

Low-temperature spin glass in IV-VI semimagnetic semiconductors

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(Received 8 May 1989)

We investigate an indirect exchange interaction between the ions of the magnetic component of a solid solution. It is shown that the indirect interaction in IV-VI semimagnetic semiconductors via the virtual electron-hole excitations is anisotropic, and it has the non-Heisenberg form.

The small-gap semimagnetic semiconductors (Pb,Sn,Ge)_{1-x}Mn_x(Te,Se) have been a subject of considerable recent interest.¹ The most essential events, that have happened recently, are the discovery of the spin-glass (SG) state in Pb_{1-x}Mn_xTe,¹ and the observation of the complex phase diagrams in SnMn-Te and Pb-Sn-Mn-Te with paramagnetic, ferromagnetic, and SG phases.²⁻⁴ In order to analyze the experiments on the thermodynamical properties of the alloys (e.g., magnetization is an external magnetic field, magnetic susceptibility, heat capacity) we have to investigate the particularities of the magnetic interactions. Therefore, the calculations of the spatial and angular dependences of the indirect interactions are needed, especially for the elucidation of the reason for the SG phase.

The polarization indirect exchange is due to *s-d* or *s-f* interactions, which depend on the energy-band structure and the wave functions of *s,d* electrons. If we assume the *d* electrons to be localized with the δ -type wave function, then the *s-d* interaction is a contact interaction. All the updated calculations were based on this assumption, and we shall use it without discussion. In the case of a metal with free *s* electrons the second-order *s-d* indirect interaction is isotropic. It is the oscillating function of the distance *R* between localized spins with the period π/k_F (k_F is the Fermi wave vector), depending on *R* as R^{-3} .⁵ The oscillating sign of this interaction is commonly used to explain the SG transition in magnetically doped metals⁶⁻⁸ and degenerate semiconductors.⁴ We have to note the isotropy and Gaussian distribution of random exchange interaction.⁹

In semiconductors with parabolic energy spectra, the exchange interaction has a definite sign. Moreover, it decreases exponentially at large distances, with the interaction length being proportional to the square root of the energy gap.¹⁰

The indirect exchange of alternating sign, leading to the SG transition, has been obtained in the framework of the model of the indirect-band semiconductor. In this case the period of the oscillations appeared to be proportional to the inverse separation of the conduction- and valence-band extrema in momentum space.¹¹ When the oscillating factor is absent, as in nondegenerate direct-gap semiconductors, the sign of the exchange interaction is important. If the exchange interaction is antiferromagnetic, and the magnetic atoms are distributed randomly, the SG phase is likely.

For Pb_{1-x}Mn_xTe alloys the calculations performed in

Ref. 12 suggest the ferromagnetic exchange interaction which opposes the experimentally discovered SG phase of these compounds. However, taking into account the non-parabolicity within the Kohn-Luttinger model, it was shown¹³ that the exchange interaction is antiferromagnetic with a R^{-3} dependence at distances $R \ll \hbar v/E_g$ (E_g is the energy gap and *v* is the velocity interband matrix element in $\mathbf{k} \cdot \mathbf{p}$ approach).

The common feature of the above-mentioned calculations is the isotropy of the indirect exchange. This is a result of an oversimplification, related to the choice of the wave functions, depending upon the projection of the spin on the movement direction. If we take into account this dependence, we immediately obtain the anisotropic non-Heisenberg exchange. For example, in the crystals of zinc-blende structure the exchange has been shown to display such an anisotropy provided that the spin-orbit coupling has been taken into account.¹⁴

In this paper we investigate a nontrivial example of the complex exchange interaction that takes place in IV-VI crystals with the Dirac Hamiltonian of free electrons and holes.

By using the Luttinger-Kohn representation, the Hamiltonian of the electrons in the vicinity of the *L* point of the Brillouin zone for IV-VI compounds can be described by the Dimmock model.^{15,16} We shall use the simplified model which ignores the contribution of the distant energy bands to the energy spectrum of the electrons and holes. Then the Hamiltonian in the space of four-spinors (two bands plus spins) has the form of the anisotropic Dirac model

$$H_0 = v_z \alpha_z p_z + v_{\perp} \alpha_{\perp} \cdot \mathbf{p}_{\perp} + \frac{\beta}{2} E_g, \quad (1)$$

where

$$\alpha = \begin{pmatrix} 0 & \sigma \\ \sigma & 0 \end{pmatrix}, \quad \beta = \begin{pmatrix} \mathbf{1} & 0 \\ 0 & -\mathbf{1} \end{pmatrix}, \quad (2)$$

v_z and v_{\perp} are the velocity matrix elements, E_g is the energy gap, and $\mathbf{1}$ is the two-dimensional unit matrix. The *z* axis is oriented along the [111] direction.

The interaction of the electrons with magnetic impurities in the second-quantization representation can be written as

$$H_1 = \frac{1}{N} \sum_{k,q,n} J_{ij} e^{iq \cdot \mathbf{R}_n} \mathbf{S}_n \cdot \sigma_{\alpha\beta} a_{kai}^{\dagger} a_{k+q,\beta j}, \quad (3)$$

where $J_{ij} = J \delta_{ij}$ are the matrix elements of the contact *s-d* interaction, calculated with the wave functions of $i, j = 1, 2$

band extrema, and \mathbf{R}_n are the localized spin positions. Since the values of the electron and hole integrals J_{11} and J_{22} are not known, we simply assume that $J_{11} = J_{22} = J$.

The contribution to the energy from the pair interaction of the localized spins \mathbf{S}_1 and \mathbf{S}_2 can be represented by a diagram of the second order in s - d interaction, see Fig. 1. The corresponding analytical expression is written down with the aid of the standard rules of the finite-temperature-diagram technique:¹⁷

$$H_{\text{eff}} = \frac{T}{(2\pi\hbar)^6 N^2} \sum_{\omega_n} \int d^3p d^3p' e^{i\mathbf{R} \cdot (\mathbf{p} - \mathbf{p}')/\hbar} \text{Sp}(\mathbf{S}_1 \cdot \boldsymbol{\sigma}) \hat{J} \hat{G}(\mathbf{p}\omega_n) (\mathbf{S}_2 \cdot \boldsymbol{\sigma}) \hat{J} \hat{G}(\mathbf{p}'\omega_n), \quad (4)$$

where N is the density of atoms in the host lattice, $\omega_n = (2n+1)\pi T$,

$$\hat{J} = J \begin{pmatrix} 1 & 0 \\ 0 & \perp \end{pmatrix}, \quad \hat{G} = (i\omega_n - H_0)^{-1}. \quad (5)$$

It is convenient to carry out a scale transformation under the integral in (4):

$$p_z \rightarrow \frac{v}{v_z} p_z, \quad p_{x,y} \rightarrow \frac{v}{v_{\perp}} p_{x,y}, \quad R_z \rightarrow \frac{v_z}{v} R_z, \quad R_{x,y} \rightarrow \frac{v_{\perp}}{v} R_{x,y}, \quad v = (v_{\perp}^2 v_z)^{1/3}.$$

Then we have the isotropic function of momentum under the integral (4). After calculating the trace in (4) we get

$$H_{\text{eff}} = \frac{J^2 T}{(2\pi\hbar)^6 N^2} \sum_{\omega_n} \left[\mathbf{S}_1 \cdot \mathbf{S}_2 \left(-\omega_n^2 I_1 + I_2 - \frac{v^2}{R^2} I_3 \right) + \frac{2v^2}{R^4} I_3 (\mathbf{S}_1 \cdot \mathbf{R})(\mathbf{S}_2 \cdot \mathbf{R}) \right], \quad (6)$$

where

$$I_1 = \int d^3p d^3p' \frac{\exp\left[\frac{i}{\hbar} \mathbf{R} \cdot (\mathbf{p} - \mathbf{p}')\right]}{(\omega_n^2 + E_p^2)(\omega_n^2 + E_{p'}^2)}, \quad I_2 = \frac{E_g^2}{4} I_1, \quad (7)$$

$$I_3 = \int d^3p d^3p' \frac{(\mathbf{p} \cdot \mathbf{R})(\mathbf{p}' \cdot \mathbf{R}) \exp\left[\frac{i}{\hbar} \mathbf{R} \cdot (\mathbf{p} - \mathbf{p}')\right]}{(\omega_n^2 + E_p^2)(\omega_n^2 + E_{p'}^2)}, \quad E_p^2 = \frac{E_g^2}{4} + v^2 p^2.$$

The integrals I_1 , I_2 , and I_3 can be evaluated analytically

$$I_1 = \left[\frac{2\pi^2 \hbar E_g}{R v^2} \right]^2 \exp\{i\alpha[(i\tilde{\omega}_n)^2 - 1]^{1/2}\},$$

$$I_3 = \left[\frac{\pi^2 \hbar E_g}{v^3} \right]^2 \left[\frac{\tilde{R}}{R} - i[(i\tilde{\omega}_n)^2 - 1]^{1/2} \right]^2 \exp\{i\alpha[(i\tilde{\omega}_n)^2 - 1]^{1/2}\}, \quad (8)$$

$$\alpha = \frac{2R}{\tilde{R}}, \quad \tilde{R} = \frac{2\hbar v}{E_g}, \quad \tilde{\omega}_n = \frac{2\omega_n}{E_g}.$$

To calculate H_{eff} at $T=0$, in (6) we should make the known substitution¹⁷

$$i\omega_n \rightarrow \omega + i\omega \text{sgn} \omega, \quad T \sum_n \rightarrow \frac{1}{2\pi i} \int d\omega.$$

Performing the contour integration we obtain the result in the following form:

$$H_{\text{eff}} = \frac{J^2 E_g^3}{64\pi^3 \hbar^4 N^2 R^2 v^4} \{ [K_1(\alpha) - K_3(\alpha)] \mathbf{S}_1 \cdot \mathbf{S}_2 + \left[K_3(\alpha) + \frac{2}{\alpha} K_2(\alpha) \right] (\mathbf{S}_1 \cdot \mathbf{n})(\mathbf{S}_2 \cdot \mathbf{n}) \}, \quad (9)$$

where \mathbf{n} is the unit vector along \mathbf{R} , $K_\nu(\alpha)$ is the modified Bessel function.

Now we examine the particular cases of (9). (i) At large distances, $\alpha \gg 1$,

$$H_{\text{eff}} = \frac{J^2 E_g^{5/2} e^{-2R/\tilde{R}}}{64\sqrt{2}\pi^{5/2} (\hbar v)^{7/2} N^2 R^{5/2}} \left[(\mathbf{S}_1 \cdot \mathbf{n})(\mathbf{S}_2 \cdot \mathbf{n}) - \frac{2\tilde{R}}{R} \mathbf{S}_1 \cdot \mathbf{S}_2 \right]. \quad (10)$$

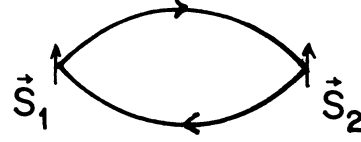


FIG. 1. The diagram for the exchange interaction of two spins.

(ii) At short distances, $\alpha \ll 1$,

$$H_{\text{eff}} = \frac{J^2}{16\pi^3 \hbar v N^2 R^5} [3(\mathbf{S}_1 \cdot \mathbf{n})(\mathbf{S}_2 \cdot \mathbf{n}) - 2\mathbf{S}_1 \cdot \mathbf{S}_2]. \quad (11)$$

As follows from (10) and (11), for two momenta localized at large distances the minimal energy corresponds to the antiferromagnetic ordering of pairs, and at short distances to the ferromagnetic one. We emphasize that the pseudodipolar contribution to the exchange interaction is not only related with the spin-orbit coupling of the band electrons. It is caused by the spin-preserving $\mathbf{k} \cdot \mathbf{p}$ contribution to the Hamiltonian as well. The calculated indirect in-

teraction has the alternating sign and depends on the angle between the localized spins and a vector connecting them. This can explain the SG ordering without additional assumption about the Ruderman-Kittel-Kasuya-Yosida interaction.^{3,4}

The expressions (9)–(11) describe the exchange interaction mediated by the virtual electron-hole pairs in the single L valley. In the isotropic case ($v_{\perp} = v_z$), the summation over four nonequivalent L valleys is trivial and leads to the multiplication by four in (9)–(11). If the spectrum is anisotropic, we have to carry out the inverse scale transformation in (9)–(11), i.e., to substitute

$$\mathbf{R} \rightarrow \frac{v}{v_{\perp}} [R^2 + \beta(\mathbf{L}_m \cdot \mathbf{R})^2]^{1/2}, \quad \mathbf{S} \cdot \mathbf{R} \rightarrow \mathbf{S} \cdot \mathbf{R} + \gamma(\mathbf{S} \cdot \mathbf{L}_m)(\mathbf{R} \cdot \mathbf{L}_m), \quad \beta = \frac{v_{\perp}^2}{v_z^2} - 1, \quad \gamma = \frac{v_{\perp}}{v_z} - 1,$$

where \mathbf{L}_m are the unit vectors along the four nonequivalent $\langle 111 \rangle$ directions. After that the summation over m must be done. For instance, at short distances we find from (11)

$$H_{\text{eff}} = \frac{J^2}{2\pi^3 \hbar v N^2 R^5} \left\{ -\mathbf{S}_1 \cdot \mathbf{S}_2 + \frac{3v_{\perp}^2}{8v^2} \sum_{m=1}^4 \frac{[\mathbf{S}_1 \cdot \mathbf{n} + \gamma(\mathbf{S}_1 \cdot \mathbf{L}_m)(\mathbf{n} \cdot \mathbf{L}_m)][\mathbf{S}_2 \cdot \mathbf{n} + \gamma(\mathbf{S}_2 \cdot \mathbf{L}_m)(\mathbf{n} \cdot \mathbf{L}_m)]}{1 + \beta(\mathbf{L}_m \cdot \mathbf{n})} \right\}. \quad (12)$$

The temperature of the SG freezing T_g is determined by the magnitude of the interaction at mean distances. There is a characteristic relative magnetic impurity concentration $x_0 = (aE_g/\hbar v)^3$ (a is the lattice constant) and two regions $x \gg x_0$ and $x \ll x_0$, where T_g is determined by the formulas (10) and (11), respectively. For the parameters of $\text{Pb}_{1-x}\text{Mn}_x\text{Te}$, $a = 6 \text{ \AA}$, $E_g = 0.1 \text{ eV}$, $v = 3 \times 10^7$

cm/sec we have $x_0 = 0.03$. Assuming $J = 1 \text{ eV}$ and $x = 0.1$, we obtain from (12) that $T_g = 2.5 \text{ K}$ in good accordance with the experimental data. Note that the concentration dependences of T_g are different in the two above-mentioned regions, i.e., $T_g \sim x^{5/6} \exp[(-x_0/x)^{1/3}]$ at $x \ll x_0$ and $T_g \sim x^{5/2}$ at $x \gg x_0$.

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