JOURNAL OF OPTOELECTRONICS AND ADVANCED MATERIALS Vol. 9, No. 4, April 2007, p. 911 - 915

Magnetic transitions in metallic compounds determined by RKKY interactions

E. BIRSAN*, C. CANDIN[a], M. RACUCIU
Physics Department, University Lucian Blaga, str. Dr I.Ratiu, No.5-7, 550024, Sibiu, Romania
[a]*Faculty of Engineering "Hermann Oberth", University Lucian Blaga of Sibiu, Romania*

We study the role of RKKY (Ruderman-Kittel-Kasuya-Yosida) interactions in magnetic transitions observed in various metallic compounds. These transitions were found to play an important role in problems involving the interaction of localized moments in a metal, via polarization of conduction electrons. We show the existence of the antiferromagnetism-ferromagnetism transition in metallic compounds generated by RKKY interactions. We study the role of interstitials impurities in this process and we provide the influence of exchange coupling constant. Indirect exchange couples moments over relatively large distances. It is the dominant exchange interaction in metals where there is little or no direct overlap between neighboring magnetic electrons. It therefore acts through an intermediary, which in metals are the conduction electrons (itinerant electrons). The RKKY exchange coefficient oscillates from positive to negative as the separation of the ions changes and has the damped oscillatory nature. Therefore, depending upon the separation between a pair of ions their magnetic coupling can be ferromagnetic or antiferromagnetic. A magnetic ion induces a spin polarization in the conduction electrons. This spin polarization in the itinerant electron system is felt by the moments of other magnetic ions within range, leading to an indirect coupling.

(Received November 2, 2006; accepted February 28, 2007)

Keywords: Itinerant ferromagnetism, RKKY interactions, Magnetic transitions

1. Introduction

The RKKY interaction was found to play an important role in various problems involving the interaction of localized moments in a metal via polarization of conduction electrons [1], [2].

It was demonstrated that at large distances r the interaction decays as $1/r^3$ and has the $2k_F$ (k_F been the Fermi wave-vector) oscillation with the Fermi momentum k_F that can be the premise of the magnetic transition band-ferromagnetism (FM) $\rightarrow$ band-antiferromagnetism (AFM) and reverse. At intermediate temperatures, when the width of the filled part of the band is comparable to $k_B T$ (k_B is the Boltzmann constant), the amplitude of the oscillation is diminished. At high temperature, for the Boltzmann gas, the magnetization has a Gaussian shape, where the reciprocal wave number of the electron with energy $k_B T$ i.e. $\hbar/\sqrt{2mk_B T}$, is the decay length [3]. Thus we take in account the Fermi–Dirac distribution law [4]:

$$f_{FD}(\varepsilon) = \frac{1}{\exp\left(\frac{\varepsilon - \mu}{k_B T}\right) + 1} \tag{1}$$

At high temperatures, there are many unoccupied states witch excited electrons can occupy. Thus, the Pauli Exclusion Principle and Fermi – Dirac statistics are useful in describing low temperature behavior (different from $T = 0K$). As the temperature increases, only electrons with $k_B T$ energy closed to ε_F will be thermally excited [5]. The total energy of the system in this case is:

$$U_{total} = \int_0^\infty \varepsilon N(\varepsilon) f_{FD}(\varepsilon) d\varepsilon \tag{2}$$

RKKY theory explains that a magnetic ion is able to spin polarize surrounding conduction electrons with λ_F [6]. In turn, these spin polarized electrons can couple to the spin of a nearby ion, thus creating a cooperative interaction between distant magnetic ions. The oscillatory nature of the polarization at large distance is of the form:

$$f(a) = \frac{\cos(2k_F a)}{a^3} \tag{3}$$

where "a" is the distance from the local moment, thus, there are regions in which the spins are polarized successively in the up and down configurations with respect to the magnetic ion. Whether ferromagnetic or antiferromagnetic behavior is favored is dependent on the distance between the conduction electron and the magnetic ion.

2. The physical model

We propose a model that include the RKKY interaction, the Hubbard model and the double exchange interaction, in fact a hybrid model between the itinerant and locals moments magnetic behavior. We study this model in a numerical way (Monte Carlo simulation) and we find the influence of the RKKY interaction in the stability of the condensed state.

2.1 The Hubbard model

The Hubbard model is the simplest band model that proposes an electronic correlated approach and the Hamiltonian interaction is given by [7]:

$$H_{Hubbard} = -\sum_{i,j,\sigma} t_{ij} c_{i\sigma}^{+} c_{j\sigma} + U \sum_{i} n_{i\uparrow} n_{i\downarrow} \tag{4}$$

where, t_{ij} is a general hopping matrix element between sites i and j. in this Hamiltonian $c_{i\sigma}^{+}, c_{j\sigma}$ are the creation and annihilation operators, respectively and U is the Coulomb interaction. Also, $n_{i\uparrow}, n_{i\downarrow}$ are the number particle operators with spin up and spin down. Thus, the Hubbard interaction is interplay between the kinetic and the Coulomb interaction terms.

2.2. The double exchange (DE) model

The double exchange model is described by the following Hamiltonian [8]:

$$H_{\mathrm{DE}} = H_{\mathrm{hopping}} + H_{\mathrm{interaction}} \tag{5}$$

where:

$$H_{\mathrm{hopping}} = \sum_{i,j} \left(t_{ij} - \mu \delta_{ij} \right) c^{+}(i) c(j) \tag{6}$$

$$H_{\mathrm{interaction}} = -J_{sd} \sum_{i} \vec{s}(i) \vec{S}(i) \tag{7}$$

$c(i)$ and $c^{+}(i)$ are the annihilation and creation operators for electrons at site $\vec{R}_i$ in the spinor notation $c^{+}(i) = \left(c_{\uparrow}^{+}(i), c_{\downarrow}^{+}(i) \right)$, μ is the chemical potential, t_{ij} is the hopping integral, $\vec{s}(i)$ is the spin density operator of the conduction – electrons and is given by:

$$\vec{s}(i) = \frac{1}{2} c^{+}(i) \cdot \vec{\sigma} \cdot c(i) \tag{8}$$

$\vec{\sigma}$ denotes the vector of Pauli matrices and $\vec{S}(i)$ is a localized spin, J_{sd} is the indirect exchange integral.

2.3. The RKKY interaction

The RKKY interaction is a well know instrument for magnetic impurities properties development. This magnetic interaction is a basic model in the physics of magnetism [3]. The diagram formalism is described in some good monograph [4], [5]. The s-d indirect exchange interaction is determined by the Hamiltonian:

$$\hat{H}_{sd} = -J_{sd} \left(\hat{s}\left(\vec{R}_i\right) \cdot \vec{S}_j + \vec{s}\left(\vec{R}_j\right) \cdot S_j \right) \tag{9}$$

and is considered like a small perturbation. Here $\vec{R}_i$ and $\vec{R}_j$ are the space – vectors of the impurities, $\vec{S}_i$ and $\vec{S}_j$ are the spins impurities and $\hat{s}\left(\vec{R}_i\right)$ and $\hat{s}\left(\vec{R}_j\right)$ are electron spin operators in the corresponding space points. Using the simple matrix equation:

$$Tr\left(\sigma \cdot \vec{S}_i \sigma \cdot \vec{S}_j \right) = 2 \vec{S}_i \vec{S}_j \tag{10}$$

we obtain the space dependence of the indirect exchange:

$$\hat{H}_{RKKY} = -J\left(\vec{R}_{ij}\right) \vec{S}_i \vec{S}_j \tag{11}$$

where $\vec{R}_{ij} = \vec{R}_j - \vec{R}_i$, $R_{ij} = \left| \vec{R}_{ij} \right|$.

Here $J\left(\vec{R}\right)$ means the RKKY exchange integral and is given by [9]:

$$J(R) = B \int_{\varepsilon(K)\langle\mu} d^3k \int_{\varepsilon(h')\rangle\mu} d^3k' \frac{2\cos\left(\left(\vec{k} - \vec{k}'\right) \cdot \vec{R}\right)}{\frac{\hbar^2}{2m^*} \left(\vec{k}'^2 - \vec{k}^2 + i \frac{2m^*}{\hbar \tau} \right)} \tag{12}$$

where $B = \left(\frac{J_{sd}}{2} \right)^2 \left(\frac{a_0}{2\pi} \right)$ and $\mu = \frac{k_F^2 \hbar^2}{2m^*}$ is the Fermi energy and $\hbar k_F$ is the Fermi momentum [6].

The expression of the exchange integral is:

$$J(R) = \left(J_{sd}\right)^2 A \cdot F\left(2 k_F R\right) \tag{13}$$

where:

$$A = \left(\frac{a_0 k_F}{2} \right)^6 \frac{8}{\pi^3 \mu} e^{-R/\lambda} \tag{14}$$

and

$$\lambda = \frac{2k_F}{q^2} = \frac{\hbar k_F \tau}{m^*} \tag{15}$$

is the electron mean path [6]. We see that the function that describes the RKKY interaction is:

$$F(x) = \frac{x \cos x - \sin x}{x^4} \tag{16}$$

with

$$x = 2 k_F R_{ij} \tag{17}$$

The exchange interaction decrease like $1/R_{ij}^3$ in $R_{ij} \to \infty$ limit and oscillate with $2/k_F$ periodic distance. Thus the polarization of the conduction electrons induced by the local magnetic spins is not uniform and decrease with distance. Thus the interaction between the nearest–neighbor may be ferromagnetic or antiferromagnetic and the sign of this interaction is given by the RKKY function F(x) [3], [9].

2.4. The hybrid model

We propose a hybrid model that arises from these three models: RKKY, Hubbard, and DE. Our Hamiltonian is given by:

$$H = H_{Hubbard} + H_{RKKY} + H_{DE} \tag{18}$$

The latest two terms in this Hamiltonian are correlated by J_{sd} (the indirect exchange interaction).

We apply the Monte Carlo algorithm to describe this simulation magnetic model for a fcc lattice with parameter a_0 and for a perturbated lattice with a centered interstitial atom. We find that the magnetic properties are changing in this way. The condensed phase is changing and is find a magnetic transition AF → FM (antiferromagnetism → ferromagnetism). The generalized infinite dimensional band fcc lattice, with hopping scaled as [10]:

$$t_{ij} = \frac{1}{\sqrt{Z_{ij}}} \tag{19}$$

(where Z_{ij} is the number of neighbors) has a density of states (DOS), given by [11]:

$$N(\varepsilon) = \frac{\exp\left(-1\left(1+\sqrt{2}\varepsilon\right)/2\right)}{\sqrt{\pi\left(1+\sqrt{2}\varepsilon\right)}} \tag{20}$$

that presents a square–root singularity at the lower band edge.

Our parameters of work are the matrix (t_{ij}) of hopping, the Coulomb interaction (U), the constant of the lattice (a_0), the chemical potential μ, the Fermi wave – vector k_F, the indirect exchange integral J_{sd}. Thus, we can study the role of inter–site hybrid interactions in deciding ferromagnetic (or antiferromagnetic) state in the itinerant electron (narrow band) systems like the transition metals. We have considered Hubbard like tight binding model along with exchange and hybrid interactions. All these interactions have been treated within mean – field approximation [3]. It is well known that the onset of ferromagnetism in itinerant electron solid need not be due to the relative shift of the position of majority and minority spin bands as a result of large intra–atomic Coulomb interactions. The inter–atomic exchange interactions may play crucial roles for the onset of ferromagnetic state. It has been argued that the ferromagnetic state may be realized with inter–atomic exchange interactions alone. The onset of ferromagnetism is manifested through the narrowing and widening of, respectively, (majority) up and (minority) down spin electrons.

3. Monte Carlo algorithm

The Monte Carlo (MC) simulations are done an N^3 cubic lattice with periodic boundary conditions. Although the simulation MC is a semi-quantum concept (we use the standard Metropolis algorithm), for the localized spins we use the classic considerations and the itinerant electrons are considered in an quantum picture. The standard Metropolis algorithm that was use is in the fact the follow the spin random distribution is considered in start of algorithm and the random reorientation on the site **i** (for example), will induce the energy change ΔE. If the quantity $\exp(-\Delta E / k_B T)$ is smaller than a random number between 0 and 1, the change is allowed, otherwise it is rejected. In this mode we can determine different physics parameters of the considered lattice by the calculation of the statistical average of those parameters:

$$\langle A \rangle = \frac{\sum_i A_i \exp(-\beta E_i)}{\sum_i \exp(-\beta E_i)} \tag{21}$$

where $\beta = \frac{1}{k_B T}$ is the Boltzmann factor and A_i is a physical quantity A, in microstate "i". An obvious physical studied quantity is the magnetization given by: $M = \sum_{i=1}^{N} s_i$ and the magnetization per spin $m = \frac{m}{N}$. Usually we are interested in the average values $\langle M \rangle$ and the fluctuations $\langle M^2 \rangle - \langle M \rangle^2$. The zero field magnetic susceptibility χ is an example of a linear response function, because it measures the ability of a spin to respond to a change in the external magnetic field. χ is related to the fluctuations of the magnetization:

$$\chi = \frac{\langle M^2 \rangle - \langle M \rangle^2}{k_B T} \tag{22}$$

Thus we can determine the magnetic properties of the simulated model and next we can find the others characteristics parameters.

4. Results

a) Magnetic transition

Take in account the oscillatory behavior of the RKKY interaction we explain the magnetic transitions that appears in fcc Mn as a result of interstitial impurities inclusion (N for example). Thus, Mn_4N present band ferromagnetism while the pure Mn is antiferromagnetic. This is due to the dilatation of the crystalline lattice and thereafter, the increment of the distance between Mn ions. The lattice parameter in Mn_4N (solid solution) is 3.861 Å and x is 14.061. Thus the RKKY integral can change the sign and the magnetic transition is possible.

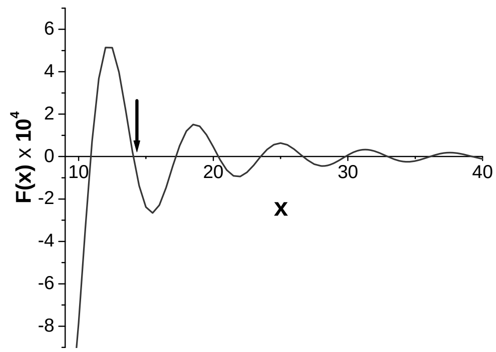

Fig.1. RKKY oscillating function.

b) Magnetization

We studied the magnetization in the MC simulation for our hybrid model for different values of $J_{sd}^{*} = J_{sd}/t$ (where t is the hopping factor).We find that the condensed phase is influenced by the coupling factor. The critical temperature is influenced by the indirect exchange coupling (see in Fig. 2):

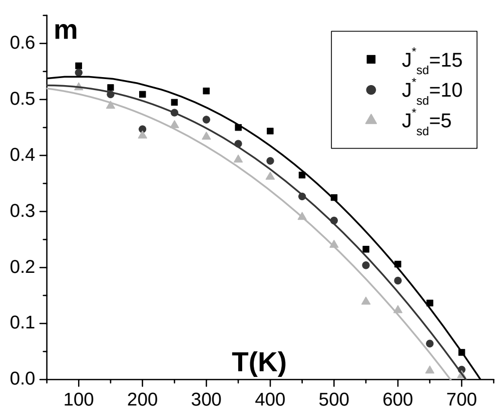

Fig. 2. The relative magnetization in fcc lattice with impurities (U=5).

c) The magnetic relative susceptibility

The magnetic relative susceptibility in a fcc lattice with interstitial impurities is investigated by our model in object to find the nature of the magnetic state. It is important to see (Fig. 3.) that the relative susceptibility (χ/χ_0) is enhanced by the existence of the impurities. (χ_0 is the corresponding susceptibility without impurities). The linear dependence of susceptibility (versus quadratic temperature) denotes a itinerant origin of the ferromagnetic state.

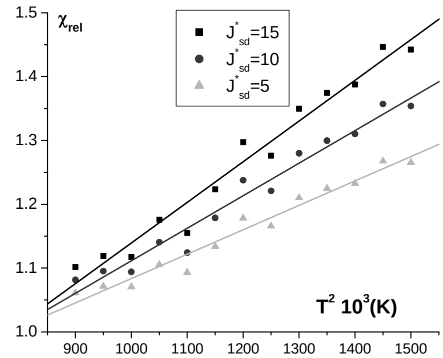

Fig. 3. Quadratic temperature dependence of the magnetic relative susceptibility (U=5).

d) Magnetic phase diagram

The phase diagram dependence by the indirect exchange integral is plotted in fig.4. The ferromagnetic domain is enhanced with the indirect exchange increasing (n is the filling level with itinerant electrons).

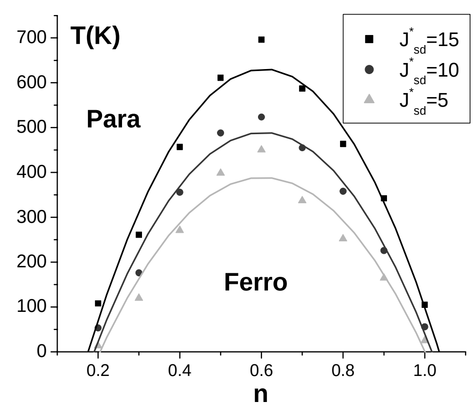

Fig.4. T vs. n phase diagram of the hybridmodel for a fcc lattice, for different J_{sd} (U=5).

5. Conclusions

The MC simulation is used to test the hybrid model that includes a composite between itinerant and localized standpoints. We investigated the influence of the impurities in the observed magnetic transition (band antiferromagnetism-band ferromagnetism). We take in account in our simulation the existence of non-magnetic interstitials impurities by the factor of hopping (in this way the lattice is transformed in a simple cubic lattice). By the magnetically point of view, the fcc structure is modified (the spin arrangement is parallel), thus the new ordered state is ferromagnetic. We verified the itinerant behavior of the condensate state and the dependence of the model by the indirect exchange coupling. We found the phase diagrams (critical temperature versus the filling level).

References

[1] M. A. Ruderman, C. Kittel, Phys. Rev. **96**, 99 (1954).
[2] T. Kasuya, Progr. Theoret. Phys. (Kyoto) **16**, 45 (1956).
[3] D. C Mattis, The Theory of Magnetism (Harper & Row, New York, 1965).
[4] E. M. Lifshitz, L. P. Pitaevskii, Course on Theoretical Physics, vol. 9, Statistical Physics, Part 2 (Pergamon Press, Oxford, 1982).
[5] A. A. Abrikosov, L. P. Goikov, I. E Dzyaloshinski, Methods of Quantum Field Theory in Statistical Physics (Dover, New York, 1963).
[6] T. M. Mishonov, T. I. Valchev, L. A. Atanasova, P. A. Ivanov, RKKY interaction in framework of T=0 Green function method, cond-mat, 28,548, (2003).
[7] J. Hubbard: Proc. Roy. Soc. London A **276**, 238 (1963).
[8] C. Zener, Phys. Rev. **82**, 403 (1951).
[9] A. A. Abrikosov, Fundamentals of the theory of metals, North-Holland, Amsterdam (1988).
[10] W. Metzner and D. Vollhardt, Phys. Rev., Lett. **62**, 324 (1989).
[11] E. Müler-Hartmann: Proc. V Symp. Phys. of Metals, ed. E.Talik and J. Szade, p22 (Poland 1991)
[12] T. Dietl, H. Ohno, F. Matsukura, Phys. Rev. B **63**, 195205 (2001).
[13] M. J. Calderón, G. Gómez-Santos, L. Brey, Phys. Rev. B **66**, 075218 (2002).
[14] A. Kaminski, S. Das Sarma, Phys. Rev. Lett. **88**, 247202 (2002).
[15] T. Hayashi, Y. Hashimoto, S. Katsumoto, Y. Iye, Appl. Phys. Lett. **78**, 1691 (2001).
[16] E. Muller-Hartmann, Z. Phys. B **74**, 507 (1989).
[17] R. Aguado, D. C. Langreth, Phys. Rev. B **67**, 245307 (2003).
[18] R. M. Fye, Phys. Rev. B **41**, 2490 (1990).
[19] J. E. Hirsch and R. M. Fye, Phys. Rev. Lett. **56**, 2521 (1981).
[20] R. Sordan, K. Balasubramanian, M. Burghard, K. Kern, Appl. Phys. Lett. **87**, 013106 (2005).

*Corresponding author: beneugen10@yahoo.com