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Monte Carlo simulation on the indirect exchange interactions of Co-doped ZnO film

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Monte Carlo simulations using a three-dimensional lattice model studied the Ruderman–Kittel–Kasuya–Yosida (RKKY) indirect exchange interaction of doped magnetic Co ions in ZnO films. The results of the calculations show that the RKKY interaction in Co-doped ZnO is long ranged and its magnitude is proportional to R^{-1} (inverse of the distance R from a central Co^{2+} ion). The sign oscillates with a frequency that depends on the concentration of the carrier. The long-distance sum of the RKKY indirect exchange energies is positive indicating that these materials are ferromagnetic, in direct correlation with previously reported results. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1508168]

Recently, diluted magnetic semiconductors have been proposed as possible materials for "spintronic" (spin + electronic) devices. Such devices may have many applications in a new generation of information storage. For example, the electronic properties of spintronic devices allow for the possibility of temporary information storage (i.e., memory cache). The ferromagnetism of spintronics media makes them good candidates for permanent information storage devices.

In p-doped $Zn_{1-x}Mn_xTe$ epilayers[1] and lead-salt materials $Pb_{1-x-y}Sn_yMn_xTe$,[2] the injection of the magnetic atoms into the semiconductor matrix has proven to be a good way to produce carrier-induced ferromagnetism. In this study, we will focus on the transition-metal-doped zinc oxide (n-type $Zn_{1-x}M_xO$: M=Co, Ni, Cr, and Mn). $Zn_{1-x}Co_xO$ was chosen not only because it forms transparent ferromagnetic material[3] but also because recent calculations suggest a high Curie temperature, T_C.[4,5] Ueda *et al.* considered three possible origins for the ferromagnetism in $Zn_{1-x}Co_xO$. The first is the ferromagnetism of CoO, the second is the micro-Co cluster, and the third is what is known as the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction.[6] The RKKY interaction between two magnetic spin impurities across a tunneling junction is studied when the system is driven out of equilibrium through biasing the junction. Due to the weak ferromagnetism of CoO and the large magnitude of magnetization of $Zn_{1-x}M_xO$, the first possibility is immediately excluded. The second possibility is also eliminated because experiments show that the Co phase, as well as the wurtzite structure, is not changed, as the Co ions are randomly substituted for Zn sites.

The RKKY interaction theory seems to be a relatively reasonable explanation for the origin of ferromagnetism in these materials. This phenomena has been observed in several varieties of semiconductors, like p-(In,Mn)As/GaSb (Ref. 7) as well as in $Cd_{1-x}Mn_xTe$.[8]

The polarization of the conduction electrons in the n-type $Zn_{1-x}Co_xO$ will lead to an $s-d$ exchange process. The final result of the RKKY exchange coupling[6] is given by

$$H_{\text{eff}}^{\text{ex}}=\frac{9\pi J_0}{2\ E_F}\left(\frac{n_e}{N}\right)^2\sum_{\substack{lm\\ l\neq m}} F(2k_FR_{lm})S_l\cdot S_m, \tag{1}$$

$$F(x)=\frac{x\cos x-\sin x}{x^4}, \tag{2}$$

where N [in Eqs. (1) and (2)] is the density of the localized spins carried by Co ions, E_F denotes the Fermi energy, J_0 is the exchange constant that only relates to the ions type of the localized magnetic spin (S_l and S_m). Thus, J_0 is the square of the interaction between a localized spin and the conduction electron–electron spins. Finally, the density of the conduction electrons is expressed by n_e. For the three-dimensional material, the wave vector at the Fermi surface is given by[9]

$$k_F=(3\pi^2n_e)^{1/3}. \tag{3}$$

R_{lm} is the relative distance between the two localized spins (S_l and S_m). For the convenience of computation, we can transform the RKKY expression into another similar form

$$H_{\text{eff}}^{\text{ex}}=\frac{9\pi J_0}{2\ E_F}\left(\frac{n_e}{N}\right)^2(S\cdot S)\sum_R F(2k_FR)G(R), \tag{4}$$

where $G(R)$ is the average number of Co^{2+} ions that can be found in a three-dimensional material with the relative distance R from a central Co^{2+} ion. $G(R)$ is the Co^{2+} ion spherical distribution function. In this RKKY expression, we have assumed that only the indirect exchange interaction between localized magnetic Co^{2+} ions, which are induced by the conduction electrons, contributes to the RKKY exchange energy of the material. We also assume that the magnitude of

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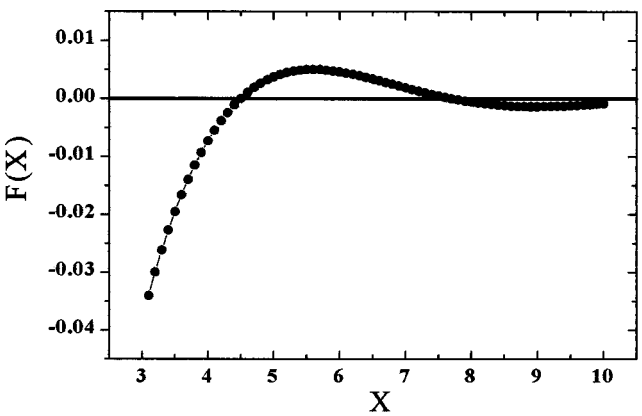

FIG. 1. Evaluation of the $F(X)$ function.

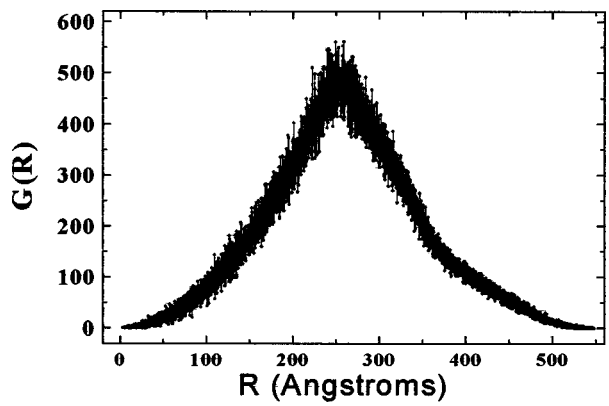

FIG. 2. Co^{2+} spherical distribution function of $Zn_{0.85}Co_{0.15}O$. We can omit the right-hand side relative to the peak. The left-hand side part of the graph is shown in Fig. 3.

the Co^{2+} ions is constant, and this value for Co^{2+} is equal to $S=3/2$. We can also assume that all of the localized spins point in the same direction. So $S\cdot S$ is positive under this condition. Since $9\pi/2J_0/E_F(n_e/N)^2(S\cdot S)$ is independent of the relative distance R, we can denote it as a positive constant C. The final expression of the RKKY indirect exchange energy is given as

$$H_{\text{eff}}^{\text{ex}}=C\sum_R F(2k_FR)G(R). \tag{5}$$

Now, the approach is used to determine the distribution function $G(R)$ and to sum its product with the known oscillating function $F(2k_FR)$ over all possible relative distances, R. Because C is positive, ferromagnetism is favored if the sum is negative, otherwise, RKKY would suggest the material is not ferromagnetic.

We can easily see from Eq. (2), that as $F(x)\rightarrow-\infty$, $x\rightarrow\infty$. From Fig. 1, we can see that the magnitude of $F(x)$ drops abruptly with increasing x values, while the sign oscillates. The least positive solution of

$$\frac{x\cos x-\sin x}{x^4}=0 \tag{6}$$

is determined to be at $x=4.5$. For most of the n-type $Zn_{1-x}Co_xO$ semiconductors,[3] the carrier concentration is below 1.0×10^{20} cm^{-3}, so k_F is less than $0.1(\text{Å})^{-1}$. Except in extremely diluted systems, the average distance of the nearest-neighbor magnetic atoms is on the order of several angstroms. Therefore, $k_FR_{\text{nearest}}<4.5$, which means that the first several layers of the neighbor cobalt ions usually favor ferromagnetism. To improve the efficiency of the computer calculations, we set the upper limit of relative distance as

$$R_{\text{cutoff}}=\frac{2\times4.5}{2\times k_F}=\frac{4.5}{k_F}. \tag{7}$$

The exactness of the cutoff distance will be addressed in this letter.

A crystalline lattice of dimensions $N\times N\times N$ was constructed. First, a single Co^{2+} ion was fixed at the geometrical center. Then, M additional Co^{2+} ions were randomly placed on the lattice (here M is determined by the total number of lattice cites times the molar percent of the Co^{2+} ions). Finally, the distance between each Co^{2+} ion and the central ion was determined and the distribution function, $G(R)$, computed. This procedure was repeated 100 times with different random seeds for each $Zn_{1-x}Co_xO$ sample to improve the results of the numerical simulation.

ZnO is a well-known wurtzite hexagonal-close-packed structure.[10] The geometrical parameters of the crystal structure we used are $a_0=3.24$ Å and $c_o=5.18$ Å.[11] We studied two samples, $Zn_{0.85}Co_{0.15}O$ (denoted as sample 1 or S1) and $Zn_{0.75}Co_{0.25}O$ (denoted as sample 2 or S2). For S1, the lattice dimension was set to $100\times100\times100$. The carrier concentration is 2.9×10^{20} cm^{-3}, and thus $k_F=(3\pi^2\times2.9\times10^{20}\text{ cm}^{-3})^{1/3}\approx0.205$ Å^{-1}. Figure 2 shows the Monte Carlo simulation result of the calculation of $G(R)$ with a step size of $\Delta R=0.1$ Å. There is a peak on the plot, but it is due to the limit size of the bulk that we use. This value indicates that the relative distance R reaches the boundary of the lattice. We should omit the right-hand side relative to the peak. If we do this, it is reasonable because the magnitude of $F(2k_FR)$ will be very small beyond that boundary. The left-hand side part of $G(R)$ is shown in Fig. 3 and $F(2k_FR)\times G(R)$ is shown in Fig. 4. The $\Sigma F(2k_FR)G(R)$ give the final result as -1.0826, which means the RKKY indirect exchange interaction, does favor the ferromagnetism for S1.

Now let us examine the exactness of the upper limit when the effective distance of the RKKY interaction is $R_{\text{cuteff}}=4.5/0.205\approx21.4$ Å. The data show that when R is greater than R_{eff}, the function $G(R)$ becomes continuous, and so we can transform this summation into an integral. A

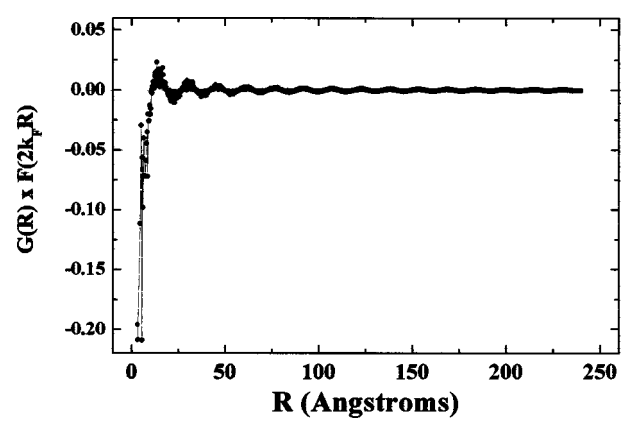

FIG. 3. Co^{2+} spherical distribution function of $Zn_{0.85}Co_{0.15}O$. [left-hand side part of $G(R)$].

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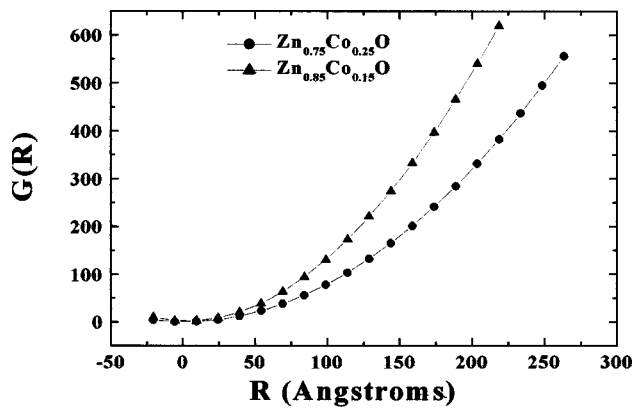

FIG. 4. $G(R)*F(2k_FR)$ distribution function over R for $Zn_{0.85}Co_{0.15}O$.

polynomial fit (Fig. 5) gives the following expression of $G(R)$ as

$$G(R)=0.81178-0.02062R+0.00813R^2-1.82671\times10^{-7}R^3. \quad (8)$$

Then, $\int_{R_{\text{cutoff}}}^{\infty} G(R)F(2k_FR)dR \approx -0.0092$ has a magnitude that is much less than the whole summation (equal to -1.0826), so we can confidently neglect the RKKY exchange interaction beyond the R_{cuteff} value. For S2, a $80\times80\times80$ crystal lattice was employed. The value of k_F is determined to be 0.0329 Å^{-1} based on the carrier concentration value of $1.2\times10^{-18}\ \text{cm}^{-3}$. The sum of $\Sigma F(2k_FR)G(R)$ for this sample is -460.13. The polynomial fit gives the expression of $G(R)$ as

$$G(R)=2.2397-0.096\,97R+0.014\,74R^2-6.3321x10^{-6}R^3. \quad (9)$$

The integral $\int_{R_{\text{cutoff}}}^{\infty} F(2k_FR)G(R)dR$ above R_{eff} gives a value of -1.8941, which poses a magnitude that is much less than the total sum.

The negative values for $\Sigma F(2k_FR)G(R)$ indicate that the RKKY indirect exchange interaction favors ferromagnetism for both Co-doped ZnO spintronics samples considered in this study. This result is in agreement with the values of Ueda *et al.*[3] In Eq. (4), J_0 only depends on the type of the localized ions. The difference of e_F in the two samples (S1 and S2) can be neglected when compared to the difference of the carrier concentration. It is thus possible to express their ratios as

$$\frac{H_{\text{eff}}(S1)}{H_{\text{eff}}(S2)} \approx 137.4. \quad (10)$$

Because the Curie temperature and the magnetization under external magnetic fields increase with the greater exchange interaction, the much higher RKKY of S1 (over 100 times greater than S2) can be explained in terms of its higher Curie temperature and greater magnetization.

In both of the samples examined here, a_2 is much greater than a_3 in the following general expression for $G(R)$:

$$G(R)=a_0+a_1R+a_2R^2+a_3R^4. \quad (11)$$

When R is very large,

$$G(R)\propto R^2, \quad (12)$$

and $|F(2k_FR)|\propto R^{-3}$, thus the RKKY interaction decays with the inverse of the relative distance, as given by

$$|H_{\text{eff}}|\propto R^{-1}. \quad (13)$$

This result explains why the final result is insensitive to the cutoff distance (R_{cutoff}).

Since the magnitude of the RKKY interaction decays with increasing relative distance, the sign of the RKKY interaction of several nearest layers of the localized magnetic ions will have the greatest effect in determining the sign of the final calculated result. We can approximately determine whether the ferromagnetism of the crystal is favored by the RKKY interaction by simply multiplying k_F by all the possible nearest relative distances between the localized magnetic atoms or ions. If $2k_FR_{\text{nearest}}<4.5$, then it is favored; otherwise, it is unfavorable.

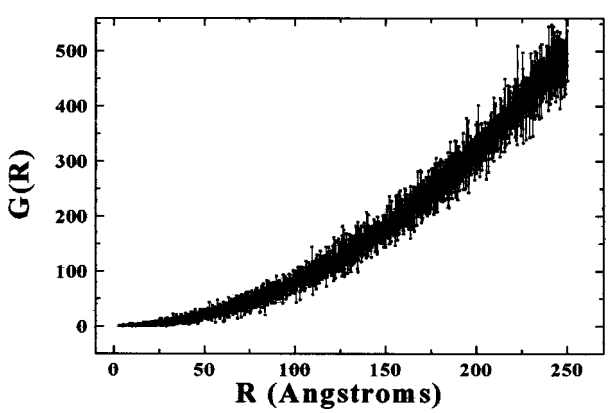

FIG. 5. Polynomial fit of the $G(R)$ function.

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