Dilute ferromagnetic alloys with long-range exchange interaction

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We investigate the properties of dilute alloys of nonmagnetic metals or of nondegenerate semiconductors with magnetic impurities. Two situations are considered, in which indirect exchange interaction via the conduction electrons leads to ferromagnetic order in the alloys: 1) the characteristic period of the Kittel-Ruderman potential is larger than the distance r_c between the impurities, 2) the exchange potential consist of two parts, one with a large and the other with a small period of the spatial oscillations compared with r_c. It is shown that at low temperatures most per-unit thermodynamic quantities and the electric resistivity contain increments proportional to the temperature and due to local spin flips. These increments decrease exponentially with increase of the impurity density n_i in the first case and are independent of n_i in the second case.

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1. Magnetic ordering in alloys of most nonmagnetic metals is due to an indirect exchange interaction, that oscillates with the distance, between the impurity spins via the conduction electrons of the matrix. The period of the spatial oscillations of the potential is of the order of k_0^{-1}, where k_0 is the characteristic momentum determined by the topology of the Fermi surface,[1]; this potential has the ferromagnetic sign at $r \ll k_0^{-1}$. Therefore the type of ordering depends on the concentration and the spectrum of the electrons. If the characteristic momentum k_0 for a given impurity density n_i for most electrons is such that $k_0 n_i^{-1/3} \gg 1$, then an ordering of the "magnetic glass" type[2] is produced in the alloy at sufficiently low temperature. But if $k_0 n_i^{-1/3} \ll 1$, then most impurity spins are ferromagnetically ordered. It is then necessary to distinguish between two cases.

First, the smallness of k_0 may be due to the low density of the conduction electrons. In this case the interaction energy of spins separated by a distance of the order of or smaller than the average distance $r_c \approx n^{-1/3}$ is positive, and the isolated spins farther away than $k_0^{-1} \gg r_c$ from the neighbors may be in a zero or negative molecular field.

The contribution made by spin flips whose energy in the molecular field is of the order of the temperature T to most thermodynamic quantities and to the kinetic coefficients is proportional to the temperature and decreases exponentially with increasing impurity concentration.

Second, the smallness of k_0 may be connected with the complicated shape of the Fermi surface. It is known[1] that in this case $\mathbf{k}_0$ is a vector joining on the Fermi surface points the normals to which have opposite directions. There may be several such vectors, so that the interaction potential can be approximated by a sum of several terms of the RKKY type, each characterized by a different $\mathbf{k}_0$. We consider a situation in which several of the characteristic momenta are smaller than $n_i^{1/3}$ and the others are larger. Then the main magnetic properties of the alloy can be understood by representing the interaction potential as a sum of two terms, one of which, $V_f(r)$, is characterized by large-scale fluctuations of period $k_1^{-1} > n_i^{-1/3}$, and the other, $V_a(r)$, by small-scale fluctuations of period $k_2^{-1} < n_i^{-1/3}$. The ferromagnetic ordering arises in the system when the molecular field produced by the potential V_f is larger than $V_a(R_c)$. Since $V_a(r)$ increases with decreasing r, the inverse inequality may hold at $r \ll r_c$, i.e., the potential of the antiferromagnetic interaction of a pair of closely-lying spins may differ little from the energy of the spin in the molecular field. The spin levels in the pair are then close to each other and the transitions between them determine the behavior of the thermodynamic quantities. The corresponding contribution per impurity is proportional to T and inversely proportional to n_i.

Thus, increments that are anomalous in comparison with the ordered ferromagnet are added to the thermodynamic and kinetic quantities in both cases; these increments can exceed the ordinary spin terms at low temperatures.

2. Impurity ferromagnetism of GeTe alloys with Mn was observed in[2]. The ferromagnetic ordering is brought about in this case by the first of the mechanisms considered by us, namely, indirect exchange via free carriers in the matrix (holes), with density on the order of 10^{21} cm^{-3}, so that the parameter $k_0 r_c < 1$ for the samples used in[3], with impurity concentration ~1 at.%. The results obtained by us in Sec. 4 are fully applicable to these alloys. It is however impossible to compare them with experiment at present, since there are no data for sufficiently low temperatures.

In the alloys $La_{1-x}Gd_x$ ($x \lesssim 6$ at.%) there was observed[4] an anomalous behavior of the magnetic susceptibility at low temperatures; this behavior seems to indicate a ferromagnetic transition. At a certain temperature, the magnetic susceptibility has a sharp maximum, and in the high-temperature region it obeys the Curie-Weiss law $\chi^{-1} \sim T - \Theta$, with $\Theta \sim x$. Since the Fermi surface of lanthanum is quite complicated and has many sections with small characteristic momenta, the ferromagnetic character of the indirect exchange may be due to one of the mechanisms indicated above. The same situation seems to obtain apparently in $LaAl_2$–$GdAl_2$ alloys, in which ferromagnetic order was observed recently.[4]

Measurement of the temperature and concentration dependences of various physical quantities and their comparison with theory would make it possible to understand the mechanism that produces ferromagnetism in these alloys.

3. The indirect interaction of the localized alloys is described by the Hamiltonian

$$\mathscr{H}=-{}^1/{}_2\sum_{i,j}V(\mathbf{r}_i-\mathbf{r}_j)\,(\mathbf{S}_i\mathbf{S}_j). \tag{1}$$

Here i and j are sites at which magnetic impurities are located, and S_i is the spin localized at the site i. The potential V(r) for a spherical Fermi surface is given by[6]

$$V(r)=V_0\frac{\sin 2k_Fr-2k_Fr\cos 2k_Fr}{(k_Fr)^4},\quad V_0\approx\frac{I^2}{\varepsilon_F}\tilde{n}_e^2 \tag{2}$$

where $\tilde{n}_e$ is the number of electrons per matrix atom, k_F is the Fermi momentum, and I is the s-d exchange interaction.1)

If $k_Fr_c < 1$, then $V(r_c) > 0$ and the Hamiltonian (1) leads to ferromagnetic ordering, with many impurities contained in a sphere of radius k_F^{-1}. The Curie temperature can therefore be determined by the molecular-field method:

$$T_c={}^2/{}_3S(S+1)\nu V_0, \tag{3}$$

where

$$\nu=4\pi n_i/k_F^3\gg 1. \tag{4}$$

Owing to this inequality, the fluctuations of the interaction energy of a given spin with the surrounding are small, despite the randomness in the spin distribution. The spin-wave spectrum is therefore determined in the usual manner:

$$\omega_q=2S\nu V_0(1-{}^1/{}_2f(q/2k_F)). \tag{5}$$

For a spherical Fermi surface we have

$$f(x)=1+\frac{1-x^2}{2x}\ln\left|\frac{1+x}{1-x}\right|.$$

The damping of the spin waves is small if their wave vector $q \ll r_c^{-1}$.[7] At small momenta $q \ll 2k_F$ it follows from (5) that

$$\omega_q=\Theta(q/2k_F)^2, \tag{6}$$

where $\Theta = ({}^4/{}_3)\nu SV_0$. The density of states per impurity, of spin waves with frequency $\omega \ll \Theta$, is given by

$$\rho(\omega_q)=\frac{1}{2\pi^2n_i}q^2\frac{dq}{d\omega_q}=\frac{8}{\pi\nu}\frac{\omega_q^{1/2}}{\Theta^{3/2}}. \tag{7}$$

The appearance of the factor ν^{-1} in this expression for ρ is due to the long-range character of the interaction potential.

In ordered ferromagnets, the spin waves determine completely the magnetic part of the thermodynamic quantities at $T \ll T_C$. In this case, owing to the random distribution of the impurities, an important role may be played by the additional contribution connected with local spin flips. We now proceed to calculate this contribution.

4. We consider first a situation in which there is one characteristic momentum k_0, and $k_0n_i^{-1/3} < 1$. To take local spin flips into account, we can use the molecular-field approximation, so that the Hamiltonian (1) takes the form

$$\mathscr{H}=-\sum\omega_jS_j^z, \tag{8}$$

where

$$\omega_j=\sum_j V(\mathbf{r}_i-\mathbf{r}_j)\langle S_j^z\rangle, \tag{9}$$

z is the magnetization direction.

Since most spins are oriented at $T \ll T_C$ in parallel and are magnetized almost to saturation, $\langle S_j^z\rangle$ in (9) can be replaced by S. We introduce the molecular-field distribution function

$$W(\omega)=\frac{1}{v^N}\int d^3\mathbf{r}_1\ldots d^2\mathbf{r}_N\delta\Big(\omega-\sum_j\omega_j\Big). \tag{10}$$

Here v is the volume of the crystal and N is the total number of spins. After the usual transformations we obtain

$$W(\omega)=\frac{1}{2\pi}\int_{-\infty}^{\infty}d\rho e^{-i\rho\omega+D(\rho)}. \tag{11}$$

Here

$$D(\rho)=n\int d^3r(e^{iSV(r)\rho}-1)=\nu\int dx\,x^2(e^{iV(x)\rho}-1), \tag{12}$$

where $x = k_Fr$ and ν is determined by (4).

We obtain first W(0). This quantity determines the concentration dependence of the thermodynamic quantities at low temperatures $T \ll V_0$. Since $\nu \gg 1$, we can calculate W(0) by the steepest-descent method.

We shall show that in the complex ρ plane the derivative $D'(\rho)$ vanishes at a certain point on the imaginary axis. From the condition $D'(\rho) = 0$ we have

$$\int_0^\infty dx\,x^2V(x)e^{-\rho_2SV(x)}\cos(V(x)\rho_1)=0,$$

$$\int_0^\infty dx\,x^2V(x)e^{-\rho_2SV(x)}\sin(V(x)\rho_1)=0,$$

where $\rho = \rho_1 + i\rho_2$. On the imaginary axis the second equation is satisfied identically, and ρ_2 is determined from the equation

$$f(\rho_2)=S\int_0^\infty dx\,x^2V(x)e^{-\rho_2SV(x)}=0. \tag{13}$$

It is easily seen that this equation has a solution, and furthermore a unique one. Indeed, f(0) is proportional to the average molecular field and is consequently positive. At large ρ_2 the main contribution to the integral (13) is made by the regions of the variable x in which $V(x) < 0$. Consequently, at large ρ_2 we have $f(\rho_2) < 0$. The derivative $f'(\rho_2) < 0$. Thus, $f(\rho_2)$ decreases monotonically with increasing ρ_2 and vanishes at a certain $\rho_2 = \rho_0 > 0$. Since

$$\left.\frac{\partial^2D(\rho)}{\partial\rho^2}\right|_{\rho=i\rho_2}<0,$$

the direction of the steepest descent is the straight line $\rho_2 = \rho_0$ parallel to the real axis. Thus,

$$W(0)=e^{D(\rho_0)}/(2\pi|D''(\rho_0)|)^{1/2}. \tag{14}$$

It is easily seen that $D(\rho_0) < 0$. This follows from the fact that $D(0) = 0$ and $\partial D(\rho_2)/\partial\rho_2 = -f(\rho_2) < 0$ at $\rho_2 < \rho_0$. Therefore

$$W(0)\sim e^{-\alpha\nu}/SV_0\nu^{1/2}, \tag{15}$$

where α is a number of the order of unity. This conclusion does not depend on the concrete form of the potential V(r). Expressions (14) and (15) are valid if the following conditions are satisfied: $\nu \gg 1$, V(x) is an alternating-sign function of x, and $\int_0^\infty dx\,x^2V(x) > 0$.

Equation (13) with potential (2) was solved numerically. The result was

$$\rho_0=\frac{1.50}{SV_0},\quad D(\rho_0)=0.845\nu,\quad D''(\rho_0)=-0.371(SV_0)^2\nu,$$

so that

$$W(0)=\frac{0.654}{SV_0\nu^{1/2}}e^{-0,845\nu}. \tag{16}$$

At low frequencies $\omega \ll \nu SV_0$ we can calculate $W(\omega)$ as before by the steepest-descent method. The saddle point is now determined from the equation

$$\nu S\int dx\,x^2V(x)\exp[-V(x)\rho_0(\omega)S]=\omega. \tag{17}$$

The quantity ρ_0 decreases with increasing ω. If $\omega \ll \nu SV_0$, then $\rho_0(\omega)$ differs little from $\rho_0(0)$ and Eq. (17) can be solved by iterating with respect to the small parameter $\omega/\nu V_0$. As a result we get

$$W(\omega)=W(0)\exp\left\{\omega\rho_0(0)-\frac{\omega^2}{2\nu|D''(\rho_0)|}\right\}. \tag{18}$$

We have discarded in the argument of the exponential the terms proportional to ω^3, which is justified if $\omega \ll \nu^{2/3}SV_0$.

At frequencies ω close to the average molecular field energy $\overline{V} = 2\nu SV_0$ we can obtain $W(\omega)$ by expanding the exponential in (12) in powers of $V(x)$ accurate to $V^2(x)$. We obtain for $W(\omega)$, naturally, the Gaussian distribution

$$W(\omega)=\sqrt{\frac{15}{\pi\nu}}\frac{1}{8SV_0}\exp\left\{-\frac{(\omega-\overline{V})^2}{\Delta}\right\},\quad \Delta=\frac{16\pi}{15}\frac{\overline{V}^2}{\nu}. \tag{19}$$

The number of spins directed against the magnetization at zero temperature is

$$n_\downarrow=n_i\int_{-\infty}^{0}W(\omega)\,d\omega. \tag{20}$$

Noting that the principal role is played in this integral by $|\omega| \sim SV_0$, we get

$$\frac{n_\downarrow}{n_i}=\frac{0.435}{\nu^{1/2}}e^{-0,845\nu}. \tag{21}$$

This quantity is small even at values of ν not greatly exceeding unity.

The heat capacity per impurity is

$$C_M=\int_{-\infty}^{\infty}d\omega\,W(\omega)c_M(\omega), \tag{22}$$

where

$$c_M(\omega)=\beta\omega^2\left\{\frac{1}{4}\operatorname{sh}^{-2}\frac{\beta\omega}{2}-\left(S+\frac{1}{2}\right)^2\operatorname{sh}^{-2}\left(S+\frac{1}{2}\right)\beta\omega\right\}.$$

At low temperatures $T \ll SV_0$, the main contribution to (22) is made by frequencies $\omega \sim T$, so that according to (18) we can replace $W(\omega)$ by $W(0)$, and

$$C_M=\frac{4\pi^2S}{3(2S+1)}W(0)T. \tag{23}$$

The ratio of this heat capacity to the heat capacity C_S due to the spin waves is of the order of

$$\frac{C_M}{C_s}\approx\sqrt{\frac{SV_0}{T}}\nu^2e^{-0.845\nu}. \tag{24}$$

At sufficiently low temperatures this quantity can exceed unity.

In an external magnetic field H we have in place of (22)

$$C_M(H)=\int_{-\infty}^{\infty}W(\omega-\mu H)c_M(\omega)\,d\omega, \tag{25}$$

where μ is the effective magneton.

At temperatures $T < SV_0$ and in fields $\mu H < \nu^{2/3}SV_0$ the heat capacity is

$$C_M(H)=C_M(0)\exp\left(-\frac{3\mu H}{2SV_0}-\frac{1.35}{\nu}\left(\frac{\mu H}{SV_0}\right)^2\right). \tag{26}$$

The heat capacity depends little on the magnetic fields up to fields of the order of $SV_0/\mu > T\mu^{-1}$. At the same time, the spin-wave heat capacity in fields $\mu H > T$ decreases with increasing field like $\exp(-\mu H/T)$. Therefore in a sufficiently strong magnetic field the entire spin heat capacitance is determined by the local spin flips. In fields $\mu H > SV_0$ the heat capacity decreases exponentially with increasing field.

Since part of the spins at $T = 0$ is directed against the magnetization, the magnetic susceptibility χ differs from zero.

It is obvious that the susceptibility per impurity is

$$\chi=-2S\mu\lim\frac{dn_\downarrow(H)}{dH}\bigg|_{H\to 0}.$$

Using (18) and (20) we find that

$$\chi=2S\mu^2W(0)\sim\nu^{-1/2}e^{-0.845\nu}. \tag{27}$$

As seen from (23) and (27), the ratio of the susceptibility per impurity to the heat capacity C_M does not depend on the impurity concentration. A temperature and a field dependence analogous to those of the heat capacity are possessed at $T < SV_0$ by all quantities that are even functions of ω prior to averaging over ω. For example, for the resistivity due to scattering of S electrons by localized spins we have

$$\frac{\Delta\rho}{\rho_0}=\frac{I^2}{U^2}\left\{-3S^2+TW(-\mu H)\int_{-\infty}^{\infty}dx\left[4(S^2-B_S^2(x))\right.\right.$$
$$\left.\left.+x\operatorname{cth}\frac{x}{2}\left(\left(S+\frac{1}{2}\right)^2\operatorname{sh}^{-2}\left(S+\frac{1}{2}\right)x-\frac{1}{4}\operatorname{sh}^{-2}\frac{x}{2}\right)\right]\right\}, \tag{28}$$

where I is the s-d exchange interaction constant, U is the energy of the Coulomb interaction, $\Delta\rho = \rho_0$, and $B_S(x)$ is the Brillouin function.

Formula (28) is obtained by averaging over ω the expression for the resistivity due to electron scattering by the impurities in an external field.[8] We see that at low T the temperature-dependent part of the resistivity is proportional to T, whereas the spin-wave scattering makes a contribution proportional to $T^{3/2}$.[7,9] With increasing H, the resistance due to local spin flips, just as the heat capacity, decreases more slowly than the spin-wave part. Therefore in sufficiently strong fields the temperature-dependent part of the resistance, connected with the spin scattering of the electrons, is due to local spin flips.

5. We consider in this section the properties of an alloy whose Fermi surface contains, besides parts with large characteristic dimensions ($k_{01} \sim \pi/a$, where a is the lattice constant), also sections with dimensions k_{02} that are small in comparison with π/a. If $k_{02}n^{-1/3} < 1$, then the small sections make a ferromagnetic contribution $V_f(r)$ the indirect exchange, which can turn out to be much stronger than the oscillating indirect interaction $V_{osc}(r)$ due to the large sections of the Fermi surface.

Let the Fermi surface comprise an assembly of a large sphere-like region and several small sections (stubs, necks, pockets, etc.) Since $V_f(r)$ does not oscillate over distances $r \lesssim k_{02}^{-1}$, and since $k_{02}^{-1} > r_c$, it follows that, as in the preceding section, the interaction energy of each spin with all the remaining spins coincides with the average energy

$$\bar{V}_f(r) \sim n_i I^2 \chi(0) \sim n_i I^2 \rho_2(\varepsilon_F),$$

where $\chi(q)$ and $\rho_2(\epsilon_F)$ are respectively the susceptibility and the summary density of states of the s electrons in the small sections. On the other hand, $V_{osc}(r)$ oscillates strongly over distances on the order of r_c, and therefore the interaction energy of each spin with the remaining ones is of the order of $V_{osc}(r_c)$, so that for a sphere it is of the order of $I^2\rho_1(\epsilon_F)n_i$, where $\rho_1(\epsilon_F$ is the density of states of the sphere.

Thus, the condition under which ferromagnetic ordering is produced in the alloy at $T = 0$ reduces to the inequality $\rho_2(\epsilon_F) > \rho_1(\epsilon_F)$. A similar inequality is obtained also when the large section is a cylinder.

The role of the fluctuations of the ferromagnetic interaction was considered above. It was shown that the linearly-temperature-dependent parts of the thermodynamic quantities decrease exponentially with increasing impurity concentration. In the situation considered now, the oscillating potential, as we shall show, makes a per-impurity contribution inversely proportional to n_i to the thermodynamic quantities. Therefore the fluctuations of the ferromagnetic potential can be neglected.

The problem reduces thus to an investigation of the properties of a system of spins situated in the molecular field $H_f = V_f\mu^{-1}$ produced by the ferromagnetic potential and interacting via the oscillating potential

$$V_{osc}(r) = A\frac{\cos 2k_F r}{(k_F r)^3}. \tag{29}$$

For impurities located at the average distance we have $\mu H_f \gg V_{osc}(r)$, so that the oscillating potential is insignificant. Their contribution to the thermodynamic quantities decreases exponentially with increasing T. The principal role is now played by impurity pairs that have come close together to such an extent that the energy of the antiferromagnetic interaction between them differs from μH_f by a value on the order of T.

To calculate the free energy of the pairs we can use the Larkin and Khmel'nitskiĭ method of virial expansions.[10] For the heat capacity per impurity we have

$$C_M = \frac{4\pi^2}{9}S(S+1)\frac{n_i AT}{k_F^3\mu(H_F+H)^2} \tag{30}$$

where H is the magnetic field. At $H = 0$ the heat capacity is $C_M \sim Tn_i^{-1}$. The specific heat is independent of the impurity concentration.

The longitudinal susceptibility is

$$\chi = \frac{4}{3}S(2S+1)\frac{An_i}{k_F^3(H_F+H)^2}. \tag{31}$$

At $H = 0$ we have $\chi \sim n_i^{-1}$. The ratio

$$C_M/\chi T = \pi^2/3\mu^2 \tag{32}$$

depends only on the effective magneton, i.e., on the properties of the impurity and the alloy.

It is easy to show that the temperature-dependent part of the resistivity, just as the heat capacity, is proportional to $n_i T(H_f + H)^{-2}$. We emphasize that both the thermodynamic quantities and the resistivity depend little on the external magnetic fields up to fields $H \approx H_f \approx T_c/\mu$.

APPENDIX

The potential of the indirect interaction of the impurities is given by

$$V(\mathbf{r}) = I^2\sum_{\mathbf{q},\mathbf{k}}\frac{f_\mathbf{k}-f_{\mathbf{k}+\mathbf{q}}}{\varepsilon_{\mathbf{k}+\mathbf{q}}-\varepsilon_\mathbf{k}}e^{i\mathbf{q}\mathbf{r}}, \tag{A.1}$$

where f_k is the Fermi distribution function. We calculate the contribution made to $V(r)$ by a Fermi-surface section in the form of a cylinder of base radius k and height p. We have

$$V(r) = \frac{I^2\Omega^2}{8\pi^6}\frac{\sin^2 pz}{z^2}\int d^2\mathbf{q}_\perp\, d^2\mathbf{k}_\perp \frac{f_{q_\perp}}{\varepsilon_{k_\perp}-\varepsilon_{q_\perp}}\exp i(\mathbf{q}_\perp-\mathbf{k}_\perp, \mathbf{r}_\perp), \tag{A.2}$$

where Ω is the volume of the unit cell, and $\mathbf{k}_\perp$, $\mathbf{q}_\perp$, and $\mathbf{r}_\perp$ are vectors in the plane perpendicular to the cylinder axis (the z axis),

$$\varepsilon_{k_\perp} = k_\perp^2/2m.$$

After integration we get

$$V(\mathbf{r}) = \frac{I^2\Omega^2 mk^2}{\pi^4}\frac{\sin^2 pz}{z^2}\Phi(kr_\perp), \tag{A.3}$$

where

$$\Phi(x) = -\frac{\pi}{4}[J_0(x)N_0(x)+J_1(x)N_1(x)],$$

$J_l(x)$ and $N_l(x)$ are cylindrical functions. At $x \ll 1$ we have with logarithmic accuracy

$$\Phi(x) = \frac{1}{2}\ln\frac{1}{2x},$$

and at $x \gg 1$

$$\Phi(x) = \frac{\sin 2x}{(2x)^2}.$$

This asymptotic form was obtained in[1].

[1)] An expression for V(r) in the case of a cylinder is given in the Appendix.

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