the square of the s - d mixing interaction, we find for the relaxation rate:

$$\frac{1}{T_{dl}^{(1)}} = \frac{C}{25} \left[7 \left(\frac{1}{U_+^2} + \frac{1}{U_-^2} \right)^2 - \frac{20}{U_+^2 U_-^2} + \frac{8}{J^2} \left(\frac{1}{U_+} + \frac{1}{U_-} \right)^2 + \frac{8}{J} \left(\frac{1}{U_+} + \frac{1}{U_-} \right) \left(\frac{1}{U_+^2} + \frac{1}{U_-^2} \right) \right] \quad (8)$$

b. *Calculation of $T_{dl}^{(2)}$.* The transitions from the state $|m, k_{\downarrow}\rangle$ to the state $|m-2, k'_{\uparrow}\rangle$ are calculated in the same fashion and the transition rates $W_{m-2,m}$ are given by an expression analogous to Eq. (5). To obtain the corresponding relaxation rate we note that the relaxation process is completed only when the conduction-electron spin is restored to its original value, i.e. after a subsequent transition $|m-2, k'_{\uparrow}\rangle \rightarrow |m-1, k''_{\downarrow}\rangle$ occurs. Since this transition occurs via the exchange coupling which is large compared to λ , the rate-determining matrix element is $W_{m-2,m}$ but the net change in the Zeeman energy is that of one spin moment instead of two. The relaxation rate is then

$$\frac{1}{T_{dl}^{(2)}} = \frac{1}{4} \frac{\sum_m W_{m-2,m} (E_m - E_{m-2})^2}{\sum_m E_m^2} \quad (9)$$

as follows from Eq. (6) for equally spaced spin levels. Making use of the calculated values of $W_{m-2,m}$ we find:

$$\frac{1}{T_{dl}^{(2)}} = \frac{4C}{25} \left[\frac{1}{U_+^2} + \frac{1}{U_-^2} + \frac{2}{J} \left(\frac{1}{U_+} + \frac{1}{U_-} \right) \right]^2 \quad (10)$$

c. *Calculation of T_{sl} .* Even though the calculation of T_{sl} (transitions $|m, k_{\uparrow}\rangle \rightarrow |m, k'_{\uparrow}\rangle$) and of the corresponding T_{eff} has previously been done [8] in the Hartree-Fock (*HF*) approximation, we do it over by perturbation theory for purposes of comparison. The matrix element $M(m, k'_{\downarrow}; m, k_{\uparrow})$ is calculated according to Eq. (3) and after averaging over k and k' the rate T_{sl}^{-1} (which is proportional to the atomic concentration of $3d$ impurities, c) is found. To compare with the *HF* value of σ_{sf} given in Reference [8] we use the relation $T_{sl}^{-1} = (c v_F / \Omega) \sigma_{sf}$ where v_F is the Fermi velocity and Ω the atomic volume. To compare with T_{dl} we use the relation $T_{eff} = T_{sl} [2c S(S+1) / 3 \rho_{at}(\epsilon_F) k_B T]$ which follows from Eq. (1).

We find: (1) The value of σ_{sf} calculated from perturbation theory is equal to the limiting value of the *HF* result for small (Δ/U) . This limit is obtained by letting $\sin^2 \delta_{\uparrow} \approx \Delta^2 / U_-^2$ and $\sin^2 \delta_{\downarrow} \approx \Delta^2 / U_+^2$ in the *HF* expression of Reference [8]. Thus the *HF* result, which is valid in the limit $(\Delta/U) \gg 1$, reduces correctly to the perturbation result which is valid in the opposite limit, $(\Delta/U) \ll 1$. (2) The perturbation

calculation shows that the enhancement factor $(1 - U_{orb}/U_{sp})^{-1}$ relating to $\tilde{\lambda}$, the off-diagonal component of the spin-orbit interaction in Reference [8], owes its origin to those terms in Eq. (3) where H_{so} is at the right or left position in the third order matrix element so that one of the energy denominators in Eq. (3) is \mathcal{J} . This is shown by first, verifying that it is these terms which give rise to the partial cross section σ_{od} which depends on $\tilde{\lambda}$ in Reference [8], and second, by examining the limit of σ_{od} when $\Delta \ll U$. This limit is:

$$\sigma_{od} = \frac{4\pi}{k_F^2} \frac{10}{3} \left(\frac{\lambda}{\Delta}\right)^2 \left(\frac{U_{sp}}{U_{sp} - U_{orb}}\right)^2 \frac{\Delta^4}{U_+^2 U_-^2} \quad (11)$$

The quantity U_{sp} is to be identified with $U_+ + U_-$ and the difference $U_{sp} - U_{orb} = (1/7)F^2 + (10/21)F^4$, calculated from Reference [8] is precisely equal to \mathcal{J} . This identification of the origin of an enhancement factor in the HF calculation with a group of terms in the perturbation expansion is illuminating and it provides a satisfactory bridge between the two calculations.

From the calculation of T_{sl} , the following expression for $T_{eff}^{-1} \equiv (T_{sd}/T_{ds}) T_{sl}^{-1}$ is found:

$$\frac{1}{T_{eff}} = \frac{C}{7} \left[\left(\frac{1}{U_+^2} + \frac{1}{U_-^2} \right)^2 + \frac{8}{\mathcal{J}^2} \left(\frac{1}{U_+} + \frac{1}{U_-} \right)^2 \right] \quad (12)$$

where C is the constant given by Eq. (10).

d. *Comparison with Experiment.* Because of the fact that the portion of the linewidth that is proportional to T had previously been thought to arise *exclusively* from the T_{sl} process, all of the temperature-dependent linewidth has been interpreted [1, 2] as a T_{sl} process. However all three processes, T_{sl} , $T_{dl}^{(1)}$ and $T_{dl}^{(2)}$ give linewidths proportional to T so that only their combined effect given by Eq. (2) is obtained from the temperature-dependent linewidth. Thus the value $1/T_{sl} = 4.8 \times 10^7 \text{ sec}^{-1}/\text{ppm}$ deduced [1] for Cr in Cu is actually an apparent rate, given by the sum $1/T_{app} = 1/T_{sl} + X_r (1/T_{dl}^{(1)} + 1/T_{dl}^{(2)})$ where $X_r = (T_{ds}/T_{sd})$. The deduced spin-flip cross section is *not* the true cross section of the impurity for flipping the electron spin but differs from it by a factor R ,

$$R = T_{eff}/T_1 = 1 + X_r T_{sl} (1/T_{dl}^{(1)} + 1/T_{dl}^{(2)}) \quad (13)$$

which is directly obtainable from Eq's. (8), (10) and (12) when the values of U_+ , U_- , and \mathcal{J} are known.

The only S state ion with which we can compare our theory is Mn^{2+} . Shana-barger [2] measured the T dependent part of the width and found $1/\gamma(T_2)_{app} = 0.96 \text{ gauss/ppm}$. The corresponding value of the apparent spin-flip cross section is

$(\sigma_{sf})_{app} = 1.27 \times 10^{-18} \text{ cm}^2 = R\sigma_{sf}$ where σ_{sf} is the true conduction electron spin-flip cross section.

To calculate σ_{sf} and R we need to know the quantities λ , Δ , U_+ , U_- , and \mathcal{J} . The value of λ for the Mn^{2+} ion is [9] $\lambda = 330 \text{ cm}^{-1}$. The value of Δ for Mn in Cu has not been measured, but for Ni in Cu the measured value is [10] $\Delta \simeq 0.3$, and for Ti in Cu , the estimated value is [11] $\Delta = 1.0 \text{ eV}$. For Mn , we estimate $\Delta = 0.55 \text{ eV}$.

The values of U_+ and U_- are estimated from the measured [12] saturation moment of Mn which is $(n_\uparrow - n_\downarrow) \mu_B = 4.0 \mu_B$ and from the assumed total number of d electrons on Mn in solution, $n_\uparrow + n_\downarrow$, which we estimate to range between 5.0 and 5.4. Use of the Lorentzian approximation for the virtual level gives then the quantities U_+ and U_- . Finally we take $\mathcal{J} = \frac{1}{2}(U_+ + U_-)$ which is a value close to but smaller than the ${}^6S - {}^4P$ separation in the free ion.

With these values of the parameters the three relaxation times T_{st} , $T_{dl}^{(1)}$, and $T_{dl}^{(2)}$ as well as the corresponding σ_{sf} and the factor R have been calculated. The results are shown in Table 1. It is seen that the calculated σ_{sf} does not vary much as $n_\uparrow + n_\downarrow$ varies between 5.0 and 5.4, even though U_+ and U_- vary appreciably. At fixed n_\uparrow and n_\downarrow , if Δ is allowed to change, the resulting σ_{sf} varies as Δ^{-2} and R does not change. This follows since U_+ , U_- are proportional to Δ when n_\uparrow , n_\downarrow are fixed.

The agreement between the calculated $(\sigma_{sf})_{app}$ and the experimental value of $1.27 \times 10^{-18} \text{ cm}^2$ is too close to be meaningful since the actual state of the Mn ion is probably not so close to the perturbation limit. Agreement within a factor of 2 should be considered good.

Finally, the case of Cr can be qualitatively compared with the present theory. The measured value is [1] $(\sigma_{sf})_{app} = 3.6 \times 10^{-18} \text{ cm}^2$ and the value of σ_{sf} , using only the T_{st} process, and calculated in Hartree-Fock approximation is $8 \times 10^{-18} \text{ cm}^2$. A factor R between 2 and 3 gives qualitative agreement, but Cr is probably less close to the perturbation limit than Mn , and in any event a separate calculation would have to be made for Cr since it is not in an S state in the ionic limit.

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I am grateful to M. R. Shanabarger for communicating his unpublished measurements and allowing me to quote his result.

TABLE I

Calculated values of σ_{sf} and $(\sigma_{sf})_{app} = R_{sf}$ for Mn in Cu.

$n_{\uparrow} + n_{\downarrow}$	$U_{+} [eV]$	$U_{-} [eV]$	$\mathcal{J} [eV]$	$R = \frac{T_{eff}}{T_1}$	$\sigma_{sf} [\text{cm}^2]$	$(\sigma_{sf})_{app} [\text{cm}^2]$
5.4	1.46	3.60	2.53	3.12	4.22×10^{-19}	1.32×10^{-18}
5.2	1.74	2.68	2.21	2.77	4.92×10^{-19}	1.36×10^{-18}
5.0	2.12	2.12	2.12	2.68	4.62×10^{-19}	1.24×10^{-18}

The values assumed for the local moment parameter are: $n_{\uparrow} - n_{\downarrow} = 4.0$; $\Delta = 0.55 \text{ eV}$; and the values of $n_{\uparrow} + n_{\downarrow}$ listed in the first column. The values of U_{+} and U_{-} then follow from the virtual level description. \mathcal{J} is taken to be $\frac{1}{2}(U_{+} + U_{-})$. The measured value of $(\sigma_{sf})_{app}$ is $1.27 \times 10^{-18} \text{ cm}^2$.

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DISCUSSION

MONOD: Is the mechanism by which you have this spin-flip of the local moment and no spin-flip of the conduction electron just like the one that Giovannini calculated in the anomalous Hall effect, and which boils down to a spin-orbit interaction in which it is the spin of the magnetic impurity and the orbit of the conduction electron?

YAFET: If you want, yes.

MONOD: You have an interaction $\lambda^* I \cdot S$ which does the same thing as Yafet says.

ORBACH: Is that a spin-other orbit that you are talking about? This I thought was a spin-orbit on the same ion.

GIOVANNINI: This effective interaction can have various sources, and one is the one that has been described by Yafet, which gives the same functional form for the interaction.

