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Theoretical Magnon Dispersion Curves for Gd†

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The magnon dispersion curve of Gd metal has been determined from first principles by use of augmented-plane-wave energy bands and wave functions. The exchange matrix elements $I(\vec{k},\vec{k}')$ between the $4f$ electrons and the conduction electrons from the first six energy bands were calculated under the assumption of an unscreened Coulomb interaction. The results are in good overall agreement with experiment provided the $I(\vec{k},\vec{k}')$ are diminished by a constant scale factor of about 2 which may be caused by screening.

The s-d (or s-f in the rare earths) interaction model has had great success in describing a variety of phenomena ranging from conduction-electron polarization effects to properties of localized magnetic moments. The s-f exchange interaction $I_{s-f}(\vec{k},\vec{k}')$ between the localized $4f$ and the conduction electrons in the rare-earth metals is the basis of the indirect exchange mechanism of

Ruderman-Kittel-Kasuya-Yosida (RKKY) theory and is thought to be responsible for the observed exotic magnetic orderings and associated magnetic and electric properties of these metals.[1] Unlike semiempirical applications of the model which invoke simple phenomenological parameters, the first-principles approach has appeared to become increasingly complex (and increasingly remote from the original free-electron and δ-function-interaction formulation of RKKY theory) as knowledge about the rare-earth transition-metal band structures, complex Fermi surfaces, and dominant d-like nature of the wave functions has become understood.[2] In addition a recent *ab initio* calculation[3] for Gd metal has shown that the $4f$-conduction-electron exchange matrix elements $I(\vec{k}, \vec{k}')$ show a great deal of structure as functions of k and k' and are not describable by the usual replacement with the phenomenological constant parameter $I(0)$, or by the approximate term $I(q)$ where $q = |\vec{k}' - \vec{k}|$.

In view of the fundamental importance of RKKY theory, we have undertaken the first *ab initio* determination of the magnon dispersion curve for a rare-earth metal. We chose to perform calculations for the case of gadolinium for several reasons. Experimental data are available on both the magnon spectrum[4] and the magnetic form factor,[5] giving information about the exchange interaction, the local-moment distribution, and the conduction-electron polarization. It is also important that Gd is in many ways the simplest rare-earth metal because of the spherically symmetric half-filled $4f$ shell which gives the largest and most isotropic Heisenberg exchange interaction in the rare-earth metals. Furthermore the lack of orbital moment in Gd is extremely important because it is responsible for the crystalline-field anisotropy and magnetostriction being several orders of magnitude smaller than the exchange energy and hence negligible. The importance of the orbital moments for the interactions in the other rare-earth metals has been reviewed from the phenomenological viewpoint by Cooper.[6] By using the energy-band structure and wave functions obtained from a nonrelativistic augmented-plane-wave (APW) calculation for Gd metal and the actual $I(\vec{k}, \vec{k}')$ matrix elements for the first six bands, we have undertaken the most severe test of RKKY theory to date. We find the results of the calculation to be in surprisingly good agreement (to within a constant scale factor) with spin-wave experiments.[4] Surprisingly too, we find the dominant role to be played by the matrix elements $I(\vec{k}, \vec{k}')$ rather than the Fermi-surface nesting features in bringing about agreement with experiment.

The magnon spectrum for localized moments is generally developed from a Heisenberg Hamiltonian

$$H = -\sum_{ij} J(\vec{R}_{ij}) \vec{S}_i \cdot \vec{S}_j \tag{1}$$

in which localized moments with spin $\vec{S}$ are located on lattice sites (indexed by i and j) and interact with strength $J(\vec{R}_{ij})$. The Fourier transform $J(\vec{q})$ of this interaction can be directly related to the magnon spectrum,

$$\hbar\omega(\vec{q}) = 2S[J(0) - J(\vec{q})] . \tag{2}$$

Since the localized $4f$ orbitals have negligible overlap between nearest neighbors, the exchange interaction $J(\vec{R}_{ij})$ is believed to arise from the indirect coupling of the conduction electrons as described by the RKKY model. In this formulation, the expression for $J(\vec{q})$ is given by

$$J(\vec{q}) = N^{-1} \sum_{\vec{k}} \sum_{n,n'} \left[2 I_{n,n'}\left(\vec{k}, \vec{k}+\vec{q}\right) \right]^2 \frac{f_{\vec{k},n}(1 - f_{\vec{k}+\vec{q},n'})}{E_{\vec{k}+\vec{q},n'} - E_{\vec{k},n}} , \tag{3}$$

where the sum on N $\vec{k}$ values is over the whole Brillouin zone, and the $E_{\vec{k},n}$ are the energy eigenvalues with the Fermi occupation numbers $f_{\vec{k},n}$. $I_{nn'}(\vec{k}, \vec{k}+\vec{q})$ is the unscreened exchange matrix element which, for Gd metal with seven $4f$ electrons (^8S state), is given by

$$I_{nn'}(\vec{k}, \vec{k}+\vec{q}) = \frac{1}{7} \sum_{m=-3}^{+3} \iint \psi_{\vec{k},n}^*(\vec{r}_1) \varphi_{4f,m}^*(\vec{r}_2) \frac{2}{r_{12}} \varphi_{4f,m}(\vec{r}_1) \psi_{\vec{k}+q}(\vec{r}_2)\, d^3r_1\, d^3r_2 . \tag{4}$$

Here $\varphi_{4f,m}(\vec{r})$ is the $4f$ orbital in the metal with angular component m and the $\psi_{\vec{k},n}(\vec{r})$ are Bloch wave functions of wave vector $\vec{k}$ and band index n. For an hcp crystal structure with two atoms per unit cell, it is in general necessary to define a different $J(\vec{R}_{ij})$ for the interaction between spins on the same sublattice and between spins on different sublattices. However, for $\vec{q}$ along the z direction, the formulation is simplified by considering the double-zone scheme in which the Brillouin zone is extended (doubled) in the z direction. We have used the double-zone scheme and have restricted our calculations to $\vec{q}$ along

Γ to A to Γ in the double zone.

For our calculations the Bloch functions $\psi_{\vec{k},n}(\vec{r})$ were evaluated for the first six bands at 125 points in the irreducible $\frac{1}{24}$th of the Brillouin zone by use of the APW method. The details of this calculation and the characteristics of the exchange matrix elements have been described previously.[3] The matrix elements $I(\vec{k}, \vec{k}+\vec{q})$ were found to exhibit a great deal of structure as functions of both $\vec{k}$ and $\vec{q}$, and were largest for d-like conduction electrons. The structure is associated with band crossings and the orthogonality properties of the wave functions.[3]

In a pioneering calculation Liu, Gupta, and Sinha[7] evaluated $J(\vec{q})$ in Eq. (3) by using the energy eigenvalues obtained from a relativistic APW band-structure calculation[8] for Gd. They made the common assumption that the matrix elements $I(\vec{k}, \vec{k}+\vec{q})$ only depended upon $\vec{q}$, and furthermore assumed that the q dependence was Gaussian. They used only bands 3 and 4, which cross the Fermi level in their calculation. The rather good agreement between experiment and the calculations of Liu, and co-workers[7] is to some extent fortuitous, since most of their assumptions concerning the matrix elements were found to be false.

The wave-vector summation in Eq. (3) was carried out by use of the analytic tetrahedron-linear-energy method. This consists of breaking the Brillouin zone into microzones taken as tetrahedrons and assuming a linear variation of the energy eigenvalues and constant matrix elements within each microzone. With these assumptions the integration can be performed accurately with use of analytical expressions.[9,10] A total of 14000 k points in the whole Brillouin zone were considered for the bands crossing the Fermi level, and 25000 k points for other bands. The numerical accuracy of this method was tested and found to be at least as accurate as the method of Ref. 7.

The results of our calculations for $J(\vec{q})$ are shown in Fig. 1 and reveal several striking features.

(1) The bands crossing the Fermi surface are generally the most important; however, the total effect of interband scattering is for some q values just as large and cannot be neglected.

(2) The contribution to $J(\vec{q})$ from the bands crossing the Fermi surface deviates significantly with respect to the wave-vector dependence from the sum, Eq. (3), with $I_{nn'}(\vec{k}, \vec{k}+\vec{q}) = 1$. This means that the wave functions of the conduction electrons are at least as important as the Fermi-surface topology for determining the wave-vector dependence of the exchange interaction, and therefore the magnetic ordering in the rare-earth metals. A similar conclusion was reached for Cr by Winsor[11] using a tight-binding model for the matrix elements.

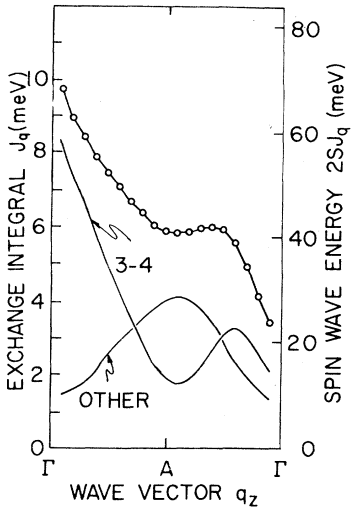

FIG. 1. The theoretical wave-vector–dependent exchange interaction J_q (left scale) and the spin-wave energy $2SJ_q$ (right scale) for Gd using the paramagnetic APW energy bands. The curve marked 3-4 is the contribution from the bands crossing the Fermi level and the "other" curve includes the rest of the contributions from the first six bands.

(3) The calculated magnitude of $J(\vec{q})$ is about a factor of 4 larger than the experimental value. From the transition temperature[1] we find $J_0{}^{T_c} = 3$ meV and the magnon spectrum[4] $2S(J_0 - J_{q\,\mathrm{zone}})^{\mathrm{exp}} = 14$ meV as compared with the theoretical values $J_0{}^{\mathrm{theory}} = 10$ meV and $2S(J_0 - J_{q\,\mathrm{zone}})^{\mathrm{theory}} = 50$ meV, respectively. This discrepancy is consistent with the fact that the calculated splitting[3] of the spin-up and spin-down bands is 1 eV, about twice the experimental splitting ~0.55 eV. The splitting is proportional to the s-f matrix element. The main wave-vector–dependent structure of the matrix elements originates from the orthogonality properties of the wave functions which make the matrix elements change drastically near band crossings. While radial wave functions contribute little to the q-dependent structure, they play an important role in determining the magnitude of the matrix element.

Although we have used the nonrelativistic APW method, it is known that the relativistic effects are important in determining the radial extent of the $4f$ orbitals.[12] Therefore the inaccuracies in

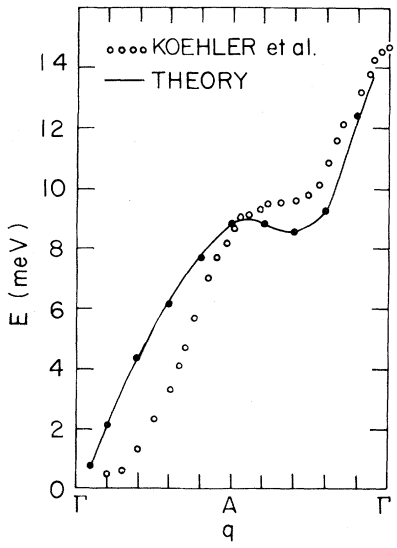

FIG. 2. The magnon spectrum obtained by Koehler *et al.* (Ref. 4) from neutron diffraction measurements and the theoretical result of this paper multiplied by a q-independent scale factor of 3.6.

the magnitude of our matrix elements are to some extent due to using nonrelativistic radial functions, and presumably to a greater extent due to neglecting screening effects in the matrix elements [Eq. (4)]. We believe that both these effects will act to scale the matrix elements rather than affect the $\vec{k}$ and $\vec{q}$ dependence. If we scale our $J(\vec{q})$ results with a $\vec{q}$-independent factor to obtain the experimental width of the magnon spectrum, our computed spectrum is in excellent overall agreement with experiment, as shown in Fig. 2. There remains a discrepancy at small $\vec{q}$ values however. The apparent lack of a q^2 dependence near Γ in the theoretical curve is simply because the calculations were not performed on a fine enough mesh near $q=0$. The apparent q^2 dependence of the experimental points over a large range of q values is only evident for the lower branch and is not present in other directions. We are presently studying the possibility of q-dependent screening and considering other mechanisms which may account for the behavior in the small-q region.

We have found that our calculated $J(\vec{q})$ is somewhat insensitive to fine details of the energy bands. The contribution to $J(\vec{q})$ from bands 3 and 4, which cross the Fermi level, is almost identical for the relativistic[8] and the nonrelativistic bands.[3] This insensitivity arises from the very dominant role played by the matrix elements, rather than the eigenvalues which enter Eq. (3). However, a rigid-band splitting of the bands by ± 0.021 Ry corresponding to that in the fully ordered magnetic state has a significant effect on $J(\vec{q})$. A similar effect was discussed for Gd, Tb, Dy, and Er with use of a simplified calculation.[13]

We have demonstrated that an *ab initio* calculation of the magnon dispersion curve for Gd metal using the actual band structure, wave functions, and the indirect (RKKY) exchange matrix elements, $I(\vec{k},\vec{k}')$, yields good agreement with experiment provided the matrix elements are scaled by a constant (q-independent) factor of about 2. The good agreement results from the dominant role played by the matrix elements (which themselves show great structure in both $\vec{k}$ and $\vec{q}$) and the inclusion of transitions between all bands below the Fermi energy to bands well above. The success of this calculation indicates that RKKY theory, when properly carried out, is probably a valid description of the exchange interaction in the rare-earth metals.

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