LONG-RANGED MAGNETIC POLARIZATION EFFECTS

Daniel Mattis
IBM Watson Research Center, Yorktown Heights, New York
(Received 15 April 1964)

Two years ago a group[1] at Bell Telephone Laboratories discovered an exceedingly long-ranged magnetic interaction among rare earth (RE) atoms dissolved in palladium. It is the purpose of the present note to suggest a theory for this phenomenon, which, it will be recalled, could not be explained by the Ruderman-Kittel interaction

$$J_{ij}^{\text{R-K}} = A(\sin x - x\cos x)/x^4, \quad x = 2k_F R_{ij}, \tag{1}$$

nor even by the longer ranged Yosida modification,

$$J_{ij}^{\text{Y}} = A'(\sin x - x\cos x)/x^3, \tag{2}$$

without assuming an unreasonably small value of k_F. In fact, the experimenters had to adopt an ad hoc interaction[1]

$$J_{ij}^{\text{BTL}} = A'', \quad R_{ij} < r_0,$$
$$= 0, \quad R_{ij} > r_0, \tag{3}$$

such that any two RE atoms separated up to a distance r_0 (~15 Å) underwent an interaction A''. No physical explanation was proposed for this novel interaction, nor do we know of any published to date.

We propose the possibility that the physical mechanism is none other than the usual indirect exchange via conduction electrons, but modified to include exchange matrix elements which change the total angular momentum of the f shell by $\pm\hbar$, and suggest Eq. (10) to replace Eq. (3). It has already been pointed out by Brout and Suhl[2] that such processes add a gap (in excess of 0.1 eV) to the Ruderman-Kittel energy denominators. Consider the complete indirect exchange Hamiltonian

$$H = -\sum J(R_{ij})\vec{J}_i\cdot\vec{J}_j, \tag{4}$$

where

$$J(R_{ij}) = B\sum f_k(1-f_{k'})\exp[i(\vec{k}-\vec{k}')\cdot\vec{R}_{ij}]$$
$$\times\Big((g_i-1)(g_j-1)(E_{k'}-E_k)^{-1} + \exp(i\pi\eta_j)$$
$$\times\{(g_i-1)+\exp(i\pi\eta_i)[S_i(S_i+1)-J_i(J_i+1)(g_i-1)^2]^{1/2}\}$$
$$\times[S_j(S_j+1)-J_j(J_j+1)(g_j-1)^2]^{1/2}(E_{k'}+\Delta-E_k)^{-1}\Big). \tag{5}$$

The quantity $(g_i - 1)$ is like a "charge": negative for the rare earths to the left of Gd in the periodic table, and positive for those to the right. The sign factor $\eta_i = 0$ or 1 depends on the relative phases of the ground state (J_i) and the excited state $(J_i \pm 1)$. Both terms in Eq. (5) contribute to the Ruderman-Kittel interaction, although the contributions may add or cancel depending on the relative sign. In addition, the second term has a monotonic long-ranged part that we isolate by means of the identity,

$$(E_{k'}+\Delta-E_k)^{-1} = (E_{k'}-E_k)^{-1}$$
$$-\Delta(E_{k'}-E_k)^{-1}(E_{k'}+\Delta-E_k)^{-1}, \tag{6}$$

The first term being of the familiar R-K form, we are led to examine the novel contribution:

$$-B'\Delta\sum\frac{f_k(1-f_{k'})\exp[i(\vec{k}-\vec{k}')\cdot\vec{R}_{ij}]}{(E_{k'}-E_k)(E_{k'}+\Delta-E_k)}\equiv I_{ij}, \tag{7}$$

in which

$$B'=B\{(g_i-1)+\exp(i\pi\eta_i)[S_i(S_i+1)-J_i(J_i+1)(g_i-1)^2]^{1/2}\}[S_j(S_j+1)-J_j(J_j+1)(g_j-1)^2]^{1/2}\exp(i\pi\eta_j). \tag{7'}$$

In the quasifree electron approximation, $E_k=\hbar^2k^2/2m$, the Fermi energy is $E_\mathrm{F}=\hbar^2k_\mathrm{F}^2/2m$, and

$$I_{ij}=-B'\Delta\exp(i\pi\eta_i)\frac{(4\pi)^2a^6}{(2\pi)^6}\int_0^{k_\mathrm{F}}dk\,k^2\int_{k_\mathrm{F}}^{\infty}dk'\,k'^2\frac{\sin kR_{ij}\sin k'R_{ij}}{(kR_{ij})(k'R_{ij})}\left[\frac{\hbar^4}{(2m)^2}(k'^2-k^2)\left(k'^2-k^2+\frac{2m\Delta}{\hbar^2}\right)\right]^{-1}, \tag{8}$$

where a^3 = volume of unit cell. The rapid convergence of the integrand when $\Delta\ll E_\mathrm{F}$ permits the substitution $\sin kR_{ij}\sim\sin k'R_{ij}\sim\sin k_\mathrm{F}R_{ij}$. Thus,

$$I_{ij}\cong-B'\Delta\exp(i\pi\eta_j)\frac{(4\pi)^2a^6}{(2\pi)^6}\left(\frac{2m}{\hbar^2}\right)^2\frac{(1-\cos2k_\mathrm{F}R_{ij})}{8R_{ij}^2E_\mathrm{F}^2}\int_0^1dx\int_1^\infty dy\left[(y-x)\left(y-x+\frac{\Delta}{E_\mathrm{F}}\right)\right]^{-1}$$

$$\cong-B'\Delta\exp(i\pi\eta_j)\frac{(4\pi)^2a^6}{(2\pi)^6}\left(\frac{2m}{\hbar^2}\right)^2\frac{(1-\cos2k_\mathrm{F}R_{ij})}{8R_{ij}^2E_\mathrm{F}^2}\left\{\frac{E_\mathrm{F}}{\Delta}\ln\left(1+\frac{\Delta}{E_\mathrm{F}}\right)+\ln\frac{E_\mathrm{F}+\Delta}{\Delta}\right\}. \tag{9}$$

Many of these factors already occur in the constant A of Eq. (1), and an order-of-magnitude approximation is

$$I_{ij}=-A\left(\frac{a}{R_{ij}}\right)^2\exp(i\pi\eta_i)\left[\ln\left(1+\frac{\Delta}{E_\mathrm{F}}\right)+\frac{\Delta}{E_\mathrm{F}}\ln\frac{E_\mathrm{F}+\Delta}{\Delta}\right], \tag{10}$$

the $(\cos2k_\mathrm{F}R_{ij})$ being in the nature of a correction to the oscillatory R-K interaction. As noted above, the derivation of this long-ranged nonoscillatory interaction is not valid if $\Delta>E_\mathrm{F}$; and in the limit $\Delta\gg E_\mathrm{F}$, the two terms in Eq. (6) actually cancel. On the other hand, if $\Delta\ll E_\mathrm{F}$, the magnitude of I_{ij} will vanish.

It may be that the d holes in palladium have just the proper Fermi energy and the RE's in that metal have just the proper excitation spectra, such that $\Delta/E_\mathrm{F}=O(\frac{1}{2})$. In that case the rapidly fluctuating R-K interaction may, in fact, average to zero, in comparison to a sizable systematic, long-ranged interaction I_{ij}.

It should be noted that if the natural choice $\eta_j=0$ is made, the long-range interaction between like atoms is antiferromagnetic, whereas the long-range interaction between atoms of opposite sign for the quantity $g-1$ is ferromagnetic. I am indebted to Dr. Shaltiel for telling me about unpublished experimental results in accordance with the above.

[1]M. Peter, D. Shaltiel, J. H. Wernick, H. J. Williams, J. B. Mock, and R. C. Sherwood, Phys. Rev. Letters 9, 50 (1962).

[2]R. Brout and H. Suhl, Phys. Rev. Letters 2, 387 (1959).