Exchange Interaction between Conduction Electrons and Magnetic
Shell Electrons in Rare-Earth Metals*

S. H. Liu[†]

Institute for Atomic Research and Department of Physics
Iowa State University, Ames, Iowa

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Energy Commission.

[†] International Business Machines Fellow, 1959-1960.

Abstract

The interaction Hamiltonian for the Coulomb exchange effect between
conduction electrons and magnetic shell electrons in rare-earth metals ^{was} ~~is~~
derived from first principles. The approximations under which the
interaction can be represented by the product of electron and ion spin
vectors are exhibited. (auth)

Introduction

The exchange interaction between conduction electrons and magnetic
shell electrons plays an important role in some of the electric and magnetic
properties of rare-earth metals. This type of interaction was first proposed

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by Zener¹ as a part of the mechanism of ferromagnetic coupling in the transition elements. Kasuya² suggested that this interaction gives the entire coupling between the ions in rare-earth metals, so that the crystal can become ferromagnetic or antiferromagnetic even though for most of the elements there apparently is very little overlapping between the magnetic 4f shells of neighboring ions. He also made a detailed calculation of the exchange interaction in gadolinium and put the interaction Hamiltonian in a form equivalent to $A \underline{s} \cdot \underline{S}$ where $\underline{s}$ is the spin vector of the conduction electron and $\underline{S}$ is the spin angular momentum of the ion. The problem for gadolinium is simple because there is no net orbital angular momentum and so $\underline{S}$ is the same as the total angular momentum $\underline{J}$. For other rare-earth metals de Gennes³ proposed that one may replace $\underline{S}$ by $(g-1)\underline{J}$, where g is the Landé factor, and obtain the form $A(g-1)\underline{s} \cdot \underline{J}$. Using this latter form he arrived at the Néel⁴ formula for the paramagnetic curie temperature of the rare-earth metals. Brout and Suhl⁵, following the suggestion of Herring⁶, proposed the same two forms $\underline{s} \cdot \underline{S}$ and $(g-1)\underline{s} \cdot \underline{J}$ and justified the use of the second form in case the multiplet splitting is large.

The anomalous resistivity in gadolinium and some other rare-earth metals has also been attributed to this exchange interaction. Kasuya⁷ and de Gennes and Friedel⁸ obtained theoretically the temperature dependence of the anomalous part of the resistivity of gadolinium. For other rare-earth metals Brout and Suhl⁵ derived a dependence of the saturation value of the anomalous resistivity on the number of electrons

in the 4f shell. The basic interaction Hamiltonian was taken as $(g-1)\underline{s} \cdot \underline{J}$. Their result is slightly different from the semiempirical dependence found by Anderson and Legvold⁹. However the uncertainty in the numerical results of the latter work was large, and so there is no decisive disagreement between the theory and the experiments. At temperatures low compared with the Curie or Néel temperature the anomalous resistivity has a T^2 dependence¹⁰. This was explained by Mannari¹¹ using a model of conduction electron-spin wave scattering through this exchange interaction.

The same exchange interaction exists between conduction electrons and rare-earth ions when these ions are dissolved in lanthanum. The net effect is that these dilute rare earth solutions have lower superconductive transition temperatures than pure lanthanum. The theoretical work of Suhl and Matthias¹² gave satisfactory explanation to the experimental results.

The effects of this interaction on other transport properties - thermal conductivity and thermoelectric power - were also studied by Kasuya¹³.

The present work is an extension of Kasuya's work to other rare-earth metals where the spin-orbit coupling in the ion should be taken into account. The basic interaction between the electrons is assumed to be the Coulomb exchange interaction. Under certain approximations, it is shown that the spin dependent part of the interaction Hamiltonian is of the form $(g-1)\underline{s} \cdot \underline{J}$. Hence this gives a fundamental proof of de Gennes' proposal.

Basic Model and Wave Functions

The model for rare-earth metals one usually employs consists of a lattice of trivalent ions in a sea of conduction electrons. Each ion has an unfilled 4f shell which is gradually filled up as the atomic number increases. There are filled 5s and 5p shells outside the 4f shell. This model describes all members of the group except the following: cerium has four conduction electrons at low temperatures; europium is divalent; and ytterbium is divalent and has a filled 4f shell. The element promethium is radioactive, so very little is known about its physical properties. The present discussion will exclude these exceptional cases.

The angular momentum and the magnetic moment of each ion are due entirely to the unfilled 4f shell since all the other shells are filled. Because of the shielding effect of the outer shells, the 4f shell has weak interactions with the surroundings. Also the crystalline field splitting is small compared with the multiplet splitting. Therefore in a first order theory one usually treats the ions as free. The electrons in a 4f shell couple their angular momenta together according to the Russell-Saunders scheme. The total angular momentum J is considered a good quantum number just as for a free ion. This picture of the magnetic shell structure is consistent with the data of paramagnetic susceptibility and saturation magnetic moment of these metals.

There are Coulomb forces between the conduction electrons and all the electrons in the ion core. However, since filled shells do not contribute any spin dependent effect, it is sufficient to consider only the

conduction electrons and the 4f shell electrons. To simplify the writing one may start by considering one conduction electron interacting with the magnetic electrons of one ion. The total interaction can be obtained by summing over all the conduction electrons and all the ions. Thus the interaction Hamiltonian may be written as

$$H_I = \sum_{i=1}^N \frac{e^2}{|\underline{r}_i - \underline{r}_{N+1}|} \quad (1)$$

where $\underline{r}_{N+1}$ is the position vector of the conduction electron and $\underline{r}_i$ is the position vector of the i-th magnetic shell electron. The summation is taken over all electrons in the shell.

The wave function for the conduction electron is of the form

$$\psi(\underline{r}, s) = u_{\underline{k}}(\underline{r}) \exp[i \underline{k} \cdot \underline{r}] \chi \quad (2)$$

which is the Pauli wave function for a Bloch wave normalized in a large volume. Here χ is the Pauli spinor. The wave function for the magnetic shell has a rather complicated structure. In the usual approximation in the theory of atomic spectra each electron in the 4f shell should have a wave function of the form

$$\psi_{l\mu}(\underline{r}) \chi = R(r) Y_{l\mu}(\theta, \phi) \chi \quad (3)$$

The wave function of the whole shell should be constructed from single particle wave functions according to the Pauli principle and Hund's rules as follows:

(a) When the magnetic shell is half filled or less than half filled ($N \leq 2l+1$), the wave function is of the form

$$\psi_{JM}(1, 2, \dots, N) = \sum_m C(L S J; m, M-m) \psi_{Lm}(1, 2, \dots, N) \times \psi_{S, M-m}(1, 2, \dots, N) \quad (4)$$

where $C(LSJ; m, M-m)$ denotes the vector coupling coefficients. The wave function ψ_{Lm} is constructed from single particle orbital wave functions $\psi_{\ell m}(\underline{r})$ and is completely antisymmetrical with respect to exchange of particles. The function $\psi_{S, M-m}$ contains only single particle spin functions and is completely symmetrical.

(b) When the magnetic shell is more than half filled ($N > 2\ell + 1$), the space and spin wave functions have more complicated symmetries. It is most convenient to express the symmetries by the Young diagrams¹⁴ shown in Fig. 1. One labels the electrons by $1, 2, \dots, N$ and arranges them in the frames such the first $(2\ell + 1)$ electrons are in the long column of the space diagram and the long row of the spin diagram. One first symmetrizes with respect to all particles in the same row and then antisymmetrizes with respect to all particles in the same column of the diagrams to obtain the wave functions $\psi_{Lm, t}(1, 2, \dots, N)$ and $\psi_{S, M-m, t}(1, 2, \dots, N)$. Here t denotes the complementary tableaux obtained by this arrangement of the particles in the frames. Similar terms can be obtained by arranging the particles in different ways in the same frames. The completely antisymmetric eigenfunction of $\underline{L}^2, L_z, \underline{S}^2, S_z$ can then be expressed by a sum of the form

$$\sum_t A_{mt} \psi_{Lm, t}(1, 2, \dots, N) \psi_{S, M-m, t}(1, 2, \dots, N)$$

where the summation is taken over all possible tableaux with the same frame. One should note that since the terms in the above sum are not linearly independent, the coefficients A_{mt} are not uniquely defined even though the sum is. Therefore the required wave function of the shell is

$$\psi_{JM} = \sum_{m, t} C(LSJ; m, M-m) A_{mt} \psi_{Lm, t} \psi_{S, M-m, t} \quad (5)$$

SPACE DIAGRAM

SPIN DIAGRAM

Fig. 1. The Young diagrams for space and spin symmetries of a more than half filled 4f shell.

Matrix Elements of the Coulomb Exchange Interaction

In this section the exchange part of the matrix elements of the interaction Hamiltonian (1) will be calculated using a wave function constructed from (2) and (4) or (5). The wave function of the system of one conduction electron and one magnetic shell is, with no regard to symmetry

$$\bar{\Psi} = \psi_{JM}(1, 2, \dots, N) \psi(N+1) \quad (6)$$

where $\psi(N+1)$ denotes $\psi(\underline{r}_{N+1}, s_{N+1})$ of the conduction electron. This wave function should be antisymmetrized with respect to all the $(N+1)$ particles, the result is

$$\bar{\Psi} = \frac{1}{\sqrt{N+1}} \left[\psi_{JM}(1, 2, \dots, N) \psi(N+1) - \sum_{i=1}^N \psi_{JM}(1, \dots, i-1, N+1, i+1, \dots, N) \psi(i) \right]$$

Now the particles are considered as completely indistinguishable, so the Hamiltonian (1) must also be symmetrized,

$$H_I = \sum_{i,j} \frac{e^2}{|\underline{r}_i - \underline{r}_j|} \quad (7)$$

where $1 \leq i, j \leq N+1$ and $i \neq j$. Consider the following (unsymmetrized) initial and final states

$$\begin{aligned} \bar{\Psi}_i &= \psi_{JM}(1, 2, \dots, N) \psi(N+1), \\ \bar{\Psi}_f &= \psi_{JM'}(1, 2, \dots, N) \psi'(N+1), \end{aligned} \quad (8)$$

where

$$\begin{aligned} \psi(N+1) &= u_{\underline{k}}(\underline{r}_{N+1}) \exp(i\underline{k} \cdot \underline{r}_{N+1}) \chi \\ \psi'(N+1) &= u_{\underline{k}'}(\underline{r}_{N+1}) \exp(i\underline{k}' \cdot \underline{r}_{N+1}) \chi'. \end{aligned} \quad (9)$$

The matrix element $(\bar{\Psi}_f, H_I \bar{\Psi}_i)$ of the Hamiltonian (7) contains the following groups of terms:

- (a) Direct interaction between shell electrons.
- (b) Exchange interaction between shell electrons.
- (c) Direct interaction between the conduction electron and the shell electrons.
- (d) Exchange interaction between the conduction electron and the shell electrons.

One is interested only in the last group of terms. If the totality of these terms is denoted by M_{ex} , it can be easily found that

$$M_{ex} = - \sum_i \int \Psi_{JM'}^*(1, 2, \dots, i, \dots, N) \Psi'^*(N+1) \frac{e^2}{|\underline{r}_i - \underline{r}_{N+1}|} \\ \times \Psi_{JM}(1, 2, \dots, N+1, \dots, N) \Psi(i) d\underline{r}_1 d\underline{r}_2 \dots d\underline{r}_{N+1} \quad (10)$$

This will be calculated separately for $N \leq 2\ell+1$ and $N > 2\ell+1$.

In the case of $N \leq 2\ell+1$, one substitutes Eq. (4) in Eq. (10), this gives

$$M_{ex} = - \sum_i \sum_{mm'} C(LSJ; m, M-m) C(LSJ; m', M'-m') \\ \int \Psi_{Lm'}^*(1, 2, \dots, i, \dots, N) \Psi_{S, M'-m'}^*(1, 2, \dots, i, \dots, N) \Psi'^*(N+1) \\ \times \frac{e^2}{|\underline{r}_i - \underline{r}_{N+1}|} \Psi_{Lm}(1, 2, \dots, N+1, \dots, N) \Psi_{S, M-m}(1, \dots, N+1, \dots, N) \\ \times \Psi(i) d\underline{r}_1 d\underline{r}_2 \dots d\underline{r}_{N+1} \quad (11)$$

For gadolinium the above expression is particularly simple because $L = 0$ and $S = J$. Hence one finds

$$\begin{aligned}
 M_{ex} = & - \sum_i \left[\int \psi_{00}^* (1, 2, \dots, i, \dots, N) u_{\underline{k}'}^*(\underline{r}_{N+1}) \exp(-i \underline{k}' \cdot \underline{r}_{N+1}) \right. \\
 & \frac{e^2}{|\underline{r}_i - \underline{r}_{N+1}|} \psi_{00} (1, 2, \dots, N+1, \dots, N) u_{\underline{k}}(\underline{r}_i) \exp(i \underline{k} \cdot \underline{r}_i) d\underline{r}_1 \dots d\underline{r}_{N+1} \\
 & \times \left[\psi_{SM'}^* (1, 2, \dots, i, \dots, N) \chi'^*(N+1) \chi(i) \right. \\
 & \left. \left. \psi_{SM} (1, 2, \dots, N+1, \dots, N) \right] \right].
 \end{aligned}$$

Due to the complete antisymmetry of the orbital wave function, the integral over the coordinate space is independent of i . One may denote it by $I'(\underline{k}, \underline{k}')$

In the product of spin functions one may replace $\chi'^*(N+1) \psi_{SM} (1, 2, \dots, N+1, \dots, N)$ by $\chi'^*(i) \psi_{SM} (1, 2, \dots, i, \dots, N)$ because the product contains only the spin functions of the rest $(N-1)$ particles. Hence the product can be written as

$$\langle SM' | \chi(i) \chi'^*(i) | SM \rangle.$$

The following four cases will be examined:

(a) $\chi = \chi' = \alpha$. It can be easily shown that

$$\chi(i) \chi'^*(i) = \frac{1}{2} [1 + \sigma_z(i)] = \frac{1}{2} + s_z(i).$$

(b) $\chi = \chi' = \beta$. In this case

$$\chi(i) \chi'^*(i) = \frac{1}{2} - s_z(i).$$

(c) $\chi = \beta, \chi' = \alpha$. In this case

$$\chi(i) \chi'^*(i) = s_-(i).$$

(d) $\chi = \alpha, \chi' = \beta$. In this case

$$\chi(i) \chi'^*(i) = s_+(i).$$

where $\underline{s}(i)$ is the spin vector of the i -th shell electron and $s_{\pm}(i) =$

$s_x(i) \pm i s_y(i)$. All these cases can be combined into one expression

$$\begin{aligned} & \langle SM' | \chi(i) \chi'^*(i) | SM \rangle \\ &= \langle SM' \chi' | \frac{1}{2} + 2 \underline{s} \cdot \underline{s}(i) | SM \chi \rangle \quad (12) \end{aligned}$$

where $\underline{s}$ is the spin vector of the conduction electron. Therefore

$$\begin{aligned} M_{ex} &= \sum_{\underline{k}, \underline{k}'} I'(\underline{k}, \underline{k}') \langle SM' \chi' | \frac{1}{2} + 2 \underline{s} \cdot \underline{s}(i) | SM \chi \rangle \\ &= I'(\underline{k}, \underline{k}') \langle SM' \chi' | \frac{N}{2} + 2 \underline{s} \cdot \underline{S} | SM \chi \rangle \end{aligned}$$

where $\underline{S} = \sum_i \underline{s}(i)$.

Hence the spin dependent part of the exchange interaction Hamiltonian may be taken as

$$H = -2 I'(\underline{k}, \underline{k}') \underline{S} \cdot \underline{S} = -2 I'(\underline{k}, \underline{k}') \underline{S} \cdot \underline{J} \quad (13)$$

This result was first derived by Kasuya² and is generally valid for any initial and final states of the conduction electron.

For other rare earths with $N < 2\ell + 1$, the Hamiltonian can be reduced to a form similar to Eq. (13) only under some restricted conditions. Each term in (11) contains an integral over the coordinate space of all the particles

$$\begin{aligned} I(\underline{k}, \underline{k}', m, m') &= \int \psi_{Lm}^*(1, 2, \dots, i, \dots, N) \psi_{\underline{k}'}^*(\underline{r}_{N+1}) \\ &\times \exp(-i \underline{k}' \cdot \underline{r}_{N+1}) \frac{e^2}{|\underline{r}_i - \underline{r}_{N+1}|} \psi_{Lm}(1, 2, \dots, N+1, \dots, N) \\ &\times \psi_{\underline{k}}(\underline{r}_i) \exp(i \underline{k} \cdot \underline{r}_i) d\underline{r}_1 \dots d\underline{r}_{N+1} \quad (14) \end{aligned}$$

and a product of spin functions

$$\psi_{S, M'-m'}^*(1, 2, \dots, i, \dots, N) \chi'^*(N+1) \chi(i) \psi_{S, M-m}(1, 2, \dots, N+1, \dots, N) \quad (15)$$

The latter can be reduced by a similar calculation to

$$\langle S, M'-m', \chi' | \frac{1}{2} + 2 \underline{S} \cdot \underline{S}(i) | S, M-m, \chi \rangle \quad (16)$$

In the space integral (14), one may expand

$$\psi_{Lm'}(1, \dots, i, \dots, N) = \sum_{\nu=-L}^L \Phi_{\nu}'(1, \dots, i-1, i+1, \dots, N) \psi_{L\nu}(\underline{r}_i)$$

and

$$\psi_{Lm}(1, \dots, N+1, \dots, N) = \sum_{\mu=-L}^L \bar{\Phi}_{\mu}(1, \dots, i-1, i+1, \dots, N) \psi_{L\mu}(\underline{r}_{N+1})$$

where

$$\bar{\Phi}_{\mu} = \int \psi_{Lm}(1, 2, \dots, N+1, \dots, N) \psi_{L\mu}^*(\underline{r}_{N+1}) d\underline{r}_{N+1}.$$

etc. Then one obtains

$$\begin{aligned} I(\underline{k}, \underline{k}', m, m') &= \sum_{\nu\mu} (\Phi_{\nu}', \bar{\Phi}_{\mu}) \\ &\times \int \psi_{L\nu}^*(\underline{r}_i) u_{\underline{k}'}^*(\underline{r}_{N+1}) \exp(-i\underline{k}' \cdot \underline{r}_{N+1}) \frac{e^2}{|\underline{r}_i - \underline{r}_{N+1}|} \\ &\psi_{L\mu}(\underline{r}_{N+1}) u_{\underline{k}}(\underline{r}_i) \exp(i\underline{k} \cdot \underline{r}_i) d\underline{r}_i d\underline{r}_{N+1} \end{aligned} \quad (17)$$

This last integral requires careful study. The conduction electrons are $6s^2$ and $5p$ electrons. If one considers that both the initial and the final states are in the s -band, the functions $u_{\underline{k}}(\underline{r})$ and $u_{\underline{k}'}(\underline{r})$ are isotropic and can all be approximated by $u_0(r)$ for $\underline{k} = 0$. The phase factor $\exp(i\underline{k} \cdot \underline{r}_i)$ can be expanded as

$$\exp(i\underline{k} \cdot \underline{r}_i) = 4\pi \sum_{l=0}^{\infty} \sum_{m=-l}^l i^l j_l(kr_i) Y_{lm}^*(\hat{\underline{k}}) Y_{lm}(\hat{\underline{r}}_i).$$

If the radius of the 4f shell is small compared with the wave length of the conduction electron, only the leading term will be of significance. If one is concerned only with the effect of the leading term, the integral can be evaluated by standard techniques to obtain

$$I(\underline{k}, \underline{k}', m, m') \cong \sum_{\mu\nu} (\Phi_\nu', \Phi_\mu) I(k, k') \delta_{\mu\nu} \\ = I(k, k') \langle L m' | L m \rangle \quad (18)$$

where

$$I(k, k') = \frac{4\pi e^2}{2l+1} \int R^*(r_i) R(r_{N+1}) U_0^*(r_{N+1}) U_0(r_i) \\ \times j_0(k' r_{N+1}) j_0(k r_i) \frac{(r_<)^l}{(r_>)^{l+1}} r_i^2 r_{N+1}^2 dr_i dr_{N+1}$$

Substituting (16) and (18) into (11), one obtains

$$M_{ex} = - \sum_i \sum_{mm'} C(L S J; m, M-m) C(L S J; m', M'-m') \\ \times I(k, k') \langle L m' | L m \rangle \\ \times \langle S, M'-m', \chi' | \frac{1}{2} + 2 \underline{S} \cdot \underline{S}(i) | S, M-m, \chi \rangle \\ = - I(k, k') \langle J M' \chi' | \frac{N}{2} + 2 \underline{S} \cdot \underline{S} | J M \chi \rangle, \quad (19)$$

Within the manifold of ground state J value, $\underline{S}$ may be replaced by its projection along $\underline{J}$, namely $(g-1)\underline{J}$ where g is the Landé factor. Therefore the spin dependent part of the Coulomb exchange interaction Hamiltonian is

$$H = -2 I(k, k')(g-1) \underline{S} \cdot \underline{J} \quad (20)$$

which is the de Gennes Hamiltonian. If either the initial or the final state or both of the conduction electron is a 5d state, the integral $I(k, k', m, m')$ can not in general be put into the simple form in Eq. (18). Then the Coulomb exchange interaction can no longer be expressed by a simple product of spin vectors.

In the case of $N > 2l+1$, one uses the wave function in Eq. (5). A similar calculation shows that the matrix elements of the interaction can be put into the form in Eq. (19) under the same approximations. Hence the Hamiltonian (20) also applies.

Discussion

It has been shown that the Coulomb exchange interaction between conduction electrons and 4f shell electrons in rare earths can be represented by a Hamiltonian $-2 I(k, k')(g-1) \underline{S} \cdot \underline{J}$ if the following approximations are made:

1. The conduction electrons are the s electrons so their wave functions have spherical symmetry.
2. The wave length of the conduction electron is large compared with the size of the 4f shell so the phase factor $\exp(i\mathbf{k} \cdot \mathbf{r})$ can be approximated by the leading term of its multipole expansion.

Hence a simple spin product Hamiltonian follows only if one neglects the dependence of the conduction electron wave functions on the direction in space.

The first approximation is rather difficult to justify because of the lack of knowledge about the various wave functions. Since most of these wave functions are oscillatory in space, so it is very important to know their space dependences to some detail. This is a task which is beyond the scope of this discussion.

By the method of screening constant of Pauling¹⁵ the size of the 4f shell is estimated to be about $0.4 \text{ \AA}$. Using a free electron model one estimates the wave number at the Fermi energy to be $k \cong 1.5 \times 10^8 \text{ cm}^{-1}$. Hence $k \cdot r \cong 0.6 < 1$. Therefore the second approximation should be fair.

The exchange integral $I(k, k')$ should be the same for all rare earths. Hence according to de Gennes³ the Curie (or Neel) temperature of rare metals should be proportional to $(g-1)^2 J(J+1)$. For the elements between Gd and Lu this reduces to

$$T_c \text{ (or } T_N) \propto S^2 \frac{J+1}{J},$$

which is the Neel formula⁴ and is verified experimentally except for Yb¹⁶.

For the elements from La to Sm the relationship reduces to

$$T_c \text{ (or } T_N) \propto S^2 \frac{J}{J+1}.$$

This however is not in agreement with the experiments. The saturation value of the anomalous resistivity in rare earths should be proportional to $(g-1)^2 J(J+1)$ according to Brout and Suhl⁵. This relationship is also

verified experimentally in the elements Gd-Lu but not in La-Sm. Hence one may conclude that in the trivalent metals praseodymium, neodymium and samarium the simple theories do not apply.

These three metals have other peculiar properties as well. It is known that they have strange crystal structures instead of the hexagonal closed-packed structure of gadolinium etc¹⁶. They also have different conduction band structures as exhibited by their Hall constants. Pr and Nd have positive Hall constants¹⁷, indicating hole conduction, and the Hall constant of Sm has a strange dependence on temperature and magnetic field¹⁸. These peculiar properties may have close connections with the failure of the simple theories.

On the other hand when these elements are dissolved in lanthanum the expected result of the exchange interaction is observable¹². Therefore, when the ions of these elements are put in proper surroundings the interaction Hamiltonian (20) does apply.

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