RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 33, NUMBER 1 1 JANUARY 1986

Long-range interactions between local moments in metals: Application to spin-glass systems

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(Received 11 October 1985)

We have calculated the pair interaction in *Cu*Mn by using the Friedel-Anderson model of local moments in metals. Over distances $a < R < 15a$ the decay of the interaction is considerably slower than $1/R^3$. With this coupling we are able to account for the magnitude of the spin interaction constant V_0 found for *Cu*Mn alloys in the ppm range, as well as the dependence on concentration of the glass temperature of more concentrated alloys after this temperature has been corrected for mean-free-path effects.

The sublinear dependence on concentration of magnetic impurities of the spin-glass temperature of metallic alloys has been heretofore explained by conduction electron mean-free-path effects.[1] This implies that the range of interaction between impurities decays faster than $1/R^3$. Here we show that, on the contrary, this dependence is an indication that the decay of the interaction is much *slower* than $1/R^3$. The long-range interaction we develop gives the correct spin interaction constant V_0 found in dilute *Cu*Mn alloys. In addition, it reproduces the dependence on concentration and gives reasonable estimates of the spin-glass temperatures (T_g) of more concentrated *Cu*Mn alloys.

The spin-glass temperature is a measure of the collective behavior of a set of randomly distributed spins. In the mean-field approximation, and for $S >> \frac{1}{2}$, it is defined as

$$k_B T_g = \tfrac{1}{3} S(S+1) \left\{ \sum_{\substack{\text{OCCUPIED SITES} \\ (j \neq i)}} J_{ij}^2 \right\}^{1/2} , \tag{1}$$

where the sum is over occupied sites. In metallic alloys where the magnetic interactions are fairly long range we approximate the sum in Eq. (1) by an integral[2]

$$\sum_{\substack{\text{OCCUPIED SITES} \\ (j \neq i)}} J_{ij}{}^2 \simeq c\rho \int_{R_0}^{\infty} (c) J^2(R)\, d^3R , \tag{2}$$

where c is the concentration of magnetic impurities, $\rho = N/V$ is the density of sites in the lattice, and the lower limit of integration is taken as the radius of the sphere corresponding to the volume of the excluded impurity i. For the fcc lattice $(4\pi/3)R_0^3 = (a^3/4)(1/c)$, i.e., $R_0(c) = \sqrt[3]{3a^3/16\pi}\, c^{-1/3} = 0.39ac^{-1/3}$ where a is the lattice constant. For the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, $J(R) \sim \cos(2k_F R)/R^3$, and after averaging over the oscillations, one finds that T_g is directly proportional to the concentration c. Intermediate scattering of the conduction electrons, i.e., finite mean free path (MFP), leads to an exponential damping of the RKKY interaction;[3] the damping constant is proportional to the concentration of impurities (nonmagnetic as well as magnetic). For a fixed concentration c this reduces T_g; as a function of c this yields a sublinear dependence of T_g on c, i.e., $T_g \sim c^{\alpha}$ where $\alpha < 1$.

Recently Vier and Schultz have shown that mean-free-path effects are responsible in part for the sublinear dependence of T_g on the concentration.[4] However, their results also show that these effects do not fully account for their data for *Cu*Mn. Indeed, it was previously noted that the scattering cross section needed to explain concentration dependence of T_g for *Cu*Mn was about *six times larger* than that necessary to explain resistivity data.[1] We have calculated the interaction $J(R)$ by using the Friedel-Anderson model of local moments in metals and the results of a self-consistent cluster calculation for the d-state resonance positions and widths of manganese in copper.[5] We find $J(R)$ has large preasymptotic corrections which make it more slowly varying than $1/R^3$ at intermediate distances $R \lesssim 15a$. We will show that at least in *Cu*Mn it is these corrections rather than finite MFP effects that are the primary cause for the deviations of $T_g(c)$ from linearity.

The coupling between local moments in metals takes place through the conduction electrons except for neighboring spins where one has direct and superexchange. At large distances the phase of the wave function oscillates so rapidly that only the spin scattering *at* the Fermi level contributes to the coupling, and the pair interaction is proportional to $1/R^3$ modulo a slow exponential decay when one takes into account MFP effects. At moderate distances, spin scattering from states in the energy range $\delta E \sim E_F/k_F R$ contribute to the coupling. In order to calculate the coupling it is necessary to know the energy dependence of the spin scattering. For the conventional RKKY interaction the assumption is made that conduction electrons are scattered by a delta function potential. The matrix elements of this scattering in k space (energy) are all equal, i.e., independent of energy.

For transition-metal ions in copper, for example, the archetypical spin-glass alloy *Cu*Mn, the spin scattering of the conduction electrons by the local moments comes from s-d mixing.[6] This d-wave scattering depends on the energies of the states involved, and the coupling between two moments deviates from the RKKY form for all but the largest distances, i.e., there are preasymptotic corrections to RKKY due to the *angular* and *energy* dependence of the spin scattering.[7] In the Hartree-Fock approximation, the interaction between two local moments in a metal is written as[7]

$$E^{\sigma\sigma'}(R) = -\frac{1}{\pi} \mathrm{Im} \sum_m \int_{E_F}^{\infty} G_1^{0\sigma}(\epsilon) F_m(\epsilon, R) G_2^{0\sigma'}(\epsilon)\, d\epsilon , \tag{3}$$

where

$$G_i^{0\sigma} = \frac{1}{\epsilon - (E_d^{\sigma} - i\Delta)} \tag{4}$$

is the Green's function for the local d state at one site (i), and

$$\sum_m F_m(\epsilon, R) = \frac{(2l+1)^2}{64\pi^4} \frac{V^4(k)}{R^2} e^{2ikR} Q \left(\frac{1}{ikR} \right| \tag{5}$$

represents the coupling between d electrons through the electron gas. The s-d mixing is represented by $V(k)[k=\sqrt{2m\epsilon/\hbar^2}]$, and Q is a polynomial given by Price[7] for d-wave scattering ($l=2$). For s-wave scattering $l=0$ and $Q=1$. To account for the finite lifetime of the conduction electron states, we add an imaginary part to the energy in the Green's function for the conduction electrons entering Eq. (3), i.e., $\tilde{G}_k=(\epsilon+i\lambda-\epsilon_k)^{-1}$, where $\lambda/\hbar$ is the inverse of the lifetime of the state k.[3] This has the effect of displacing the line of integration by λ above the real axis in the complex energy plane, and replacing Δ by $\Delta-\lambda$ in Eq. (4). To relate the Hartree-Fock energy to an effective coupling we use

$$J(R)=\frac{E^{\uparrow\downarrow}(R)-E^{\uparrow\uparrow}(R)}{2S^2} \quad . \tag{6}$$

To illustrate our point, we were guided by the results of Ref. 5 for the d resonances of manganese in copper. To simplify our calculation we took $Z_d^+ + Z_d^- = 6$ and $Z_d^+ - Z_d^- = 2S = 4$. By using the Friedel sum rule we find $\eta^+(k_F)=\pi$ and $\eta^-(k_F)=\pi/5$, and we can confine our attention to the one resonant level ($\sigma=\sigma'$), i.e., the open d resonance above the Fermi level. If we had used $Z_d^+ = Z_d^- = 5$, we would have to consider the contribution to Eq. (3) from both levels, i.e., $\sigma=\pm$, and our interaction would not have changed very much, e.g., the asymptotic value of Eq. (3) changes by less than 10%. We have numerically evaluated the coupling Eq. (3) by using $E_d-E_F=0.7$ eV, $\Delta=0.51$ eV, and $E_F=7$ eV for manganese in copper.[5] As we have set $\eta^-=\pi/5$ we *fixed* Δ by using the relation $\tan\eta^-=\Delta/(E_d-E_F)$ and the result of Ref. 5 for E_d-E_F. The expression for the interaction Eq. (3) valid for all values of $k_F R$ is unwieldy; see Price, Ref. 7. While we used it to evaluate the spin-glass temperature it is not reproduced here.

Mean-free-path effects depend on concentration; for the range of concentration we consider they can be included in the range function by writing

$$J_\lambda(R)=e^{-(\lambda/2E_F)k_F R}J_0(R) \quad . \tag{7}$$

In any case these effects have been removed from the data[4] so that we can set $\lambda=0$ and compare our results to the $T_g(\rho=0)$ given by Vier and Schultz.[4] To arrive at the spin-glass temperature we average over the oscillatory part of the coupling $J(R)$; therefore we plot in Fig. 1 the *amplitude* of the coupling $A(R)$, and we show the RKKY interaction for comparison. As we have multiplied our coupling by R^2, a pure $1/R^2$ interaction would show up as a horizontal line. In Fig. 1 we focus on the region $R>2a$ because there are relatively few spins in the region $R<2a$ for concentrations less than 10%; in fact, for $c<0.05$ (5%) the average between neighboring spins, is greater than $2a$. In the region $R<2a$ $J(R)$ $[A(R)]$ goes to zero as $R\rightarrow 0$ because we have considered d-wave scattering. The conventional RKKY interaction is based on s-wave scattering and has $J(R)\rightarrow\infty$ as $R\rightarrow 0$.

In the range $R/a>2$, we are able to fit the envelope of our coupling Eq. (3) to the form

$$A(R)=\frac{1}{R^2}\left[Ae^{-\beta x}+\frac{B}{x}[1-e^{-\gamma x}]\right] \quad , \tag{8}$$

where $x=R/a$, $A=-2.2\times10^{-3}(\mathrm{eV}\ \mathrm{\AA}^2)$, $B=0.27(\mathrm{eV}\ \mathrm{\AA}^2)$, $\beta=2.6\times10^{-3}$, and $\gamma=0.23$.

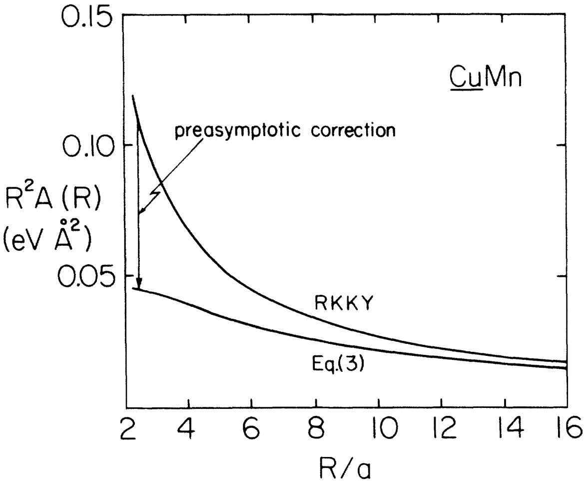

FIG. 1. Envelope function of pair interaction between two manganese impurities in copper, as calculated by Eq. (3), with $\lambda=0$, and the standard RKKY interaction $A(R)\sim R^{-3}$. The differences are preasymptotic corrections to the RKKY interaction.

For large R our coupling agrees with the asymptotic form

$$J(R)=V_0\cos(2k_F R+2\eta^-)/R^3 \quad , \tag{9}$$

where

$$V_0=\frac{25}{2S^2\pi}\frac{E_F}{k_F^3}\sin^2\eta^- \quad .$$

For the parameters quoted above and by using $S=2$ we find $V_0=15.6\times10^{-37}$ ergs cm^3. This value is very close to the one found from low-field magnetization measurements on dilute alloys,[8] $V_0=14.2\times10^{-37}$ ergs cm^3. When we attempt to fit the envelope function Eq. (8) in the range $1<R/a<15$ with the form V_0'/R^3, we find $V_0'=7.0\times10^{-37}$ ergs cm^3. This value is very close to that found when high-field magnetization studies of dilute alloys are analyzed using this (incorrect) form,[9] $V_0'=7.50\times10^{-37}$ ergs cm^3.

The deviations of the actual coupling from the RKKY form, see Fig. 1, are known as preasymptotic corrections.[7] They are governed by the distance $|k_d-k_F|^{-1}$ where $k_d=\sqrt{2\pi[E_d-i\Delta]/\hbar^2}$; the spatial extent of the corrections increase as the center of the d-state resonance approaches the Fermi level and the narrower the resonance is. If one halves $|k_d-k_F|$ the range over which preasymptotic corrections are noticeable is approximately doubled.

We have used the interaction Eq. (3) to calculate the spin-glass temperature as a function of concentration, see Eqs. (1) and (2), for *Cu*Mn. As Eq. (1) is a mean-field estimate of T_g we must normalize our results before comparing them to data. By reducing our mean-field estimates of T_g by a factor of 2.1 we are able to fit the recent data on *Cu*Mn,[4] see Fig. 2. In particular, the preasymptotic corrections are able to explain the sublinear concentration dependence of T_g after MFP effects are removed, i.e., $T_g(\rho=0)$.[4] Furthermore, by introducing $\lambda=1.6cE_F$ in Eq. (7) we are able to fit the data on $T_g(\rho)$[4] which contains the self-damping by the magnetic impurities. The magnitude of the reduction used to fit our mean-field estimates to the data, i.e., 2.1, seems reasonable; however, there is no guidance from theory on this reduction at the present time.[10]

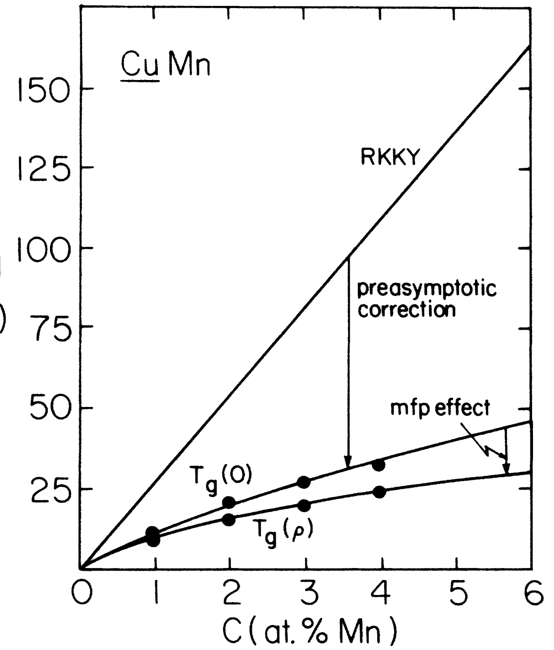

FIG. 2. The spin-glass temperature of *Cu*Mn alloys as a function of manganese concentration. Experimental points are taken from Ref. 4. Calculated curves for T_g were normalized as discussed in the text, $T_g(0)$ was calculated using Eq. (3) using $\lambda = 0$, and $T_g(\rho)$ using $\lambda = 1.6cE_F$.

Of the two origins of the deviations of $T_g(c)$ from linearity, we see from Fig. 2 that preasymptotic corrections are seen to be far more important than MFP effects. If we tried to explain $T_g(\rho)$ with *only* MFP effects, say for $c = 6\%$, we would need $\lambda = 8.8cE_F$, which is 5.5 times larger than that found from Ref. 4. This explains why in previous attempts to fit $T_g(c)$ it was necessary to use these unusually large scattering cross sections.[1] Finally, in addition to explaining the slope of T_g versus concentration for *Cu*Mn in the range $c = 0.001$ to 0.1 (0.1 to 10 at. %), we find in the ppm range $T_g \sim c^{0.94}$. While there is no transition in this range of concentration, this nonlinear behavior is very close to that observed for the spin interaction temperature which characterizes collective behavior in the ppm range,[8] $T_i \sim c^{0.89}$. This is noteworthy in that the MFP of the conduction electrons has a negligible effect on T_i in this range of concentration.

While we concentrated on *Cu*Mn the above considerations apply to other metallic alloys, e.g., *Au*Mn and *Ag*Mn.[11] One can determine the magnitude and range of the pair interactions once the single-ion parameters k_d, k_F, Δ, and E_F are reliably known. This analysis is applicable also to systems as *Au*Fe in as much as the local moment is due to spin alone, as Eq. (3) neglects orbital contributions to the moments.[12]

In summary, we have shown that the decay of the interaction between local moments in metals as *Cu*Mn is considerably slower than $1/R^3$. We have seen that this interaction is able to account for the magnitude of the spin interaction constant V_0 and it gives reasonable estimates of the spin-glass temperature of *Cu*Mn alloys over a wide range of concentrations. As we have not taken into account short-range interactions in estimating T_g, our results suggest that short-range order does not have an appreciable effect on T_g, rather the spin-glass temperature is an indication of long-range *collective* behavior. Furthermore, correlations above T_g start to grow under a potential given by Eq. (8); only when $R > 10a$ does the system realize the asymptotic form R^{-3}. For a sample with $c = 0.05$ (5%) as many as 160 spins are coupled through the more slowly varying preasymptotic portion of the interaction.

We would like to thank A. J. Bray and M. A. Moore for their extensive discussions and correspondence with us, and H. Alloul, I. A. Campbell, A. Fert, J. Friedel, S. M. Goldberg, P. Monod, and J. A. Mydosh for helpful discussions. We gratefully acknowledge the hospitality of the Laboratoire de Physique des Solides at the Université de Paris-Sud during our stay in France. This work was supported in part by the National Science Foundation under Grant No. DMR 81-20673 and by the United States–France program of scientific cooperation through National Science Foundation Grant No. INT 82-12503.

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[2] To evaluate the sum in Eq. (1) one should proceed as one does for dipolar sums, i.e., by doing a discrete sum over neighboring sites where a continuum approximation would be inappropriate, and calculating the remainder by an integral. For the discrete sum over the near neighbors, contributions from direct and superexchange couplings should be added to the indirect coupling through the conduction electrons; this introduces additional parameters. The lower limit of integration is large enough so that the main contribution to T_g comes from regions where the discreteness of the lattice and the additional couplings do not play an important role. Therefore, we neglect these effects and approximate the sum as shown in Eq. (2).

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