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Indirect Exchange Model for Ferromagnetic Metals

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The ferromagnetic properties of rare-earth metals and their alloys are discussed in terms of the indirect exchange model. It is shown by the molecular-field approximation that, in calculating the Curie temperatures of these metals, the simple theory of Fröhlich and Nabarro and Zener is applicable. The second-order energy terms calculated by Ruderman and Kittel, Kasuya, and Yosida are important in discussing the low-temperature properties. Some numerical results are obtained which are in good agreement with the experiments.

INTRODUCTION

THE theory of ferromagnetism has been based on the spin exchange interaction between magnetic atoms or ions. Heisenberg's[1] direct exchange interaction between nearest neighbors has been considered as the basic interaction that gives rise to ferromagnetism in transition elements. The necessary condition for this interaction to take place is that the magnetic electrons of neighboring ions have overlapping orbits. Consequently, this model does not seem to apply to the magnetic rare-earth metals in which the magnetic shells of different ions have little or no overlapping. Even for transition metals the recent calculation of Stuart and Marshall[2] shows that the Heisenberg's coupling is not sufficient to account for the observed magnetic properties.

The indirect exchange model was first proposed by Fröhlich and Nabarro[3] for the ferromagnetism of the nuclear-spin system in some metals. They showed that, due to the hyperfine interaction between the conduction electrons and the nuclear magnetic moments, the nuclear spins tend to line up ferromagnetically at low temperatures. The basic idea is as follows. The hyperfine interaction can be represented by the following interaction Hamiltonian

$$H_I = \sum_{i,j} A\,\mathbf{s}_i\cdot\mathbf{I}_j, \tag{1}$$

where $\mathbf{s}_i$ is the spin operator of the conduction electron and $\mathbf{I}_j$, the nuclear-spin operator. When the nuclear spins are aligned, this interaction produces an internal field that removes the spin degeneracy of the conduction electrons. As a result, the electrons will distribute themselves unevenly between the two spin states in order to minimize the total energy. It turns out that the total energy of the electron sea is lower in this case than in the case of unpolarized electrons. The latter case holds when the nuclear spins are oriented at random. Hence, at low enough temperatures, the ferromagnetic state is actually more stable. In 1946 Vonsovsky[4] suggested that, in magnetic metals, there exists an exchange interaction between the conduction electrons and the magnetic electrons in the ions. The Hamiltonian of this interaction was shown to be of the form

$$H_I = \sum_{i,j} A\,\mathbf{s}_i\cdot\mathbf{S}_j, \tag{2}$$

where $\mathbf{S}_j$ is the spin of the ion. This Hamiltonian has the same form as that in Eq. (1). Zener[5] independently proposed this interaction Hamiltonian and argued that a stable ferromagnetic state for the ionic spins should be possible at low temperatures as a result of the Fröhlich-Nabarro mechanism. This so-called indirect exchange interaction has been investigated in detail by Ruderman and Kittel,[6] Kasuya,[7] and Yosida.[8] These authors worked out a perturbation calculation up to the second order and showed that the effective coupling between different ions can be represented by the Hamiltonian

$$H_{\mathbf{S}\cdot\mathbf{S}} = -\sum_{i,j} J(R_{ij})\mathbf{S}_i\cdot\mathbf{S}_j, \tag{3}$$

where $J(R_{ij})$ is a function of R_{ij}, the distance between the ions. Due to the complicated space dependence of the coupling energy $J(R_{ij})$, it seems very difficult to obtain quantitative estimations of the important magnetic properties.

In this paper it is shown that the simple theory of Fröhlich and Nabarro and Zener does give good quantitative estimation of the Curie temperatures of rare-earth metals and their alloys if the experimental values of the electron-ion coupling energy are used. In the molecular-field approximation, the second-order energy terms tend to cancel out each other near the Curie temperature. At low temperatures, one must take into account the second-order terms in order to explain the spin-wave phenomena.

SIMPLE THEORY

In this section, a review of the simple theory based on the indirect exchange model is given. A solution of a magnetic metal in a nonmagnetic metal is considered. The ions of the solute and of the solvent are assumed to have the same volume. The total Hamiltonian of the material is assumed to contain the following terms:

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the kinetic energy of the electrons; the interactions between the magnetic ions and the external magnetic field, between electrons and magnetic ions, and between the electrons and the field. Hence

$$\mathcal{H}=\sum_i(p_i^2/2m)+\sum_{ij} I\Omega_0\mathbf{s}_i\cdot\mathbf{S}_j\delta(\mathbf{r}_i-\mathbf{R}_j) -\sum_j g\mu\mathbf{H}\cdot\mathbf{S}_j-\sum_i 2\mu\mathbf{H}\cdot\mathbf{s}_i, \quad (4)$$

where Ω_0 is the volume of the ion, I the energy of electron-ion coupling, μ the Bohr magneton, g the Landé factor, and $\mathbf{H}$ the external field. The position vectors of the electrons and the ions are denoted by $\mathbf{r}_i$ and $\mathbf{R}_j$, respectively. The electron-ion interaction is localized as indicated by $\delta(\mathbf{r}_i-\mathbf{R}_j)$. The free-electron model is used for the electrons. Hence, the wave function for an electron with wave vector $\mathbf{k}$ and spin σ is taken as

$$\psi(\mathbf{r})=\Omega^{-\frac{1}{2}}u_\sigma\exp(i\mathbf{k}\cdot\mathbf{r}), \quad (5)$$

where Ω is the volume in which the wave function is normalized, and u_σ is the Pauli spinor. From the Hamiltonian in Eq. (4) and the wave function in Eq. (5), one finds that an electron in the spin-up state has the energy

$$\epsilon_{k\uparrow}=(\hbar^2k^2/2m)+(I/2N)\sum_j S_j^z-\mu H,$$

and an electron in the spin-down state has

$$\epsilon_{k\downarrow}=(\hbar^2k^2/2m)-(I/2N)\sum_j S_j^z+\mu H, \quad (6)$$

where $N=\Omega/\Omega_0$, the number of ions in Ω. In later calculations, Ω will be taken as 1 cc, so N is the number of ions in a unit volume. The direction of H is taken as the z direction for convenience. Hence, when the ions are lined up ($\sum_j S_j^z\neq 0$), the effect of the electron-ion coupling is to produce an internal field that polarizes the electrons. The total effective field is

$$H_{\text{eff}}=H-(I/2\mu N)\sum_j S_j^z. \quad (7)$$

At 0°K, it can be shown that (see reference 8) the energy of the electron sea in the presence of this field is

$$\frac{1}{5\pi^2}\left(\frac{2m}{\hbar^2}\right)^{\frac{3}{2}}(E_F)^{\frac{5}{2}}-\tfrac{1}{2}\chi\left(H-\frac{I}{2\mu N}\sum_j S_j^z\right)^2, \quad (8)$$

where E_F is the Fermi energy and χ the paramagnetic susceptibility of the electron sea,

$$E_F=(\hbar^2/2m)(3\pi^2N')^{\frac{2}{3}}, \quad \chi=3\mu^2N'/2E_F,$$

and N' is the density of electrons. The result in Eq. (8) does not change appreciably with temperature due to the nature of the Fermi-Dirac statistics. The total energy of the electron-ion system is then

$$E=E_0-\tfrac{1}{2}\eta(\sum_j S_j^z)^2-\left(g\mu-\frac{I\chi}{2\mu N}\right)H\sum_j S_j^z-\tfrac{1}{2}\chi H^2, \quad (9)$$

where

$$E_0=\frac{1}{5\pi^2}\left(\frac{2m}{\hbar^2}\right)^{\frac{3}{2}}(E_F)^{\frac{5}{2}}$$

and

$$\eta=\chi\left(\frac{I}{2\mu N}\right)^2=\frac{3N'}{2E_F}\left(\frac{I}{2N}\right)^2. \quad (10)$$

In Eq. (9), the term quadratic in H is the energy due to the paramagnetism of the free electrons. From the term linear in H one finds that, because of the polarization of the electrons, the g factor of the ions are modified to

$$g'=g-\frac{I\chi}{2\mu^2N}=g-\frac{3N'I}{4E_FN}. \quad (11)$$

Since $\eta>0$, the term $-\tfrac{1}{2}\eta(\sum_j S_j^z)^2$ in Eq. (9) indicates that a state of ordered spin ($\sum_j S_j^z\neq 0$) has lower energy than a state of random spin ($\sum_j S_j^z=0$). This is the fundamental mechanism that makes a ferromagnetic state more stable at low temperatures than a random spin state or an antiferromagnetic state.

At $T=0$°K, the energy is a minimum when all the spins are aligned. Taking N as the number of ions per cc, one finds the spontaneous magnetization to be

$$M_0=g'\mu S\xi N, \quad (12)$$

where ξ is the concentration of magnetic ions. Using Eq. (11), one finds that

$$M_0=M_i+M_e, \quad (13)$$

where M_i is the ionic contribution given by

$$M_i=g\mu S\xi N,$$

and M_e is the electronic contribution given by

$$M_e=-3N'I\mu\xi S/4E_F.$$

At finite temperatures, the ionic spins are no longer completely aligned, due to thermal excitation. If one identifies $g'\mu\sum_j S_j^z$ as the spontaneous magnetization, then the energy associated with the alignment of spins is $-\tfrac{1}{2}\eta(g'\mu)^{-2}M^2$. Hence, the quantity $\eta(g'\mu)^{-2}$ can be identified as the molecular-field constant in the conventional theory. From the relationship between the Curie temperature T_c and the molecular-field constant, one finds

$$T_c=S(S+1)\xi\eta N/3k, \quad (14)$$

where k is the Boltzmann constant.

For alloys of two magnetic metals, it can be easily shown that the Curie temperature should depend linearly on the composition, i.e.,

$$T_c=\xi T_{c1}+(1-\xi)T_{c2}, \quad (15)$$

where T_{c1} and T_{c2} are the Curie temperatures of the two metals in pure state. In an actual alloy system, the linear relationship may be distorted by other effects.

MORE RIGOROUS THEORY

Ruderman and Kittel,[6] Kasuya,[7] and Yosida[8] have calculated the energy of indirect exchange up to the second order of perturbation. The result given by Yosida is

$$E=E_0-g'\mu H\sum_j S_j{}^z-\tfrac{1}{2}\chi H^2-\frac{3N'}{32E_F}N^{-2}$$
$$\times\sum_m\sum_n\sum_{\mathbf{q}} I^2(\mathbf{q})f(q)\exp(i\mathbf{q}\cdot\mathbf{R}_{nm})\mathbf{S}_n\cdot\mathbf{S}_m, \quad (16)$$

where

$$f(q)=1-\frac{4k_m{}^2-q^2}{4k_mq}\ln\frac{2k_m-q}{2k_m+q}, \quad \mathbf{R}_{nm}=\mathbf{R}_m-\mathbf{R}_n,$$

and k_m is the size of the wave vector at Fermi level. The electron-ion interaction is, strictly speaking, not localized. Hence, the coupling constant I should depend on the initial and final values of the electron momentum. However, one usually makes the simplified assumption that I depends only on the momentum transfer, which is characterized by the wave vector $\mathbf{q}$. Thus, $\mathbf{q}$ is a lattice vector in the reciprocal lattice.

The last term in the above expression can be separated into two parts according to whether $m=n$ or $m\neq n$. The first part is

$$E_0'=-\frac{3N'}{32E_F}N^{-2}\sum_n\sum_{\mathbf{q}} I^2(\mathbf{q})f(q)\mathbf{S}_n\cdot\mathbf{S}_n$$
$$=-\frac{3N'}{32E_F}N^{-1}S(S+1)\sum_{\mathbf{q}} I^2(\mathbf{q})f(q), \quad (17)$$

which corresponds to a shift in total energy of the crystal independent of the spin ordering. The second part is the coupling term between the ions and can be written as

$$E_{\mathbf{s}\cdot\mathbf{s}}=-\sum_n \mathbf{S}_n$$
$$\cdot\left[\sum_{m,m\neq n}\sum_{\mathbf{q}}\frac{3N'}{32E_F}N^{-2}I^2(\mathbf{q})f(q)\exp(i\mathbf{q}\cdot\mathbf{R}_{nm})\mathbf{S}_m\right]. \quad (18)$$

Hence, one finds that the molecular field acting on the spin $\mathbf{S}_n$ is

$$\mathbf{H}_n=\frac{3N'}{16E_F}N^{-2}(g'\mu)^{-1}$$
$$\times\sum_{m,m\neq n}\sum_{\mathbf{q}} I^2(\mathbf{q})f(q)\exp(i\mathbf{q}\cdot\mathbf{R}_{nm})\mathbf{S}_m. \quad (19)$$

Near the Curie point, the spins are very much at random, so one may substitute for $\mathbf{S}_m$ in Eq. (19) the mean value $\langle\mathbf{S}\rangle$ of all the spins. In an alloy where not all the ions are magnetic, one has to make another averaging process. If the concentration of magnetic ions is ξ, then the probability that there is a magnetic ion at the site $\mathbf{R}_m$ is ξ. Hence, in this case, one should replace $\mathbf{S}_m$ by $\xi\langle\mathbf{S}\rangle$. Thus,

$$\mathbf{H}_n=\frac{3N'}{16E_F}\xi N^{-2}(g'\mu)^{-1}\langle\mathbf{S}\rangle\sum_{m,m\neq n}\sum_{\mathbf{q}} I^2(\mathbf{q})f(q)\exp(i\mathbf{q}\cdot\mathbf{R}_{nm}).$$

Carrying out the summation over m, one finds that

$$\sum_{m,m\neq n}\exp(i\mathbf{q}\cdot\mathbf{R}_{nm})=(N-1)\delta_{q0},$$

because $\mathbf{q}$ is a reciprocal lattice vector. Hence,

$$\mathbf{H}_n=\frac{3N'}{16E_F}N^{-2}(g'\mu)^{-1}\xi\langle\mathbf{S}\rangle I^2(0)f(0)(N-1).$$

Since $f(0)=2$, $I^2(0)=I^2$, $N-1\cong N$, one obtains

$$\mathbf{H}_n=\eta(g'\mu)^{-2}\mathbf{M}, \quad (20)$$

where

$$\mathbf{M}=g'\mu\xi N\langle\mathbf{S}\rangle.$$

The molecular-field constant is, therefore,

$$\lambda=\eta(g'\mu)^{-2},$$

which is just what has been found in the last section. Therefore, as far as the calculation of the Curie temperature is concerned, the simple theory gives the correct result.

At low temperatures, the excitation of the spin system is better described by spin waves. For a spin-product Hamiltonian of the form appropriate to indirect exchange coupling

$$H_{\mathbf{s}\cdot\mathbf{s}}=-\sum_n\sum_{m,m\neq n} J(\mathbf{R}_{mn})\mathbf{S}_m\cdot\mathbf{S}_n,$$

one can readily show that, in a pure material, the dispersion relation for the spin waves is

$$E(\mathbf{q})=2S[\mathcal{J}(0)-\mathcal{J}(\mathbf{q})], \quad (21)$$

where

$$\mathcal{J}(\mathbf{q})=\sum_m J(\mathbf{R}_{mn})\exp(-i\mathbf{q}\cdot\mathbf{R}_{mn}).$$

Physically, the ground state of the system is ferromagnetic if $\mathcal{J}(\mathbf{q})$ has a maximum at $\mathbf{q}=0$. Therefore, for small q, one must have

$$E(q)=\zeta q^2,$$

where

$$\zeta=-\frac{1}{2}\frac{\partial^2}{\partial q^2}\mathcal{J}(\mathbf{q})>0.$$

This dispersion relation leads to the $T^{\frac{3}{2}}$ law for the dependence of the spontaneous magnetization on temperature.

It should be noted that the above result holds only if $\mathcal{J}(\mathbf{q})$ varies smoothly with $\mathbf{q}$ in the neighborhood of $\mathbf{q}=0$. From Yosida's formula Eq. (16), one finds

$$\mathcal{J}(\mathbf{q})=\frac{3N'}{32E_F}N^{-1}I^2(\mathbf{q})f(q),$$

which satisfies the requirement. However, from Eq. (9), one finds that the simple theory gives

$$\mathcal{J}(\mathbf{q})=\tfrac{1}{2}\eta\delta_{q0},$$

which is singular at $\mathbf{q}=0$. Therefore, without the second-order terms, the theory gives an incorrect description of the magnetic properties of the metal at low temperatures.

Physically the second-order terms have an oscillatory space dependence. As a result they only contribute when the spin arrangement deviates from a perfectly ordered state in a periodic fashion. This condition does not hold either at absolute zero or near the Curie point. Therefore the Curie temperature, which depends on the difference in energy between these two states, is correctly estimated by ignoring these terms. However, when the spins undergo a collective motion as in spin waves, the condition of periodic spin deviation is satisfied. Hence, the second-order terms are important in this case.

DISCUSSION

For pure gadolinium, the Curie temperature is 289°K. From Eq. (14), with

$$\xi=1, \quad N=3.0\times10^{22} \text{ per cc}, \quad S=7/2,$$

one finds

$$\eta=2.54\times10^{-37}.$$

From Eq. (10), with $N'=3N=9.0\times10^{22}$ per cc and an effective mass ratio of unity, one finds

$$\begin{aligned}|I|&=2.78\times10^{-13} \text{ erg}\\&=0.174 \text{ ev}.\end{aligned}$$

The interaction Hamiltonian (2) has also been used by Suhl and Matthias[9] to explain the reduction of superconducting transition temperature of lanthanum when there is gadolinium impurity; and by Kasuya[10] and de Gennes and Friedel[11] to explain the anomalous resistivity of magnetic metals. In the first case, the estimated value of I given by reference 9 is

$$|I|=0.165 \text{ ev}.$$

In the second case, one estimates

$$|I|\cong0.2 \text{ ev}$$

from the formula in references 10 and 11. The value of I measured by Jaccarino *et al.*[12] is about 0.1 ev.

The Landé factor of the ions is

$$g'=g-(3N'I/4E_FN),$$

from Eq. (11). Using $I=-0.174$ ev and $g=2$, one finds

$$g'-g=0.055.$$

From the experimental value[13] of the effective number of Bohr magnetons per ion, one finds that

$$g'-g=0.04\pm0.01.$$

The agreement is reasonably good because this quantity itself is of the same order of magnitude as the experimental error. The contribution of the free electrons to the magnetic moment amounts to approximately 2% of the total value. In order to be consistent with the experiment the sign of I assumed here is opposite to that measured by Jaccarino *et al.*

For solutions of magnetic metals in nonmagnetic metals, Eq. (14) shows that the Curie temperature is directly proportional to the concentration of magnetic ions provided that the crystal constants do not change appreciably with composition. This behavior has been observed in $GdRu_2$-$CeRu_2$ alloys[14] and Gd-Y alloys.[15] From the T_c vs concentration curve of the latter group, one finds

$$|I|=0.21 \text{ ev}.$$

The alloys of gadolinium and lanthanum show more complicated behavior.[15]

For rare-earth metals other than gadolinium, the spin-orbit coupling in the magnetic shell is important. The appropriate Hamiltonian for the electron-ion interaction has been shown to be[16–18]

$$H_I=I(g-1)\mathbf{s}\cdot\mathbf{J}, \tag{22}$$

where J is the total angular momentum of the ion. The coupling constant I is approximately the same for all the elements. Therefore, the Curie temperatures of these elements are given by

$$T_c=(g-1)^2J(J+1)\eta N/3k. \tag{23}$$

Since these elements have nearly the same lattice parameters, η and N are roughly the same for all of them. Hence, one obtains

$$T_c\propto(g-1)^2J(J+1), \tag{24}$$

which is just the Néel[19] and de Gennes[16] formula for the Curie temperature of rare-earth metals.

From Eqs. (11) and (22), one finds that the change in Landé factor for rare-earth metals is

$$\Delta g=g'-g=-(3N'I/4E_FN)(g-1). \tag{25}$$

The calculated values of Δg using $I=-0.174$ ev are compared with the experimental values in Table I. The experimental values for Gd, Tb, and Dy are calculated from the saturation magnetizations at 0°K; the others are calculated from the paramagnetic

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TABLE I. The change in g factor due to the polarization of conduction electrons in rare-earth metals; the theoretical values are calculated from Eq. (25) with $I=-0.174$ ev.

Element	Δg (theoret)	Δg (exp)
Gd	0.055	0.04 ± 0.01
Tb	0.028	0.04 ± 0.01[a]
Dy	0.018	0.017 ± 0.009[b]
Ho	0.014	0.03[c]
Er	0.011	0.05[c]
Tm	0.009	0[d]

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susceptibilities. The theoretical and the experimental values agree in order of magnitude.

Many rare-earth metals and alloys undergo a spontaneous magnetic transition from ferromagnetic to antiferromagnetic ordering. However, these materials obey the Curie-Weiss law in the paramagnetic temperature region with positive Curie points. This is a good indication that the basic interaction in the materials is ferromagnetic. In such cases, Eqs. (14) and (23) should apply to the paramagnetic Curie temperatures rather than the Néel temperatures.

The indirect exchange model may apply to the transition elements as well. However, the problem is hard to analyze because of the complicated band structures.

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Current-Carrier Transport with Space Charge in Semiconductors

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Differential equations are given for a general formulation of current-carrier transport that includes space charge. Arbitrary dependences of diffusivities and magnitudes of drift velocities on electrostatic field are considered, and extension is made for applied magnetic field. Though excess electron and hole concentrations are not equal, the small-signal recombination rate depends on a single lifetime, the "diffusion-length lifetime," τ_0. The formulation is applied to one-dimensional drift with recombination for an injected pulse of electron-hole pairs. The exact electron and hole distributions are obtained in closed form for the linear small-signal case. The condition for linearity is given; it is usually the same as that for substantially unperturbed applied field, E_0. There are two principal types of solution, essentially according to whether τ_0 is larger or smaller than the dielectric relaxation time, τ_d. For $\tau_0 > \tau_d$, the electron and hole distributions in not too strongly extrinsic material are ultimately similar Gaussian distributions displaced by the "polarization distance," x_P, the distance electrons and holes drift apart in time $(\tau_d^{-1} - \tau_0^{-1})^{-1}$. These distributions drift at a velocity that differs from the ambipolar velocity by an amount which, besides being small for small τ_d/τ_0, vanishes for equal mobilities. They spread, exhibiting an apparent diffusion. A "pseudodiffusivity," D_v, is defined. For $\tau_0 \gg \tau_d$ and constant mobilities, D_v is proportional to $\tau_d E_0^2/\sigma_0^2$, with σ_0 the conductivity. The ambipolar diffusivity and D_v are additive. They are equal in intrinsic material for E_0 equal to kT/e divided by the Debye length $(kT\epsilon/8\pi n_i e^2)^{\frac{1}{2}}$, or 10 v/cm for silicon at 300°K. An extension to a nonlinear case involving high-level injection is given; concentration-dependent D_v and velocity function are defined. For sufficiently strongly extrinsic material and $\tau_0 > \tau_d$, the minority carriers drift in a delta pulse that leads the majority carriers distributed in an exponential tail of characteristic length x_P, which may be quite large. For nonconstant mobilities and $\tau_0 > \tau_d$, ambipolar velocity in the majority-carrier or "reverse" direction may occur. For $\tau_d > \tau_0$, the other principal type of solution gives distributions that in general (and for constant mobilities) drift in the reverse direction. Involving also regions of local carrier depletion, and thus generation as well as recombination, these distributions may persist for times long compared with τ_0, being attenuated then with time constant τ_d.

1. INTRODUCTION

WITH carrier injection and transport in semiconductor material of high resistivity, the widely used approximation of local electrical neutrality frequently does not apply. Thus, for various experiments and for a number of devices, including semiconductor detectors of nuclear particles, solutions are needed that take space charge into account. Extending results previously reported,[1] this paper presents a general formulation of transport with space charge, including applied magnetic field, and gives solutions for various cases of one-dimensional drift with recombination. An injected pulse of electron-hole pairs is considered. For linear small-signal cases, with relatively small perturbation of applied electrostatic field, exact solutions are obtained

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