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SIMPLE CONSIDERATIONS ON THE NUCLEAR RELAXATION OF DIAMAGNETIC IMPURITIES IN FERROMAGNETIC METALS

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Résumé. — La relaxation nucléaire d’impuretés diamagnétiques dissoutes dans des métaux ferromagnétiques peut être influencée par des processus d’ordre élevé mettant en jeu les excitations des électrons magnétiques. Nous étudions ce problème à l’aide d’un modèle très simple (polarisation uniforme), en utilisant un formalisme de susceptibilités et en écrivant des équations de mouvements couplées pour les électrons ferromagnétiques et les électrons de conduction. Nous discutons en détail l’interprétation physique des différentes susceptibilités qui apparaissent dans le problème, et nous faisons le raccord avec la théorie élaborée par Giovannini, Heeger, Pincus, Gladstone pour traiter la relaxation nucléaire de la matrice dans les métaux diamagnétiques contenant des impuretés paramagnétiques.

Abstract. — In ferromagnetic metals, high order processes involving the excitations of the magnetic electrons may contribute to the nuclear relaxation of diamagnetic impurities. Here this problem is investigated on the basis of a very simple model (uniform polarization), with the help of a susceptibility formalism and of coupled equations of motion for the ferromagnetic and conduction electrons. The physical interpretation of the various susceptibilities which enter the problem is discussed at length, and a connection is made with the Giovannini, Heeger, Pincus, Gladstone treatment of the nuclear relaxation of the matrix in diamagnetic metals containing paramagnetic impurities.

1. Introduction. — Nuclear relaxation times of diamagnetic impurity nuclei in ferromagnetic metals at low temperatures have recently been measured by NMR and nuclear orientation techniques [1], [2], [3]. In principle, this relaxation proceeds through the coupling of the nucleus with the conduction electrons. But one may wonder if, in addition to the simple Korringa process, higher order processes involving the excitations of the ferromagnetic system cannot take a part in the relaxation.

This problem has been considered in the appendix of a recent paper by Kontani et al. [1] who use a susceptibility formalism and coupled equations of motion. But they do not explain the transition from their eq. (A.4) to (A.5) (which involves a change in the notations, (A.4) being expressed in terms of $\chi_a^0$ and (A.5) in terms of $\chi_a$ with no very precise definition of $\chi_a^0$ and $\chi_a$). This transition is indeed the delicate point of this type of calculation. For this reason, our own work, although undertaken before the publication of the paper by Kontani et al., will be presented here as a comment of their appendix.

In the same way as Kontani et al. we consider an atom with nuclear spin $I$ which may be either a diamagnetic impurity or an atom of the matrix. In both cases, this particular atom will be denoted as « the impurity ». This atom is embedded in a ferromagnetic metallic matrix which contains two systems of electrons: conduction electrons ($s$) with a spin density $\sigma(R)$, and ferromagnetic electrons ($m$) which may or may not be strictly localized, giving rise to a spin $S$ per atom ($S > J_0/2$). In practice, these magnetic electrons will be $d$ electrons in transition metals and $f$ electrons in rare earth metals.

Between the two types of electrons, there exists an exchange coupling:

$$\mathcal{H}_{sm} = - \frac{2 I_{sm}}{N_v} \sum_q \sigma_q \cdot S_q$$

where $N_v$ is the number of magnetic atoms per unit volume and the Fourier components $\sigma_q$ and $S_q$ of $\sigma(R)$ and $S(R)$ are defined as in reference [1]; $q$ lies in the first Brillouin zone.

The hyperfine hamiltonian of the impurity may be written as:

$$\mathcal{H} = \frac{A}{N_v} \sigma(R) \cdot I + KS \cdot I$$

(*) Associé au CNRS.
where $K$ is non zero only when the impurity is an atom of the matrix. In view of the present uncertainty concerning the origin of the hyperfine fields on impurities in metals [4], [5], [6], $A$ will be considered as an adjustable parameter.

If we admit that the presence of the impurity does not greatly alter the properties of the matrix and assuming a uniform polarization model (1), the nuclear resonance frequency is given by:

$$\hbar \omega_n = KE + \frac{A}{2N_s} (N_{s+} - N_{s-})$$

or, since

$$\frac{J_{sm} S}{E_F} \ll 1$$

$$\hbar \omega_n = KE + AJ_{sm} SN_s(E_F)$$

where $N_s(E_F)$ is the density of $s$ electrons per atom and per spin at the Fermi level ($N(E_F) = 3p/4$ energy per $s$ electron and a free electron band).

Let us now consider two small transverse fictitious fields $h_{qw}$, $h_{sm}$ at circular frequency $\omega$, the first of which only acts on $s$ electrons, the second one only acting on $m$ electrons

$$\mathcal{J} = -g\mu_B h_{qw} \sigma_{qw} - g\mu_B h_{sm}' S_{qw}.$$  

By definition of the effective transverse susceptibilities $\tilde{\chi}_{sw}$, $\tilde{\chi}_{sm}$ and $\tilde{\chi}_s$, we shall write:

$$g\mu_B \sigma_{qw} = \tilde{\chi}_s(q, \omega) h_{qw} + \tilde{\chi}_{sm}(q, \omega) h_{sm}$$

$$g\mu_B S_{qw} = \tilde{\chi}_{sm}(q, \omega) h_{qw} + \tilde{\chi}_s(q, \omega) h_{sm}.$$  

From which we deduce that:

$$\tilde{\chi}_{sm}(q, \omega) = \frac{\tilde{\chi}_s(q, \omega)}{1 - \frac{2U}{g^2 \mu_B^2 N_s} \chi_s(q, \omega) - \left( \frac{2J_{sm}}{g^2 \mu_B^2 N_s} \right)^2 \chi_s(q, \omega) \chi_s(q, \omega)}$$

$$\tilde{\chi}_{sm}(q, \omega) = \frac{2J_{sm}}{g^2 \mu_B^2 N_s} \chi_s(q, \omega) \tilde{\chi}_s(q, \omega)$$

$$\chi_s(q, \omega) = \chi_s(q, \omega) + \left( \frac{2J_{sm}}{g^2 \mu_B^2 N_s} \right)^2 \chi_s^2(q, \omega) \tilde{\chi}_s(q, \omega)$$

when $U = 0$, these equations are identical to (A.4) of Kontani et al. [1] except for the change in notations ($\chi_s \rightarrow \tilde{\chi}_s$, $\chi_s' \rightarrow \chi_{sw}$, $\chi_s'' \rightarrow \chi_{sm}$, $\chi_s \rightarrow \tilde{\chi}_s$).

We must now indicate what $\chi_{sm}$ and $\chi_s$ are. In fact,
these are not simple electron susceptibilities since they must \[7\] be calculated in the presence of the longitudinal effects of \( \mathcal{N}_{\text{int}} \) and \( U \).

2.2 Rare Earth Metals (f Electrons). For simplicity let us first consider the case of well localized ferromagnetic spins with \( U = 0 \) (for example gadolinium metal). Then, in the presence of an external longitudinal field \( H_0 \), the susceptibility \( \chi_f(\omega) \) in eq. (8) \((m = f)\) is given by:

\[
\frac{1}{\chi_f(\omega)} = \frac{g \mu_B H_0 + \hbar \omega + \frac{2 J_{st}}{N_s} \left( N_{s+} - N_{s-} \right)}{g^2 \mu_B^2 S N_s} \tag{12}
\]

where the third term in the numerator represents the effect, on one of the localized spins \( S \), of the exchange field resulting from the polarization of the \( s \) band due to all other magnetic ions.

Inserting (12) in (9), we get:

\[
\tilde{\chi}_f(q, \omega) = \frac{N_s S g^2 \mu_B^2}{g \mu_B H_0 + \hbar \omega + J_{st} \frac{N_{s+} - N_{s-}}{N_s} - \frac{(2 J_{st})^2}{N_s g^2 \mu_B^2} S \chi_f(q, \omega)} \tag{13}
\]

where \( \chi_f(q, \omega) \) must be the transverse susceptibility of the \( s \) electrons in the presence of the exchange \( \mathcal{N}_{st} \) interaction, which introduces a splitting \( 2 J_{st} S \) between the up and down spin electrons

\[
(E_{k_1} - E_{k_1} = - 2 J_{st} S).
\]

As is well known:

\[
\chi_f(00) = \frac{g^2 \mu_B^2}{2} \frac{N_{s+} - N_{s-}}{2 J_{st} S} \tag{14}
\]

so that, when \( q = \omega = 0 \), the two last terms in the denominator of (13) cancel out, as they should in order to preserve the rotational invariance of the \( eS \) system.

\[
\tilde{\chi}_f = \frac{N_s S g^2 \mu_B^2}{g \mu_B H_0 + \hbar \omega + \frac{(2 J_{st})^2}{N_s g^2 \mu_B^2} \left( \chi_f(00) - \chi_f(q, 0) \right) - i \frac{(2 J_{st})^2}{N_s g^2 \mu_B^2} \chi_f(q, \omega)} \tag{21}
\]

in which \( \chi_f \) is the free (unpolarized) electron susceptibility. This shows that whatever the value of \( q \) the spin wave dispersion relation is given by:

\[
\hbar \omega_q = g \mu_B H_0 + \frac{(2 J_{st})^2 S}{N_s g^2 \mu_B^2} \left( \chi_f(00) - \chi_f(q, 0) \right) \tag{22}
\]

But it is well known that in a magnetic system with exchange hamiltonian:

\[
\mathcal{H} = \sum_{ij} J_{ij} S_i \cdot S_j \tag{23}
\]

Let us now consider the case \( q \neq 0 \), and assume for simplicity that in the absence of exchange \( (\mathcal{N}_q) \), coupling the \( s \) electrons behave like free electrons. Then, the imaginary part of \( \chi_f(q, \omega) \) is different from zero only when \([1]\):

\[
\frac{q^*}{q_{\min}} < q < 2 k_F^* \tag{15}
\]

with

\[
q^*_{\min} = \frac{k_F^*}{E_F^*} \left| \frac{J_{st} S}{E_F^*} \right| \tag{16}
\]

For \( q > q^*_{\min} \) it is given by [1]

\[
\text{Im} \chi_f(q, \omega) = \frac{N_s N_s(E_F^*)}{8 E_F^*} \frac{\pi \hbar \omega}{g^2 \mu_B^2} \left( \frac{2 k_F^*}{q} \right) \tag{17}
\]

Consequently

\[
\tilde{\chi}_f(q, \omega) = \frac{N_s S g^2 \mu_B^2}{g \mu_B H_0 + \hbar \omega + D q^2} \tag{18}
\]

This represents the susceptibility of a ferromagnetic assembly of spins in the presence of spin waves with dispersion relation

\[
\hbar \omega_q = g \mu_B H_0 + D q^2 \tag{19}
\]

More generally for arbitrary \( q \), it may be shown that since \( J_{st} S/E_F < 1 \), the real part of

\[
\frac{(2 J_{st})^2 S}{N_s g^2 \mu_B^2} \left[ \chi_f(00) - \chi_f(q, \omega) \right] \tag{20}
\]

differs only slightly from the value obtained by making \( J_{st} = 0 \). Then, we may write:

\[
\text{Im} \chi_f(q, \omega) = \frac{N_s S g^2 \mu_B^2}{g \mu_B H_0 + \hbar \omega + D q^2} \tag{21}
\]

where \( J(q) \) is the Fourier transform of \( J_{st} \).

In the case we are considering here [9], the ferromagnetic spins are coupled by the Ruderman-Kittel (RKKY) mechanism for which the exchange hamil-

(2) See Appendix.
tonian in the traditional approach may be written
as \[10\] :

\[\mathcal{C} = - \sum_{q} \left( \frac{2 J_s}{N_s' g^2 \mu_B^2} \right)^2 Z_0(q) S_q \cdot S_{-q} \]  

(25)

so that :

\[2 J_s(q) = \left( \frac{2 J_s}{N_s' g^2 \mu_B^2} \right)^2 Z_0(q) . \]

Inserting (26) into (24), we get again eq. (22).

Having thus checked the internal consistency of our results, we must however notice that the rigorous spin wave dispersion relation is not given by (22), but must be obtained by equating to zero the real part of the denominator of (13)

\[g \mu_B H_0 + h \omega + J_s' \frac{N_{s+} - N_{s-}}{N_s} = \frac{(2 J_s)^2}{N_s' g^2 \mu_B^2} Z_0(q, \omega) = 0 . \]

(27)

This is an implicit equation in \(\omega\), which in addition to the spin wave branch may have other roots probably associated with higher energy electronic excitations [11].

As concerns the imaginary part, which becomes non zero when \(q > q_{\text{min}}\), it corresponds to the damping of the spin waves by the Stoner excitations of the conduction electrons. It is interesting to notice that this damping is proportional to the frequency of excitation of these spin waves.

For small \(\omega \) (\(h \omega < \hbar^2 \frac{2 q k_F - q^2}{2 m} \)) :

\[\text{Im} \chi(q, \omega) = \frac{\pi}{\text{Re} Z_0(q, \omega)} \frac{1}{q} \frac{h \omega}{k_F} \frac{1}{F(q/k_F)} \]

(28)

where \(F(q/k_F)\), the Lindhard function, is for small \(q \) :

\[F(q/k_F) \approx 1 - \frac{1}{12} \left( \frac{q}{k_F} \right)^2 . \]

Then :

\[\Delta \omega \over \omega_q = \frac{\text{Im} \chi(q, \omega)}{Z_0(00) - Z_0(q, 0)} = \frac{3 \pi}{N_s(E_F)} \left( \frac{q}{k_F} \right)^3 \]

(30)

where \(\Delta \omega \) is the damping of the spin wave (excited at \(\omega \)) and \(\omega_q\) its circular frequency, i. e. for small \(q \)

\[\omega_q = \frac{1}{6} \left( J_s \right)^2 S N_s(E_F) \left( \frac{q}{k_F} \right)^2 . \]

(31)

When the spin waves are excited at their own frequency it follows from (30) that :

\[\Delta \omega \over \omega_q = \frac{\pi}{2} \left( \frac{J_s}{E_F} \right)^2 \frac{S}{N_s(E_F)} \frac{k_F}{q} . \]

(32)

In normal rare earth metals, there are three conduction electrons per atom. Specific heat measurements lead to \(N(E_F) \sim 2/eV/at\); for a free electron like band this would correspond to \(E_F \sim 1 \text{ eV}\). On the other hand, approximating the Fermi surface by a sphere we have that \(k_F = 0.8 \text{ A. U.} = 1.5 \times 10^8 \text{ cm}^{-1}\), and, assuming a parabolic band with effective mass \(m^* = 3.1 \text{ m}_e\), we arrive at \(E_F \sim 2.5 \text{ eV} \) [12]. Finally, Freeman et al. [13] have estimated \(E_F \sim 3.4 \text{ eV}\).

Anyhow, if, following Sano et al. [5], we set \(2 J_{\text{sm}} \) equal to \(0.15 \text{ eV}\), it appears that \(\Delta \omega \omega_q < 1 \); this shows that the spin waves retain their meaning when \(q > q_{\text{min}}\).

As concerns the spin wave energies (eq. (31)) taking \(N(E_F) \sim 2/eV/at\), \(J_s \sim 0.075 \text{ eV}\), \(S \sim 7/2\) we arrive at :

\[h \omega_q = 65 \text{ meV} \times \left( \frac{q}{k_F} \right)^2 . \]

(33)

This may be compared with the neutron diffraction results for gadolinium [14]. In the \(c\) direction, when \(q = \frac{\pi}{c} \times 10^8 \sim 0.55 \times 10^8\)

the spin wave energy is of order 8 \text{ meV}. Formula (33) would lead to \(h \omega_q \sim 8.6 \text{ meV}\). But, then, at the edge of the Brillouin zone \((q = 2 \pi/c)\), we would have \(h \omega_q \sim 34 \text{ meV}\) instead of the observed 14 \text{ meV}. Then a law :

\[h \omega_q = 30 \text{ meV} \left( \frac{q}{k_F} \right)^2 \]

(34)

giving values of order of 15-20 \text{ meV} at the edges of the Brillouin zone would seem more appropriate to a rough representation of the spin wave dispersion law in this hexagonal metal in which the energies of the spin waves are fairly anisotropic and far from simple parabolic looking curves.

Besides the term \(D q^2\), the spin wave energy contains a term \(g \mu_B H_0\). The value of \(q\) for which the two contributions are equal is such that :

\[q_0 = \sqrt{\frac{g \mu_B H_0}{D}} \]

(35)

Assuming that :

\[h \omega_q = 30 \text{ meV} \left( \frac{q}{k_F} \right)^2 , \quad k_F = 1.5 \times 10^8 , \quad H_0 = 10000 \text{ Oe} \]

(which corresponds to \(g \mu_B H_0 = 1 \text{ cm}^{-1} = 1/8 \text{ meV}\), we find :

\[\frac{q_0}{k_F} = \frac{1}{15.5} ; \quad q_0 \sim 10^7 \text{ cm}^{-1} . \]

(36)

This is to be compared with the value of \(q_{\text{min}}\). Taking \(J_s \sim 0.075 \text{ eV}\), \(S = 7/2\), \(E_F = 2 \text{ eV}\) (corresponding to \(N_s(E_F) \sim 1/eV/at\), which would be consistent with eq. (34)), one gets :

\[\frac{q_{\text{min}}}{k_F} \sim \frac{1}{7.6} ; \quad q_{\text{min}} \sim 2 q_0 \times 10^7 \text{ cm}^{-1} . \]

(37)

With \(E_F = 1 \text{ eV}\), we would have obtained a value twice as big. But \(J_s\) is not well known either, so that we cannot expect to get precise estimates.
2.3 TRANSITION METALS (d ELECTRONS). — In this case, the magnetic d electrons must be treated as a band and \( U \neq 0 \). Taking into account the longitudinal effects the energies of up and down spin electrons are given, as functions of the unperturbed energies by:

\[
\begin{align*}
E_{d\uparrow}^p &= E_k - \frac{g \mu_B H^0}{2} + \frac{J_{sd}}{N_v} \sigma + U n_{d\uparrow} \\
E_{d\downarrow}^p &= E_k + \frac{g \mu_B H^0}{2} + \frac{J_{sd}}{N_v} \sigma + U n_{d\downarrow}
\end{align*}
\] (38)

in which \( n_{d\uparrow}, n_{d\downarrow} \) are the numbers of d electrons with spins \( \uparrow \) per atom.

Then, in eq. (9):

\[
\chi_d(q, \omega) = \chi_d(q, \omega) - \frac{2 U}{g^2 \mu_B^2 N_v} \chi_d(q, \omega) - \left( \frac{2 J_{sd}}{g^2 \mu_B^2 N_v} \right)^2 \chi_d(q, \omega) \chi_d(q, \omega)
\] (39)

\( \chi_d(q, \omega) \) must be the transverse susceptibility of the d electrons calculated using expressions (38) for the energies [7].

Notice that \( n_{d\uparrow} \) and \( n_{d\downarrow} \) are not arbitrary but must be determined in a self-consistent way using the relation:

\[
2 < S_z > = n_{d\uparrow} - n_{d\downarrow} = \frac{1}{N_v} \left[ \sum_k f(E_k) - \sum_k f(E_k) \right]
\] (40)

and expressions (38) for the energies (it is possible to relate the \( < \sigma > \) term to \( n_{d\uparrow} - n_{d\downarrow} \) :

\[
2 J_{sd} \frac{N_s(E_F)}{N_v} < \sigma > = J_{sd}^2 N_s(E_F) (n_{d\uparrow} - n_{d\downarrow}).
\]

The condition for the appearance of ferromagnetism is then found to be :

\[
1 - (U + J_{sd}^2 N_s(E_F)) N_d(E_F) \leq 0
\] (41)

in agreement with the classical result when one makes \( J_{sd} = 0 \) (\( N_d(E_F) = \) density of d states per atom and per spin).

In practice, \( U \) is taken to have an effective value of 1.5 eV [7],

\[
J_{sd} \leq 1 \text{ eV}, \quad N_d(E_F) \sim \frac{3 p}{4 E_F^d}
\]

(where \( p \sim 1, E_F^d \sim 5 \text{ eV} \) for iron) so that

\[
J_{sd}^2 N_s(E_F) < U
\] (42)

and the occurrence of ferromagnetism appears mainly as a consequence of the dd interaction term as is usually admitted.

Assuming that \( n_{d\uparrow} - n_{d\downarrow} \) has been determined self-consistently, let us set \( n_{d\uparrow} - n_{d\downarrow} = 2 S \). Since \( \chi_d(q, \omega) \) must be computed from the energies (38), it follows that (approximating very roughly the d band by a free electron band), \( \Im \chi_d(q, \omega) \) will be different from zero only if:

\[
q^d > q_{min}^d
\]

with

\[
q_{min}^d \sim \left[ \frac{U + J_{sd}^2 N_s(E_F)}{E_F^d} \right] S \sim \frac{U}{E_F^d}.
\] (43)

It is difficult to predict any real order of magnitude of \( q_{min}^d \), since the real d band is quite different from a free electron band [15]. Nevertheless, formulas (15) and (43) together with the numerical estimates of \( J_{sd} \) and \( U \) suggest that \( q_{min}^d \) and \( q_{min}^d \) must have comparable orders of magnitude with \( q_{min}^d > q_{min}^d \).

Let us now come back to the study of \( \chi_d(q) \). As in the case of rare earths, it may be checked that, for small \( q \):

\[
\chi_d(q) \sim \frac{N_s \mu_b^2 \mu_b^2}{g \mu_b H^0 + D q^2 + \hbar \omega} \sim \frac{N_s \mu_b^2 \mu_b^2}{\hbar \omega_q + \hbar \omega}.
\] (44)

This shows the existence of spin waves with a dispersion law:

\[
h \omega_q = g \mu_b H^0 + D q^2.
\] (45)

When \( q \) increases, the dispersion law becomes less simple and the spin waves are damped. As a matter of fact in eq. (39):

— When \( q > q_{min}^d \), \( \chi_d(q) \) is different from zero, this corresponds to the relaxation of the spin waves by the Stoner excitations of the conduction electrons.

— When \( q > q_{min}^d, \chi_d(q) \neq 0 \), this corresponds to the merging of the spin wave branch into the continuum of Stoner excitations of the d electrons.

\( D \) and \( q_{min}^d \) can be obtained experimentally by neutron diffraction [16]. In iron, \( D \) is found to be of order 280 meV. \( \AA^2 \). As for \( q_{min}^d \), it corresponds to the steep decrease in diffused intensity which accompanies the merging of the spin waves into the Stoner excitations. This occurs when \( h \omega_q = D q^2 = 85 \text{ meV} \). It follows that:

\[
q_{min}^d \sim 5.5 \times 10^7 \text{ cm}^{-1}.
\] (46)

By comparison, with [17] \( k^d_F \sim 1.3 \times 10^8 \), \( J_{sd} \sim 0.5 \) to 1 eV, \( S \sim 1, E_F^d \sim 5 \text{ eV} \), we get from eq. (15):

\[
q_{min}^d \sim 1.3 \times 10^7 \text{ cm}^{-1}.
\] (47)

Both \( q_{min}^d \) and \( q_{min}^d \) are not very small compared with the average radius of the Brillouin zone (\( \sim 1.7 \times 10^8 \text{ cm}^{-1} \)).

On the other hand, the value of \( q \) for which \( D q^2 = g \mu_b H^0 \) is of order \( q_0 \sim 2.1 \times 10^6 \) (for \( H^0 = 10^4 \text{ Oe} \)) i. e. much smaller than \( q_{min}^d \) and \( q_{min}^d \).
Before closing this paragraph, we must add a few remarks:

- In the spin wave dispersion relation \( g \mu_B H_0 \) must in reality be replaced by \( g \mu_B (H_0 + H_A - H_{dem}) \) where \( H_A \) is the anisotropy field and \( H_{dem} \) the demagnetizing field. In a multidomain sample \( H_{dem} = H_0 \) in a uniformly magnetized sphere

\[
H_{dem} = \frac{4 \pi}{3} M.
\]

- Strictly speaking, \( \chi(q) \) defined in the reduced zone scheme is a sum over the reciprocal lattice vectors [18].

\[
\chi(q) = \sum_k \chi(q + K_k, q).
\]

This is related to the possible occurrence of umklapp processes in the relaxation calculations and has important bearings on the magnetic structures of rare earth metals. But assuming a ferromagnetic rare earth metal amounts assuming that the maximum of the real \( \chi(q) \) lies at \( q = 0 \), as in the case of free electrons.

3. Application to nuclear relaxation. — 3.1 General Formulas. — Let us now come back to eqs. (7), (9)-(11). We have to extract the imaginary parts of \( \tilde{\chi}_m \), \( \tilde{\chi}_s \), and \( \tilde{\chi}_e \) taking account of the fact that:

\[
\tilde{\chi}_m, \tilde{\chi}_e \ll \tilde{\chi}_m, \tilde{\chi}_e. 
\]

Then, we find that:

\[
\begin{align*}
\text{Re } \tilde{\chi}_m &= \frac{\chi'_m}{1 - 2 \frac{U}{g^2 \mu_B^2 N_s} \chi_m - \left( \frac{2 J_{sm}}{g^2 \mu_B^2 N_s} \right)^2 \chi_e \chi_m} \\
\text{Im } \tilde{\chi}_m &= \left( \text{Re } \tilde{\chi}_m \right)^2 \left[ \frac{\chi''_m}{\chi_m^2} + \left( \frac{2 J_{sm}}{g^2 \mu_B^2 N_s} \right)^2 \chi_e \right].
\end{align*}
\]

which after substitution in (9)-(11) give again eq. A.5 of Kontani when one sets \( \chi''_m = 0 \).

After completing all the calculations, we find that:

\[
\begin{align*}
K + \frac{2 J_{sm}}{N_s g^2 \mu_B^2} A \chi'_q(q, \omega) &\approx K + \frac{2 J_{sm}}{N_s g^2 \mu_B^2} A \chi'_q(0) = \frac{\hbar \omega_q}{S} \\
&\quad + \frac{2 J_{sm}}{N_s g^2 \mu_B^2} A \chi'_q(0)
\end{align*}
\]

and, dropping some of the \( (q, \omega) \) to simplify the aspect of the formulas, eq. (51) becomes:

\[
\begin{align*}
&\quad + \frac{2 J_{sm}}{N_s g^2 \mu_B^2} A \chi'_q(0) = \frac{\hbar \omega_q}{S} \\
&\quad + \frac{2 J_{sm}}{N_s g^2 \mu_B^2} A \chi'_q(0)
\end{align*}
\]

We shall define:

\[
k_0 = \sqrt[\frac{\hbar \mu_B(H_0 + H_A - H_{dem})}{D}}.
\]

As concerns the integration over \( q \), \( \chi'_q(q, \omega) \) is different from zero only if \( q_{min} \ll q \ll 2 k_F \). Then if \( 2 k_F \ll q \) (edge of the Brillouin zone) the integration must run from \( q_{min} \) to \( 2 k_F \). Otherwise it must run from \( q_{min} \) to \( q \). We shall write

\[
q_{max} = \text{Inf } (q_B, 2 k_F).
\]
Then eq. (53) leads to:

\[ \frac{1}{T_1} = \pi (N_s (E_F))^2 \frac{\omega_n}{2} \coth \left( \frac{\hbar \omega_n}{2 k_B T} \right) \left( A^2 + \frac{A J_{sd} \hbar \omega_n}{D k_F^2} \log \left[ \frac{q_{max}^2 + k_0^2}{q_{min}^2 + k_0^2} \right] + \frac{\hbar \omega_n}{S} \left( \frac{E_F}{k_F^2} \right)^2 \frac{1}{1 + \left( \frac{k_0}{q_{min}} \right)^2} \right). \]

In the high temperature limit \( |\hbar \omega_n/k_B T| \ll 1 \)
this result is identical to eq. (A.9) of Kontani et al. [1]. One should notice that the Log term is probably overestimated since all \( q \) values contribute to it so that eq. (52) is not strictly valid. It is possible to check that the last term of eq. (57) reduces to Weger’s result [19] when \( k_0/q_{min} \to 0 \):

\[ \frac{1}{T_1} = \frac{1}{2} \frac{\omega_n^3}{\omega_{ex}^2} \coth \left( \frac{\hbar \omega_n}{2 k_B T} \right) \frac{a^2 \Sigma}{64 \pi^4 S^2} \]  

in which \( \Sigma \) is the area of the Fermi surface and \( \omega_{ex} = D/a^2 \).

Notice that in the derivation of these results a) we have neglected the umklapp processes b) we have assumed that the spin wave spectrum of the matrix is not seriously perturbed by the presence of the diamagnetic impurity [20]. Therefore the following estimates will be more qualitative than quantitative.

3.2 Rare earth metals. — 3.2.1 Diamagnetic impurity in a rare earth metal.

Then \( \chi'_m = 0 \) in eq. (53) (no intrinsic damping of the localized spins) and, using:

\[ \hbar \omega_n = A J_{sd} S N_s (E_F) \]
\[ D k_F^2 = \frac{1}{6} J_{sd}^2 S N_s (E_F) \]

the last bracket of eq. (57) takes the form:

\[ (B) = A^2 \left[ 1 + 6 \log \left( \frac{q_{max}^2 + k_0^2}{q_{min}^2 + k_0^2} \right) + \frac{36 E_F^2}{S^2 J_{sd}^2} \frac{1}{1 + \left( \frac{k_0^2}{q_{min}^2} \right)^2} \right]. \]

When the anisotropy field is weak compared with the external field: \( k_0/q_{min} \sim q_{m}/q_{min} \sim 1/2 \) in a field \( H_0 = 10^4 \text{ Oe} \) (§ 2.2). Thus the second and third terms begin to become field dependent above \( 10^4 \text{ Oe} \). As concerns the relative orders of magnitude, taking \( E_F \sim 2 \text{ eV}, J_{sd} \sim 0.075 \text{ eV}, S = 7/2 \), we find that the three terms of eq. (59) are in the ratios:

\[ \frac{1}{T_1} \sim 1 \quad ; \quad \sim 18 \quad ; \quad \sim 270. \]

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As concerns the precise evaluation of $T_1$, the inclusion of umklapp processes is found to be necessary. Similar results have been obtained in Terbium [21].

3.3 Transition metals. — In this case, we may more or less consider that we have spin waves in the whole Brillouin zone but that these spin waves become very strongly damped as soon as they become degenerate with the continuum of Stoner excitations of the $d$ electrons ($q > q_{min}^d$).

The anisotropy fields being usually small ($\sim 10^3$ Oe) we have that $k_0^2 \simeq q_0^2$.

Then from the discussion of paragraph 2.3, it follows that:

$$\frac{k_0^2}{q_{min}^2}, \frac{k_0^2}{q_{min}^2} \ll 1$$

i. e. we cannot expect to see a dependence on $T_1$ on the external magnetic field.

The dependence observed by Kontani et al. at a few kOe thus seems to be due to residual Bloch Walls and domains effects — or to arise from other phenomena.

Turning now to the expression (53) of $1/T_1$, let us estimate the ratios of the contributions due to $\chi_0^d$ and $\chi_\alpha^d$ in the last term

$$\left(\frac{\hbar}{S}\right)^2 \sum_q \left(\frac{2 \chi_0^d}{N_s \mu_B^2} \right)^2 \chi_0^d$$

Assuming that $\text{Re} \chi_0(q, \omega) \sim N_s \mathcal{S} D \mu_B^2$ and approximating very roughly $\chi_0^d(q, \omega)$ by a free electron expression, these two contributions will be respectively proportional to $(q_{max}^d = 2 k_F^d$ or $2 k_F^d$):

$$\left(\frac{2 J_{sd}}{N_s \mu_B^2} \right)^2 \frac{2}{E_F q_{min}^2} \frac{N_s(E_F) k_F^d}{\chi_0^d}$$

For iron, we can try to adopt $|J_{sd}| \sim 1$ eV, $S \sim 1$, $E_F = 5$ eV, $Dk_F^2 = E_F/12$, $q_{max}^d/q_{min}^d \sim 7.5$. Then the three terms of the bracket in eq. (64) are respectively equal to:

$$\begin{array}{c}
1 \\
1.2 \\
2.25
\end{array}$$

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Therefore if we compare the relaxation of a same well-behaved impurity (for example Ag) in a diamagnetic metal and in a ferromagnetic metal like iron, for a given value of $AN_s(E_F)$, its relaxation should be several times faster in iron ($\sim 5$ times faster with the above numerical values).

These estimates are only tentative, but the existence of such an enhancement should not be ignored in the theories where one compares $(T_1 T)^{-1}$ with the square of the hyperfine field.

$$\left(\frac{\chi_0^d q_{min}^d}{N_s(E_F)^2 q_{min}^d} \right)^2$$

3.3.1 Simple diamagnetic impurities. — Let us consider the magnitude of the different terms of eq. (57) for a diamagnetic impurity. Setting $k_0/q_{min} = 0$, we find that the bracket is equal to:

$$[B] = A^2 \left[1 + \frac{J_{sd}^2 S N_s(E_F)}{D k_F^2} \frac{q_{max}^d q_{min}^d}{D k_F^2} + \frac{N_s(E_F)^2 E_F^2 J_{sd}^2}{D k_F^2} \right].$$

For iron, we can try to adopt $|J_{sd}| \sim 1$ eV, $S \sim 1$, $E_F = 5$ eV, $Dk_F^2 = E_F/12$, $q_{max}^d/q_{min}^d \sim 7.5$. Then the three terms of the bracket in eq. (64) are respectively equal to:

1 ; 1.2 ; 2.25

Korringa Weger

Therefore if we compare the relaxation of a same well-behaved impurity (for example Ag) in a diamagnetic metal and in a ferromagnetic metal like iron, for a given value of $AN_s(E_F)$, its relaxation should be several times faster in iron ($\sim 5$ times faster with the above numerical values).

These estimates are only tentative, but the existence of such an enhancement should not be ignored in the theories where one compares $(T_1 T)^{-1}$ with the square of the hyperfine field.

3.3.2 Atom of the matrix. — In that case, as well as for impurities of the 3 d and 4 d groups the relaxation is thought to be mainly of orbital origin [22] (in connection with virtual bound state filling in the case of impurities). This process has not been considered in the above calculations.

On the other hand, if the Weger process eq. (58) happened to be dominant the quantity $\omega_s T_1 T$ should be constant for a given matrix whatever be the impurity (normal or transition).
4. Remark. — All throughout this calculation the ferromagnetic system has been treated as being at zero temperature. At finite temperature, the method which has been used would lead to a dispersion term $D q^2$ in the spin wave energy varying like $< S_\perp >$ i.e. of the form $(1 - \alpha T^{3/2})$. It is well known that in insulating ferromagnets the real variation is $(1 - b T^{5/2})$ and this must also hold for rare earth metals.

For this reason, using an expression of type (9) in order to describe the behaviour of a paramagnetic impurity in a diamagnetic metal at finite temperatures cannot be expected to give correct results (see below).

5. Relation of the present considerations to the Giovannini-Heeger-Pincus-Gladstone calculations. — These authors [23] study the relaxation of a nucleus situated in a diamagnetic metallic matrix at point $r = 0$, in the presence of a paramagnetic impurity situated at point $r = R_j$. Here we have to work in real space with susceptibilities $\chi(r, \omega)$ instead of $\chi_{\perp \nu}$, $N_v$ being the number of sites per unit volume, we assume a coupling

$$\mathcal{H} = -\frac{2J}{N_v} \sigma(R_j) \cdot S(R_j)$$

(65)

$$g \mu_B < S_j(\omega) > = \tilde{\chi}(\omega) \hbar' + \frac{2J}{g^2 \beta^2 N_v} \chi'(R_j, \omega) \tilde{\chi}(\omega) \hbar$$

$$g \mu_B < \sigma(r, \omega) > = \left( \chi'(r, \omega) + \frac{2J}{g^2 \beta^2 N_v} \right)^2 \chi'(r - R_j, \omega) \chi(R_j, \omega) N_v \tilde{\chi}(\omega) \hbar +$$

$$+ \frac{2J}{g^2 \beta^2} \chi'(r - R_j, \omega) \tilde{\chi}(\omega) \hbar'$$

(68)

These equations show that the observable susceptibility of the impurity is indeed $\tilde{\chi}(\omega)$, and that in the presence of this impurity at point $R_j$, the electron response at point $r$ to the field $\hbar$ at $r = 0$ is given by

$$\tilde{\chi}(r, \omega) = \chi'(r, \omega) +$$

$$+ \left( \frac{2J}{g^2 \beta^2 N_v} \right)^2 N_v \chi_0(r - R_j, \omega) \chi(R_j, \omega) \tilde{\chi}(\omega).$$

(69)

In particular the susceptibility to be used in the computation of the relaxation time of a nuclear spin at $r = 0$ is:

$$\tilde{\chi}(0, \omega) = \chi'(0, \omega) + \left( \frac{2J}{g^2 \beta^2 N_v} \right)^2 N_v (\chi_{\perp 0}(R_j, \omega))^2 \tilde{\chi}(\omega).$$

(70)

This equation is the strict equivalent of eq. (11). As concerns the relationship between $\tilde{\chi}(\omega)$ and $\chi(\omega)$, since $\tilde{\chi}(\omega)$ must have rotational invariance properties, $\chi(\omega)$ must include the longitudinal effects of the conduction electrons i.e. at 0 K

$$\chi(\omega) = \frac{g^2 \mu_B^2 S}{g \mu_B H_0 + \frac{(2J)^2 S}{g^2 \mu_B^2 N_v} \chi'(r = 0, 0) + \hbar\omega}.$$ 

(71)

Then

$$\tilde{\chi}(\omega) = \frac{g^2 \mu_B^2 S}{g \mu_B H_0 + \frac{(2J)^2 S}{g^2 \mu_B^2 N_v} (\chi'(r = 0, 0) - \chi'(r = 0, \omega)) + \hbar\omega}.$$ 

(72)

which reduces to

$$\tilde{\chi}(\omega) = \frac{g^2 \mu_B^2 S}{g \mu_B H_0}$$

(73)

when $\omega = 0$ as it should. As a matter of fact the existence of the $J^2$ term in eq. (72) is associated with the self energy of the Ruderman-Kittel interaction. In
the static limit \((\omega = 0)\), this energy has the form
\((2 J)^2 S(S + 1)\), or, in terms of the \(\chi(q, \omega)\)
the form
\[(2 J)^2 S(S + 1) \times \frac{1}{N} \sum_{q} \chi(q, 0, 0).
\]

It is clearly rotation invariant. On the other hand, it diverges. This last feature has been commented by Kittel [24].

The inspection of the self energy, as well as the remarks of paragraph 4 suggest that the use of a formula of type (72) in order to compute \(\chi(\omega)\) at finite temperatures cannot be expected to give good results.

It is then better to adopt an empirical and reasonable representation such as for example eq. (3.1) of reference [23] i.e., in terms of \(\chi(\omega)\) (not \(\chi(\omega)\))
\[
\tilde{\chi}(\omega) = \frac{g^2 \beta^2 < S_z >}{\omega - \omega_m + i \frac{\tau_2}{\omega}}.
\]

As a final remark let us now go back to the case of a diamagnetic impurity in a ferromagnetic metal. Then \(K = 0\) in eq. (51). Taking account of eq. (50), it appears that the second line of eq. (51) \(\propto x e X e \text{Re } \chi_m\) corresponds to the Giovannini-Heeger type of process while the third line \(\propto x e^2 \text{Im } \chi_m\) corresponds to the Benoit-de Gennes-Silhouette type of process

6. Conclusion. — In conclusion, we have shown that the nuclear relaxation of diamagnetic impurities in ferromagnetic metals may be strongly influenced by indirect processes involving virtual or real excitations of the ferromagnetic electrons. Relaxation by real spin waves is made possible by the damping of the spin waves due, either to their coupling with the conduction electrons, or to their merging into the continuum of Stoner excitations of the magnetic electrons. Formulas previously derived by other authors are obtained as particular cases of our general expression (51). As concerns numerical values, the enhancement factor of the relaxation rate by processes involving spin waves, is found to be of order one to five in the iron group and of order one hundred in the rare earth group. Although our very simple model certainly has to be improved in order to provide more precise estimates, the existence of such an enhancement should not be ignored in the theory of hyperfine interactions in metals.

Acknowledgments. — It is a pleasure to thank Dr. I. A. Campbell who called our attention to the work by Sano, Kobayashi and Itoh (ref. [5]), which was the starting point of this study.

Appendix

Computation of the transverse, frequency dependent spin susceptibility of the free electron gas in the presence of a longitudinal magnetic field \(^{(3)}\)

This problem has already been considered by de Chatel and Szabo [25], who give the result in reduced units of their own. Here we give the formulas for small \(\omega\) with notations closer to those used in the main text.

NOTATIONS. — Electron gas in the absence of magnetic field : Fermi energy \(E_F^0\), Fermi vector \(k_F^0\), \(N = k_F^0/3 \pi^2\) density of electrons of both spins.

— Electron gas in the presence of a longitudinal magnetic field \(H_0\). Response at circular frequency \(\omega\). — \(N = N_+ + N_-\) where \(N_+, N_-\) are the densities of electrons of a given spin.

We define
\[
N_- = \frac{k_F^3}{6 \pi^2}, \quad N_+ = \frac{k_F^3}{6 \pi^2}.
\]

Then
\[
E_F = \frac{\hbar^2 k_F^2}{2 m} + \frac{\hbar^2 k_F^2}{2 m} - \Delta
\]

\((3)\) In practice the (large) longitudinal fields which we are considering in the main text are the fictitious fields associated with the couplings involving \(J_{aa}, J_{ee}\) and \(U\). They only act on the spin of the electron, not on its orbital motion.
IMAGINARY PART OF THE SUSCEPTIBILITY. — We just give the result for small $\Delta/E_F$. Then $\chi'(q, \omega)$ is different from zero only when $q_{\min} < q < 2k_F^0 - q_{\min}$ and it is given by eq. (16) of the main text.

$$\chi'(q, \omega) = \chi_0 \left( \frac{k_{F_1} + k_{F_2}}{4k_F^0} + \frac{2\Delta'}{4q^2} \frac{k_{F_1} - k_{F_2}}{k_F^0} \right) +$$

$$+ \left( \frac{k_{F_1}^2}{4qk_F^0} - \frac{1}{16q^3k_F^0} (2\Delta' + q^2)^2 \right) \log \left( \frac{q^2 + 2k_{F_1}q + 2\Delta'}{q^2 - 2k_{F_1}q + 2\Delta'} \right)$$

$$+ \left( \frac{k_{F_2}^2}{4qk_F^0} - \frac{1}{16q^3k_F^0} (2\Delta' - q^2)^2 \right) \log \left( \frac{q^2 + 2k_{F_2}q - 2\Delta'}{q^2 - 2k_{F_2}q - 2\Delta'} \right).$$

It is possible to check that when $\Delta = 0$, and after performing the relevant modifications, this expression is in agreement with eq. (6.6a) of the book by Schrieffer (Theory of Superconductivity).

We have done a numerical evaluation of $\chi'(q, 0)$ for several values of the parameter $W = 2\Delta/E_F^0$. The results are reported in figure 1. It is seen that even for rather large values of $2\Delta/E_F^0$, $\chi'(q, 0)$ only slightly deviates from the standard Lindhard function.

References

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