Indirect interaction of magnetic impurities in a semiconductor in the field of a strong electromagnetic wave

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We calculate the exchange interaction between impurity spins in a nondegenerate semiconductor whose carriers interact coherently with a strong resonant light wave. We show that the exponential spatial decrease of the exchange, with a decrement equal to the coherence length ξ_0, is modulated by oscillations having a period $\sim\hbar\omega - Eg$ (detuning from resonance). The conditions for the formation of photoinduced spin glass are determined.

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1. An optically induced rise in the Curie temperature T_C in magnetic semiconductors was predicted in Ref. 1. This effect is due to the fact that electrons photoexcited into the conduction band participate in the indirect exchange. For an experimentally noticeable shift of T_C, however, the necessary density of the nonequilibrium carriers is too high.[2] In measurements of the transition-temperature shift it is necessary here to exclude the effect of crystal heating. Such an experiment was performed on an EuS crystal,[3] and the shift obtained was ~0.1 K.

In this paper we call attention to another effect connected with optical pumping in a semiconductor containing magnetic impurities. The light can produce magnetic ordering of the impurities both in the disordered-ferromagnet regime and in the spin-glass regime. Impurity spins acting as a disordered ferromagnet were investigated in Ref. 4.

Interest in the production of a disordered magnet with spin-glass behavior has stimulated the study of various versions of direct impurity-spin interactions in a crystal matrix.[5,6] The form of the exchange interaction between the impurity spins is determined by the form of the electron spectrum. In a metal, such an interaction is an RKKY potential that oscillates rapidly with distance.

If a gap exists at the Fermi level, i.e., if a threshold energy is needed to produce an electron-hole pair, the exchange between the impurity atoms decreases exponentially with distance. This situation obtains in a nondegenerate semiconductor.[7] In a semiconductor with indirect gap, the exponential decrease is modulated by oscillations whose period is determined by the separation between the extrema in momentum space. Such a potential realizes a spin-glass state in a semiconductor.

The purpose of the present paper is to clarify the effect of strong optical pumping on indirect exchange in a semiconductor with a direct gap E_g.

The field of a strong electromagnetic wave with quantum energy $\hbar\omega \gtrsim E_g$ alters the electron spectrum of the semiconductor. A gap appears in the spectrum of the electron + field quasiparticles at the Fermi level,[8] and has been observed experimentally in Ref. 9.

The effect exerted on the exchange between impurity spins by the change of the spectrum in the light-wave field can be easily understood qualitatively. The appearance of the Fermi quasilevel introduces into the system a parameter with the dimension of length, which determines the period of the oscillations. On the other hand, the gap in the quasiparticle spectrum causes an exponential decrease, with distance, of the exchange interaction of the impurity spins. It is thus to be expected that the exchange varies like an exponential modulated by an oscillating factor, and the damping decrement as a function of distance is determined by the size of the gap, while the period of the oscillations is determined by the detuning $\hbar\omega - E_g$ from resonance. A spin-glass state can arise at certain relations between the values of the gap, of the detuning, and of the impurity-spin density.

These qualitative arguments will be confirmed below by a formal calculation of the dependence of the exchange on the distance.

2. Consider the Hamiltonian of the two-band s–d model with account taken of dipole resonant transition in the electric field of a light wave:

$$H=\sum_{k i\alpha}\varepsilon_{k i}n_{k i\alpha}+\lambda\sum_{k\alpha}(a^{+}_{k 1\alpha}a_{k 2\alpha}e^{-i\omega t}+\text{H.c.})\cdot -\frac{1}{N}\sum_{kqm}\sum_{\alpha\beta ij}J_{ij}\mathbf{S}_m\sigma_{\alpha\beta}a^{+}_{k i\alpha}a_{k+q j\beta}e^{-iq\mathbf{R}_m}, \tag{1}$$

J_{ij} are the matrix elements of the contact s–d interaction on the Bloch functions of the bands, $i,j=1,\ 2$: $a_{k i\alpha}$ is the annihilation operator for an electron in a state $\mathbf{k}$ and with a spin projection α; the spectrum $\pm\varepsilon_{\mathbf{k}}=\pm(E_g/2+\hbar^2\mathbf{k}^2/2m)$ is measured from the midpoint of the forbidden band; $\mathbf{R}_m$ are the positions of the localized spins; σ are Pauli matrices; $\lambda=e\mathbf{E}\mathbf{p}/m\omega$; $\mathbf{p}$ is a matrix element of the dipole interband transitions (we assume for simplicity that it is independent of momentum[8]); $\mathbf{E}$ is the electric field strength in the wave, and N is the number of the basic atoms in the crystal.

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Under strong-field conditions $\lambda\tau > 1$ (τ is the shortest of the relaxation time on phonons, impurities, or electrons), a unitary transformation[8] can be used to change over to a representation in which the first two terms of (1) are independent of time. The magnetic impurities give rise to recombination whose rate is determined by the matrix elements J_{12}. This manifests itself formally in the appearance, after the unitary transformation, of nonresonant terms in (1) that oscillate rapidly with time. These processes hinder the establishment of the saturation state; they can be neglected by virtue of the inequality $cJ_{12}/E_g \ll 1$ (Ref. 10) (c is the relative density of the magnetic centers). Thus, we assume the stronger inequality $\lambda\bar{\tau} \gg 1$ to be satisfied ($\bar{\tau}^{-1}$ is the sum of the frequencies of the relaxation and the recombination on account of the magnetic centers). The stationary-state Hamiltonian is then

$$H = H_0 + H_{int};$$
$$H_0 = \sum_{k\alpha}\left\{\left(\varepsilon_k - \frac{\hbar\omega}{2}\right)n_{k1\alpha} + \left(-\varepsilon_k + \frac{\hbar\omega}{2}\right)n_{k2\alpha}\right\} + \lambda\sum_{k\alpha}(a^+_{1k\alpha}a_{2k\alpha} + \text{H.c.}); \tag{2}$$
$$H_{int} = -\frac{1}{N}\sum_{kqm}\sum_{\alpha\beta i} J_{it}\mathbf{S}_m\sigma_{\alpha\beta}a^+_{ik+q\alpha}a_{ik\beta}e^{-iqR_m}.$$

3. The exchange between impurity spins on account of the s–d interaction in (2) is determined by a simple loop that involves, since H_0 is not diagonal in the bands, matrix Green's functions:

$$J(R) = \frac{J^2T}{2N^2}\sum_q e^{-iqR}\sum_{nk}\mathrm{Sp}\,G(\mathbf{k}, \omega_n)G(\mathbf{k}+\mathbf{q}, \omega_n), \tag{3}$$

$$G(\mathbf{k}, \omega_n) = \frac{1}{(i\omega_n - E_{k1})(i\omega_n - E_{k2})}\begin{pmatrix} i\omega_n + \xi_k & \lambda \\ \lambda & i\omega_n - \xi_k \end{pmatrix}, \tag{4}$$

where

$$J = J_{11} = J_{22}, \quad E_{k1,2} = \pm(\xi_k^2 + \lambda^2)^{1/2}, \quad \omega_n = \pi T(2n+1),$$
$$\xi_k = \hbar^2(k^2 - k_F^2)/2m, \quad k_F^2 = m(\hbar\omega - E_g)/\hbar^2.$$

Calculating the trace in the sum over the frequencies in (3), we obtain for $J(R)$ at $T = 0$

$$J(R) = \frac{J^2}{2n^2(2\pi\hbar)^6}\int d^3p\,d^3p'\exp\left\{-\frac{i\mathbf{R}}{\hbar}(\mathbf{p}'-\mathbf{p})\right\}I(\mathbf{p}, \mathbf{p}'), \tag{5}$$

where

$$I(\mathbf{p}, \mathbf{p}') = (E_pE_{p'} - \xi_p\xi_{p'} - \lambda^2)/E_pE_{p'}(E_p + E_{p'}), \tag{6}$$

and n is the exchange density of the main atoms of the crystal. We carry out in (5) an integration over the angles, which pertains only to the exponential:

$$J(R) = \frac{J^2}{8n^2\pi^4\hbar^4R^2}\int_0^\infty p\,dp\int_0^\infty p'\,dp'\left[\cos\frac{p'-p}{\hbar}R - \cos\frac{p'+p}{\hbar}R\right]I(p, p'). \tag{7}$$

Integrals of this type are encountered in calculations of screening in an optical dielectric[11] and in the calculation of the Meissner effect in superconductors.[12]

Linearizing the spectrum near p_F and changing to integration with respect to ξ_p and $\xi_{p'}$ with the state density at the Fermi level we can, by making the change of variables $\xi_p = \lambda\sinh t$ and $\xi_{p'} = \lambda\sinh t'$ and using the condition $p_F^2 \gg m\lambda$, reduce (7) to the expression

$$J(R) = -\frac{J^2m^2\lambda}{8n^2\pi^2\hbar^4R^2}I(R)\left\{1 + \frac{G(R)}{I(R)}\cos\frac{2p_FR}{\hbar}\right\}, \tag{8}$$

where

$$I(R) = \int_{-\infty}^{\infty}\frac{d\beta}{\mathrm{ch}\,\beta}\exp\left(-\frac{2R\,\mathrm{ch}\,\beta}{\pi\xi_0}\right),$$
$$G(R) = \int_{-\infty}^{\infty}dt\,\mathrm{th}\,t\,\mathrm{sh}\,t\exp\left(-\frac{2R\,\mathrm{ch}\,t}{\pi\xi_0}\right), \tag{9}$$

$\xi_0 = \hbar v_F/\pi\lambda$ is the coherence length.

4. The function $I(R)$ differs little from a pure exponential:

$$I(R) \approx \pi\exp(-R/\xi_0).$$

The ratio G/I determines the behavior of $J(R)$. At small distances we have $R < R_0$ (R_0 is determined from the equation $G = I$) and $J(R)$ is an alternating-sign function of the distance. Numerical calculation yields $R_0 = 1.5\xi_0$.

The period of the oscillations $J(R)$, equal to $\pi\hbar/p_F$, is determined by the detuning from resonance, and the exponential decrease takes place over a length $\sim\xi_0$ that depends on the detuning and on the pump intensity.

We note that the Green's function of the quasiparticles in an impurity crystal takes the form (4) if $l > \xi_0$ (l is the electron mean free path). In the absence of nonmagnetic impurities $l \sim 1/cna^2$, where a is of the order of the interatomic distance.

Formation of spin glass is possible if the average distance between the impurity spins exceeds the period of the oscillations, i.e., subject to the inequality

$$l > R_0 > (3/4\pi cn)^{1/3} > \pi\hbar/p_F. \tag{10}$$

In the case of the inequality

$$l > (3/4\pi cn)^{1/3} > R_0 > \pi\hbar/p_F, \tag{11}$$

the impurities behave like a disordered ferromagnet.

Assuming the impurity density to be small enough,

$$(4\pi/3)^{1/3}/a^2(cn)^{2/3} \gg 1,$$

we ensure a wide parameter range satisfying (11) and (12). Since $R_0 \approx \hbar v_F/\pi\lambda$, the phase transition from spin glass to a disordered ferromagnet can be effected by changing the parameters of the external pump.

5. To estimate the coefficient preceding the curly bracket in (9) we choose the following parameters: $J \sim 1$ eV, $\hbar\omega - E_g \approx 0.1$ eV, $\lambda = 0.01$ eV, $m \sim m_0$, and $R_{av} = 30$ Å. In this case $J_{eff} \approx 0.1$ K.

The indirect interaction due to the pumping, J_{eff}, at the average distance between impurities, is of the same order as the estimated shift of T_C in a magnetic semiconductor.[13]

We note in conclusion that the problem considered in this paper was simplified by choosing for the semiconductor a symmetrical band scheme. Allowance for the noncongruence of the "electron" and "hole" Fermi surfaces of the quasiparticles makes for a more complicated $J_{eff}(R)$ dependence, since the system acquires two parameters with the dimension of length, which determine the periods of the oscillations.

In a real cyrstal, the results should also be modified by allowance for the recombination and heating of the

electrons relative to the lattice. This leads to a dependence of J_{eff} on the electron temperature and complicates the calculations in the problem, but does not affect the basic conclusions of the present analysis.

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