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Crystalline Electric Field Effects in *f*-Electron Magnetism

Edited by

Robert P. Guertin
Tufts University
Medford, Massachusetts

and

Wojciech Suski and
Zygmunt Zołnierek

Polish Academy of Sci

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CONDUCTION ELECTRON EFFECTS ON LOCALIZED SPIN EXCITATIONS IN THE RKKY-THEORY OF MAGNETISM

V. Christoph, A.L. Kuzemsky and Th. Frauenheim

Joint Institute for Nuclear Research, Dubna, 10100 Moscow
P.O. Box 79, USSR

INTRODUCTION

The magnetic scattering of thermal neutrons is a unique technique for establishing both the static and the dynamic properties of magnetic correlations. For interpreting neutron inelastic magnetic scattering data on heavy rare earth metals, the calculations of the magnetic susceptibility and the magnetic excitation spectrum are of particular interest[1-11]. In rare earth metals the exchange interaction between the localized 4f electrons and the extended (conduction) electrons (RKKY exchange) is basic for understanding their magnetic and electric properties[1].

In order to avoid the difficulties connected with crystal field and anisotropy effects, we restrict our consideration to rare earth metals like Gd. Recent detailed experimental and theoretical examinations[9-14] confirm the spin moment of the Gd ion to be a good quantum number. The d-band, having a width of 5-7 eV, lies well above the 4f level and is about one fourth occupied. The density of states of the d-electrons at the Fermi level is much higher than the density of states of the s(p) electrons[8-12]. The general conclusion drawn from these investigations was that the conduction electrons, which play an essential role in the mechanism of RKKY exchange in the heavy rare earth metals, cannot be considered as free s-electrons as assumed in early papers[2,3], but rather as similar to tight-binding d-electrons in transition metals[4-14]. In particular, the magnetic excitation spectrum of Gd has been calculated[11] taking into consideration the d-like character of the extended electrons. Starting with an APW calculation of the band structure and wave functions, the RKKY exchange matrix elements were obtained. Agreement of the calculated magnon spectrum with the experimental one[15] could be obtained by reducing the calculated values by a scale factor of about four. In a more recent paper[16] the generalized spin susceptibility has been calculated using the KKR method.

In the present report the generalized spin susceptibility and the magnon spectrum of Gd metal has been calculated. The tight-binding d-like character of the conduction electrons and the electron-electron and electron-phonon interaction are taken into account in a unified manner. The contributions of different interactions to the magnon damping are estimated, and the lower temperature dependence of the magnon width is calculated. As has been noted[17], the magnon lifetime investigations in Gd at low temperature should give information on different interactions and their roles and significance in the heavy rare earth metals.

RARE EARTH METAL MODEL

Neglecting crystal field and anisotropy effects, we describe the rare earth metal by localized 4f spins, interacting with d-like tight-binding conduction electrons. We take into consideration the electron-electron and electron-phonon interactions in the framework of a model given by S. Barisic et al.[18]. The Hamiltonian[18] is a generalization of the well known Hubbard Hamiltonian[19] and has been investigated in detail in Ref. 20.

The total Hamiltonian is:

$$\mathcal{H}=H_d+H_{d-f}+H_{d-ph}+H_{ph}, \tag{1}$$

where

$$H_d=\sum_{k\sigma} E(k)\alpha^+_{k\sigma}\alpha_{k\sigma}+\frac{U}{2N}\sum_{\substack{kk',\\ q\sigma}} \alpha^+_{k+q\sigma}\alpha_{k\sigma}\alpha^+_{k'-q,-\sigma}\alpha_{k',-\sigma} \tag{2}$$

is the Hubbard Hamiltonian[19]. For the tight-binding d-electrons we use $E(k) = 2\sum_\alpha t(\vec{a}_\alpha)\cos(\vec{k}\vec{a}_\alpha)$, where $t(\vec{a}_\alpha)$ is the hopping integral between next nearest neighbors[18], and $\vec{a}_\alpha$ (α=1,2,3) denotes the lattice vectors in a simple lattice with inversion center. The second term in (2) describes the Coulomb interaction of electrons with opposite spins at the same lattice site. The RKKY Hamiltonian describing the interaction of the total 4f spin $\vec{s}$ with the spin density of the conduction electrons has the form

$$H_{d-f}=-\frac{J}{\sqrt{N}}\sum_{kq}\{(\alpha^+_{k\uparrow}\alpha_{k+q\uparrow}-\alpha^+_{k\downarrow}\alpha_{k+q\downarrow})S^z_{-q}+(\alpha^+_{k\uparrow}\alpha_{k+q\downarrow}S^+_{-q}+\text{c.c.})\} \tag{3}$$

where J is the local RKKY exchange integral. In general the exchange integral strongly depends on the wave vectors $\vec{k}$ and $\vec{q}$ having maximum values at k=q=0[11]. For simplicity we restrict ourselves to a local exchange. The generalization to non-local exchange is straightforward. For the electron-phonon interaction we use[18]:

$$H_{d-ph}=\sum_{kq}\sum_{\sigma\nu} V^\nu(k,k+q)Q_{q\nu}\alpha^+_{k+q\sigma}\alpha_{k\sigma} \tag{4}$$

where

$$V^\nu(\vec{k},\vec{k}+\vec{q})=\frac{2iq_o}{\sqrt{NM}}\sum_\alpha t(\vec{a}_\alpha)e^\alpha_\nu(\vec{q})\{\sin\vec{a}_\alpha\vec{k}-\sin\vec{a}_\alpha(\vec{k}+\vec{q})\}. \tag{5}$$

In (5) q_o is the Slater coefficient originating in the exponential decrease of the d-functions[18], N is the number of unit cells in the crystal, and M is the ion mass. $\vec{e}_\nu(\vec{q})$ (ν=1,2,3) are the polarization vectors of the phonon modes.

For the vibrating ion system we have

$$H_{ph}=\frac{1}{2}\sum_{q\nu}(P^+_{q\nu}P_{q\nu}+\omega_o^2(q\nu)Q^+_{q\nu}Q_{q\nu}) \tag{6}$$

where $P_{q\nu}$ and $Q_{q\nu}$ are the normal coordinates and $\omega_o(q\nu)$ are the accoustical phonon frequencies. Thus, as in the Hubbard model, the d- and s-(p)bands are replaced by one "effective" band in our model[1]. However, the s-electrons give rise to screening effects and are taken into account by choosing proper values of J, U and the accoustical phonon frequences $\omega_o(q\nu)$[21].

GENERALIZED SPIN SUSCEPTIBILITY

We are interested in the Fourier transform of the generalized susceptibility of the localized f-spins $\langle\langle S^+_k|S^-_{-k}\rangle\rangle$ where

$$\langle\langle S^+_k(t)|S^-_{-k}(0)\rangle\rangle=-i\theta(t)\langle[S^+_k(t)S^-_{-k}(0)]\rangle \tag{7}$$

is the usual double-time commutator Green's function[22] and $\vec{S}_k$ is the Fourier transform of the f-spin $\vec{S}_i$. For the calculation of $\langle\langle S^+_k|S^-_{-k}\rangle\rangle$ we use the irreducible Green's function technique[23] already applied to the Hubbard model in the atomic and band limits[24].

The method of calculation is based on using the generalized matrix Green's function

$$\bar{G}=\begin{pmatrix}\langle\langle S^+_k|S^-_{-k}\rangle\rangle & \langle\langle S^+_k|\sigma^-_{-k}\rangle\rangle\\ \langle\langle \sigma^+_k|S^-_{-k}\rangle\rangle & \langle\langle \sigma^+_k|\sigma^-_{-k}\rangle\rangle\end{pmatrix}, \tag{8}$$

where the Fourier components of the conduction electron spin densities, $\sigma^+_k=\Sigma_q\alpha^+_{k\uparrow}\alpha_{k+q\downarrow}$ and $\sigma^-_k=\Sigma_q\alpha^+_{k\downarrow}\alpha_{k+q\uparrow}$, have been introduced. It may be shown that the equation of motion for G [Eq.(8)] may be exactly transformed to the Dyson equation by using the irreducible Green's functions with an explicit representation of the mass operator. The exact Dyson equation is given by

$$\bar{G}=\bar{G}_o+\bar{G}_o\bar{M}\bar{G}, \tag{9}$$

where $\bar{M}$ is the mass operator and $\bar{G}_0$ is the mean field Green's function. Hence the determination of $\bar{G}_0$ has been reduced to the determination of $\bar{G}_0$ and $\bar{M}$, when the mean field Green's function $\bar{G}_0^-$ coincides with the RPA-result[25].

DAMPING OF SPIN WAVES IN THE COUPLED LOCAL MOMENT-ELECTRON SYSTEM

In order to estimate the damping of the localized spin excitation spectrum due to electron-magnon and electron phonon scattering, we calculate the local spin susceptibility for small k and ω

$$<<S_k^+|S_{-k}^->>_\omega = \frac{2N^{-1/2}<\tilde{S}_0^z>}{\omega-\varepsilon_k-2N^{-1/2}<\tilde{S}_0^z>\Sigma(k,\omega)}\,, \tag{10}$$

which contains the matrix elements of the mass operator $\bar{M}=(M_{ij})$ in a linear approximation

$$\Sigma(k,\omega)=M_{11}+(M_{12}+M_{21})\frac{J^{1/2}\chi_o^{df}}{1-U\chi_o^{df}}+\frac{J^2N(\chi_o^{df})^2M_{22}}{(1-U\chi_o^{df})^2}\,. \tag{11}$$

Here we have used the notations

$$<\tilde{S}_o^z>=<S_o^z>(1+\frac{N_\uparrow-N_\downarrow}{2N^{1/2}<S_o^z>});\ \chi_o^{df}=\chi_o^{df}(k,\omega)=\frac{1}{N}\sum_q\frac{f_{q+k\downarrow}-f_{q\uparrow}}{\omega_{q,k}}$$

$$\omega_{q,k}=\omega+E(q)-E(q+k)-\Delta;\quad N_\sigma=\sum_q f_{q\sigma}=\sum_q<n_{q\sigma}> \tag{12}$$

$$\Delta=2JN^{-1/2}<S_o^z>+UN^{-1}(N_\uparrow-N_\downarrow).$$

The spectral density of the spin wave excitations with wave vector k then reads

$$g(k,\omega)=-\frac{1}{\pi}\mathrm{Im}<<S_k^+|S_{-k}^->>=\frac{2N^{-1/2}<\tilde{S}_o^z>\Gamma(k,\omega)}{[\omega-\varepsilon_k-\Delta(k,\omega)]^2+[\Gamma(k,\omega)]^2}\,, \tag{13}$$

where

$$\Delta(k,\omega)=2N^{-1/2}<\tilde{S}_o^z>\mathrm{Re}\Sigma(k,\omega),$$
$$\Gamma(k,\omega)=-2N^{-1/2}<\tilde{S}_o^z>\mathrm{Im}\Sigma(k,\omega+i\varepsilon) \tag{14}$$

describes the shift and the damping of the acoustical magnons.

Finally we estimate the temperature dependence of $\Gamma(\vec{k},\omega)$ due to the mass operator terms in (11). Considering the first contribution in (11) at low temperatures we obtain

$$\mathrm{Im}M_{11}\sim T. \tag{15}$$

The other electron-magnon contributions to $\Gamma(\vec{k},\omega)$ can be treated in the same way, where M_{12}, M_{21} and the electron-magnon contribution to M_{22} are proportional to T also. For the electron-phonon contribution to M_{22} we find

$$\mathrm{Im}M_{22}^{ph}\sim T^3. \tag{16}$$

Hence the damping of the acoustical magnons at low temperatures can be written as

$$\Gamma(\vec{k},\omega)|_{k,\omega\to 0}\sim\Gamma_1+\Gamma_2T+\Gamma_3T^3 \tag{17}$$

where the coefficients Γ_i (i=1,2,3) vanish for k=ω=0. For the case when J=0, the electron-electron term Γ_1 vanishes in (17).

DISCUSSION

In the present paper the spectrum of the elementary magnetic excitations and their lifetimes have been calculated for the microscopic model of heavy rare earth metals. In the mode used the electron-electron interaction of the extended (conduction) electrons and the electron-phonon interaction in the Barisic-Labbe-Friedel manner have been taken into account. Neglecting the damping terms the result obtained (10) coincides with that of L.C. Bartel[25], obtained in the RPA. Furthermore, using a generalized matrix Green's function here the expressions for the spin susceptibility of the conduction electrons and mixed terms have been derived. Therefore, starting with this formalism the contribution of the extended (conduction) electrons to the total magnetization of the rare earths[4,5] may also be considered.

The temperature dependence of the acoustical magnon damping is given mainly by the electron-magnon and electron-phonon interactions. the electron-electron contribution Γ_1 is almost temperature independent, but vanishes at J=0, also. It should be emphasized here that the real electronic structure of a rare earth metal such as Gd is much more complicated than given by our tight-binding model[1,4-14]. For a realistic first principle calculation of the stiffness constant about ten bands should be included[4,5,8,10,11]. Furthermore, the dependence of the RKKY exchange integral J(k,k+q) on the wave vector should be taken into account[11]. Nevertheless, taking into consideration all interactions playing a part in the rare earth metals, this estimate of the temperature dependence of the magnon damping should be reasonable. Unfortunately there are no neutron scattering measurements of the low temperature magnon damping in Gd[17]. Such measurements and their theoretical interpretation would greatly improve our understanding of the interactions in the heavy rare earth metals.

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COMMENTS

GREIDANUS: What is your argument for the statement that in the neutron spectrum of PrNi$_2$ the observed linewidth cannot be explained by dispersion effects?

FRAUENHEIM: I believe that dispersion averaging is important only if you average over the whole Brillouin zone, but not if you average over the directions in a polycrystal at a fixed momentum transfer. As known from the single crystal neutron inelastic measurements in PrAl$_2$, direction averaging in a polycrystal does not give such a linewidth effect. Maybe the new theoretical results from Dr. Keller's lecture may give the explanation.

OLES: You mentioned quite a big discrepancy between the measured and the theoretical values of the exchange constant in Gd. What is your explanation of that?

FRAUENHEIM: I have no explanation for the J($\vec{q}$) reduction in Gd. I am only sure that the electron correlation effects would lead to an enhancement of the J($\vec{q}$) value, which is in contrast to J. F. Cooke's explanation.