Instability of the homogeneous state in diluted magnetic semiconductors

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(Received 10 May 2010; revised manuscript received 28 June 2010; published 14 July 2010)

In diluted ferromagnetic semiconductors, with ferromagnetic exchange mediated by mobile nondegenerate carriers, the homogeneous susceptibility of the carriers rapidly falls as the spin polarization of the carriers increases inside the ferromagnetic phase. Therefore, the indirect exchange between the localized spins becomes a nonmonotonic function of the wave vector, and the ferromagnetic state is unstable against inhomogeneous fluctuations. It is shown that at a temperature close to the Curie temperature the ferromagnet enters into a modulated state, the modulation length being fixed by the Coulomb screening of the carriers. The inhomogeneous state is suppressed by an external magnetic field.

DOI: 10.1103/PhysRevB.82.033201 PACS number(s): 75.50.Pp

The current interest in diluted ferromagnetic semiconductors (DMS) is stimulated by their potential in spintronics application as well as by their unique physical properties (for reviews see Refs. 1 and 2). It is widely believed that free carriers play a crucial role in the ferromagnetic properties of these semiconductors. The free carrier mediated spin-spin coupling is long range, and therefore, different kinds of mean-field approximations were usually used to determine various thermodynamic properties of the DMS. This is, however, not always correct. It has been shown in several papers[3–5] that in metalliclike DMS with degenerate statistics of the carriers, the magnetically ordered state is unstable against inhomogeneous fluctuations on a nanometer scale, if the carrier concentration per one localized spin is smaller than some critical value.[4] Inhomogeneous ferromagnetism in (Ga,Man)As DMS on a length scale of several nanometers was observed[6] by muon spin rotation and relaxation studies. Although homogeneous above T_c, below T_c the DMS consists of ferromagnetic and paramagnetic regions of comparable volumes on both the metallic and nonmetallic sides of the metal-insulator transition.

Recently the properties of DMS with temperature-dependent carrier concentration were considered[7–9] within the molecular-field theory. The carrier mediated indirect exchange in this case is ferromagnetic and decreases exponentially with the spin separation.[10] The ferromagnetic phase exists if the carrier concentration does not fall too fast with the decrease of the temperature, and a reentrant transition into a paramagnetic state takes place at low temperatures, where the free-carrier concentration is too low to maintain ferromagnetism. It has been argued[8] that the exchange mediated by thermally excited carriers may be responsible for the high-temperature ferromagnetism observed in oxide based DMS.

In this paper, we show that, almost in the entire ferromagnetic phase, the carriers cannot support homogeneous ferromagnetism, since they become polarized in the ferromagnetic phase, and their homogeneous magnetic susceptibility rapidly decreases when moving away from T_c. Therefore, the carrier mediated exchange interaction becomes a nonmonotonic function of the wave vector, and the ferromagnet enters into a state with modulated magnetization, the modulation scale being of order of the Debye-Hükel screening length. Unlike the metal-like DMS, this happens at parametrically small $\tau \equiv 1 - T/T_c$. The inhomogeneity is characteristic only for carrier mediated exchange, and can, therefore, serve as a fingerprint of this exchange mechanism.

We start from the following Hamiltonian of the $s-d$ model

$$\mathcal{H} = \sum_{\mathbf{k},\sigma} \epsilon_{\mathbf{k}} c^{\dagger}_{\mathbf{k},\sigma} c_{\mathbf{k},\sigma} - 2J \sum_i \mathbf{S}_i \cdot \mathbf{s}_i + \mathcal{H}_C, \tag{1}$$

where $c^{\dagger}_{\mathbf{k},\sigma}(c_{\mathbf{k},\sigma})$ is the creation (annihilation) operator for a carrier with spin σ, $\epsilon_{\mathbf{k}} = \hbar^2 k^2/2m$ is the electron (hole) kinetic energy, $\mathbf{S}_i$ is the spin, localized at the lattice site i, $\mathbf{s}_i$ is the carrier spin, and $\mathcal{H}_C$ is the Coulomb interaction between the carriers given by:

$$\mathcal{H}_C = \frac{1}{2} \sum_{\mathbf{k}_1,\mathbf{k}_2,\mathbf{q}\neq 0,\sigma_1,\sigma_2} U(q) c^{\dagger}_{\mathbf{k}_1,\sigma_1} c^{\dagger}_{\mathbf{k}_2,\sigma_2} c_{\mathbf{k}_2+\mathbf{q},\sigma_2} c_{\mathbf{k}_1-\mathbf{q},\sigma_1}. \tag{2}$$

Here $U(q) = 4\pi e^2/q^2 \varepsilon v_0$, v_0 is the unit cell volume, and ε is the dielectric constant. The $q=0$ term is excluded from this sum, since it is compensated by the positive background.

In the molecular field approximation, the magnetization and the electron density are supposed to be homogeneous. Then the Coulomb interaction vanishes, and in the ferromagnetic state, when the electron polarization is finite, the Hamiltonian reads

$$\mathcal{H} = \sum_{\mathbf{k},\sigma} \epsilon_{\mathbf{k}\sigma} c^{\dagger}_{\mathbf{k},\sigma} c_{\mathbf{k},\sigma} - h \sum_i S^z_i. \tag{3}$$

Here $\epsilon_{\mathbf{k}\sigma} = \epsilon_{\mathbf{k}} \mp \Delta/2$, $\sigma = \uparrow, \downarrow$ labels the electrons with spin along the magnetization and opposite to it, $\Delta = 2Jx\langle S_z \rangle = 2Jxb(h/k_BT)$, $\langle S_z \rangle$ is the mean value of the localized spin, $b(z)$ is the Brillouin function:

$$b(z) = \left(S + \frac{1}{2}\right) \coth\left(S + \frac{1}{2}\right) z - \frac{1}{2} \coth \frac{z}{2}, \tag{4}$$

x is the concentration of magnetic impurities per unit cell, and $h(r) = J(n_\uparrow - n_\downarrow)$ is proportional to the molecular field acting on the localized spins, $n_\uparrow(n_\downarrow)$ being the number of carriers with spin up(down) per unit cell. We have for Boltzmann statistics of the carriers:

$$n_{\uparrow,\downarrow} = \frac{n(T)}{2}\left(1 \pm \tanh\frac{\Delta}{2k_BT}\right), \tag{5}$$

where $n(T)=n_\uparrow(T)+n_\downarrow(T)$ is the full carrier concentration, k_B is the Boltzmann constant.

Expanding $b(h/k_BT)$ and $n_\uparrow - n_\downarrow$ in powers of Δ, i.e., in powers of $\langle S_z\rangle$, one gets the well-known equation for the Curie temperature:

$$T_c = J[\alpha x n(T_c)]^{1/2}, \tag{6}$$

where $\alpha=S(S+1)/3$. As shown in Refs. 7 and 9 this equation has two solutions, if n does not fall to strong with the decrease of T. When lowering the temperature the system first undergoes a transition from the paramagnetic state into a ferromagnetic one at $T=T_c^+$. When the temperature is further lowered, a reentrant transition into the paramagnetic state takes place at a temperature T_c^-. Suppose that $n(T)$ has a simple activation form, $n=n_0\exp(-E/k_BT)$.[11] Then Eq. (6) has two solutions, when E is smaller than $2e^{-1}J\sqrt{\alpha x n_0}$, which is equivalent to $E<2T_c^+$. For larger E this equation has no solutions. In the following we consider the behavior of the system in the vicinity of T_c^+. For simplicity, we will denote this temperature as T_c. Assuming first that one may neglect the dependence of n on T at small τ, and solving the molecular field equations, one gets:

$$\langle S_z\rangle = \sqrt{6\alpha n\tau/x}, \quad \Delta = 2k_BT_c\sqrt{6\tau}, \tag{7}$$

Thus, at τ of order one, the ratio Δ/k_BT_c is larger than unity, i.e., the electrons are almost polarized. On the contrary, $\langle S_z\rangle/S$, is still small at such temperatures, since usually n is much smaller than x.

To account for the spatial fluctuations of the magnetic moment, we consider the correlation function of the longitudinal components of the localized spins. It can be written as[3]

$$K_{zz}(\mathbf{q}) = \frac{k_BTb'}{k_BT - b'J_{eff}(\mathbf{q})}, \tag{8}$$

Here $b'(z)=db/dz$, $J_{eff}(\mathbf{q})$ is the Fourier transform of the carrier mediated exchange between localized spins averaged over the random distribution of the impurity spins. It is proportional to the Fourier transform of the carrier susceptibility

$$J_{eff}(\mathbf{q}) = J^2x\chi(\mathbf{q}). \tag{9}$$

In the paramagnetic phase the susceptibility, χ_p, of electrons described by Eq. (1), is the same as for noninteracting electrons:

$$\chi_p(\mathbf{q}) = 2\sum_{\mathbf{p}}\frac{f(\mathbf{p}+\mathbf{q}) - f(\mathbf{p})}{\epsilon(\mathbf{p}) - \epsilon(\mathbf{p}+\mathbf{q})}, \tag{10}$$

where $f(\mathbf{p})$ is the Boltzmann distribution function.

We have at small q, $q \ll k_T=(2mk_BT)^{1/2}/\hbar$:

$$\chi_p(\mathbf{q}) = \frac{n}{k_BT}\left(1 - \frac{q^2}{6k_T^2}\right). \tag{11}$$

From Eqs. (8), (9), and (11) the usual Ornstein-Zernicke form of the correlation function follows:

$$K_{zz}(q) = \frac{\alpha}{2|\tau| + q^2/6k_T^2}, \tag{12}$$

with T_C given by Eq. (6).

The polarization of the carriers in the ferromagnetic phase drastically changes the behavior of the susceptibility. It was shown long ago[12,13] that in the random phase approximation the susceptibility of carriers with a finite degree of polarization depends crucially on the carrier-carrier Coulomb interaction as

$$\chi_f(\mathbf{q}) = \frac{\Pi_\uparrow(\mathbf{q}) + \Pi_\downarrow(\mathbf{q}) + 4U(q)\Pi_\uparrow(\mathbf{q})\Pi_\downarrow(\mathbf{q})}{1 + U(q)[\Pi_\uparrow(\mathbf{q}) + \Pi_\downarrow(\mathbf{q})]}. \tag{13}$$

Here Π_σ is given by Eq. (11), with the full concentration n replaced by n_σ. For non-polarized electrons with $\Pi_\uparrow=\Pi_\downarrow$, $U(q)$ cancels out, and Eq. (13) reduces to Eq. (11). One gets for small q

$$\chi_f(q) = \chi_f(0)(1-\gamma^2\tilde{q}^2)\frac{\tilde{q}^2\cosh^2(\Delta/2k_BT) + 1 - \gamma^2\tilde{q}^2}{1 + \tilde{q}^2(1-\gamma^2)}. \tag{14}$$

Here $\gamma^2=\kappa^2/6k_T^2$, κ is the inverse Debye-Hükel screening length, $\kappa^2=4\pi e^2 n/v_0\varepsilon k_BT$, $\tilde{q}=q/\kappa$, and the uniform susceptibility, $\chi_f(0)$, is equal to

$$\chi_f(0) = \frac{n_\uparrow n_\downarrow}{nk_BT} = \frac{n}{k_BT\cosh^2(\Delta/2k_BT)}. \tag{15}$$

The parameter γ is usually small for nondegenerate semiconductors.[14]

As shown above, the carriers become polarized at temperatures close to T_c, when τ is still smaller than unity. At such temperatures, $n_\downarrow$ and, hence, $\chi(0)$ are small. Therefore, the carrier-mediated exchange cannot maintain the uniform long-range order. Indeed, it follows from Eq. (14) that the susceptibility monotonically decreases with the increase of q only very close to T_c. At lower temperatures $\chi(q)$ reaches a maximum at $\tilde{q}$ given by:

$$\tilde{q}_m^2 = \frac{1}{\gamma}\tanh\frac{\Delta}{2k_BT} - 1. \tag{16}$$

The maximum susceptibility is:

$$\chi_f(q_m) = \frac{n}{k_BT}\left(1 + \gamma^2 - \gamma\tanh\frac{\Delta}{2k_BT}\right)\left(1 - \gamma\tanh\frac{\Delta}{2k_BT}\right), \tag{17}$$

and, unlike $\chi_f(0)$, it practically does not depend on the carrier polarization.

The wave-vector $\tilde{q}_m$ increases with the decrease of the temperature. When the electrons become almost polarized, $\tilde{q}_m$ reaches the value $1/\sqrt{\gamma}$. Equation (16) shows that at small γ the susceptibility monotonically decreases with increase in q only in the vicinity of T_c, when the inequality $\Delta/2k_BT_c < \gamma \ll 1$ holds.

One gets at small τ,

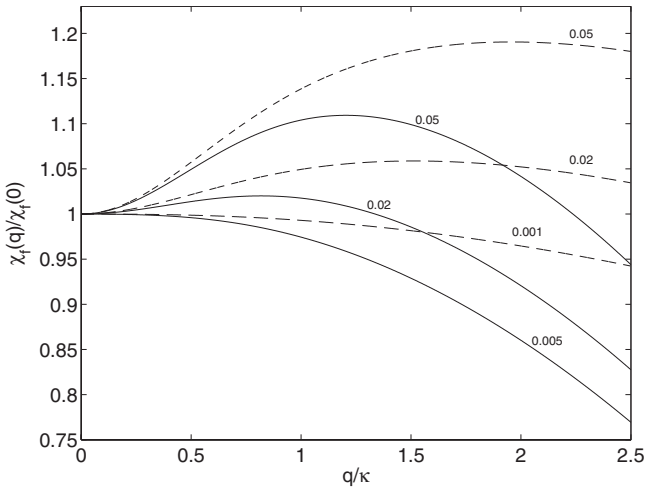

FIG. 1. The carrier susceptibility versus q in the ferromagnetic phase at different τ. Full lines—γ=0.2, broken lines—γ=0.1. The numbers near the lines show the values of τ.

$$\tilde{q}_m^2 = \frac{1}{\gamma}\sqrt{6\tau} - 1. \tag{18}$$

The dependence of χ on q at different γ and τ is illustrated in Fig. 1.

When τ is larger than $\gamma^2/6$, and the susceptibility is nonmonotonic, the correlation function can be written as

$$K_{zz} = \frac{\alpha(1+\tilde{q}^2)}{4\tau + \tilde{q}^2(\gamma^2 - 2\tau) + \gamma^4\tilde{q}^4} = \frac{\alpha\gamma^2(1+\tilde{q}^2)}{(\tau_+ - \tau)(\tau - \tau_-) + \gamma^4(\tilde{q}^2 - \tilde{q}_0^2)^2}. \tag{19}$$

Here $\tau_+ = 4.949\gamma^2$, $\tau_- = 0.0505\gamma^2$, and

$$\tilde{q}_0^2 = \frac{\tau}{\gamma^2} - \frac{1}{2}, \quad \tilde{q}_0(\tau_+) = \tilde{q}_m(\tau_+) = 2.11. \tag{20}$$

This is our main result. It shows that in the vicinity of T_c, at $\tau = \tau_+ \ll 1$, the uniform magnetic order becomes unstable, and the DMS undergoes a phase transition into a modulated state with a pitch wave vector q_0 of order of the inverse Debye-Hükel screening length. The result is robust. It follows from the basic properties of the susceptibility of polarized carriers and of the carrier-carrier correlation function. It should, therefore, also be qualitatively valid for more realistic models than the simple one considered in this paper.

It follows from the inequalities $n \ll x$, $\gamma \ll 1$, and $k_{T_C} = (2mT_c)^{1/2}/\hbar \ll (x/v_0)^{1/3}$ that the screening length, κ^{-1}, and, hence, the modulation length, q_0^{-1}, exceed the mean distance, $(v_0/x)^{1/3}$, between the magnetic impurities.

When one increases the temperature starting from the reentrant paramagnetic phase and one crosses T_c^-, one again enters into a ferromagnetic state with small but finite carrier polarization. It follows from the above that the homogeneous ferromagnetic state will only be stable at temperatures close to T_c^-. At higher T the system enters into a modulated state, which is stable until T reaches the value $T_c(1-\tau_+)$.

At $\tau = \tau_+$ the polarization of the carriers is weak. In contrast, as mentioned above, in metalliclike DMS the transition into the modulated state should happen at τ of order unity,[4] when the carriers are fully polarized. The reason for this difference between degenerate and non-degenerate carriers is related to the difference in the dependence of the carrier kinetic energy on the polarization degree. The kinetic energy of degenerate carriers increases with the increase of Δ. Therefore, the transition into the modulated state takes place, when the gain in magnetic energy of the localized spins caused by the local increase of $\langle S_z \rangle$ owing to the spatial redistribution of the carriers, overcomes the loss in the carriers kinetic energy. In DMS with non-degenerate carriers the carrier kinetic energy does not depend on Δ, and the transition is determined only by the temperature, where q_m reaches a value of order of the screening length.

If the dependence of n on the temperature is taken into account, the pitch vector q_0 is given by

$$\tilde{q}_0^2 = \frac{(1-\rho)}{2\gamma^2} + \frac{1}{2}\rho\cosh^2(\Delta/2K_BT) - 1, \tag{21}$$

where $\rho = (T^2/T_c^2)n(T_c)/n(T)$.

The condition $q_0^2 > 0$ in the vicinity of T_c leads to the inequality

$$\rho < 1 - \gamma^2. \tag{22}$$

Assuming that $n(T)$ is an exponential function, $n = n_0 \exp(-E/k_BT)$, one gets from Eq. (22) at small τ

$$\tau > \frac{\gamma^2 T_c}{2T_c - E}. \tag{23}$$

Thus, if E is not close to the critical value $2T_c$, when the ferromagnetic phase disappears, the transition into the ferromagnetic state is always accompanied by a transition into a modulated state at τ of order of γ^2.

Consider the effect of an external magnetic field, H, on the transition into the modulated state. It follows from the molecular field equations that in a low magnetic field, which satisfies the inequality

$$\mu H \ll k_B T_c \tau^{3/2}\sqrt{n/x\alpha}, \tag{24}$$

where μ is the magnetic moment of the localized spins, the spin gap in the electron spectrum is given by

$$\Delta = 2T_c\left(\sqrt{6\tau} + \frac{\eta}{4\tau}\sqrt{\frac{x\alpha}{n}}\right), \tag{25}$$

where $\eta = \mu H/k_BT_c$.

The above growth of Δ comes from the increase in the mean value of the localized spin and, hence, from the increase of the exchange field acting on the carriers, while the direct effect of the field on the carriers is smaller by a factor $k_BT_c/Jx = (n/x)^{1/2} \ll 1$. Inserting Eq. (25) into Eqs. (8), (14), and (15) we obtain that the field raises both τ_+ and q_0:

$$\tau_+(H) - \tau_+(0) = 0.112\frac{\eta}{\gamma}\sqrt{\frac{\alpha x}{n}}. \tag{26}$$

$$\tilde{q}_0(H) - \tilde{q}_0(0) = 0.0531 \frac{\eta}{\gamma^3} \sqrt{\frac{\alpha x}{n}}. \quad (27)$$

Thus, the field narrows the modulated state, and increases the stability region of the homogeneous ferromagnetic state.

In the present form, our model does not consider the effect of the direct exchange or superexchange between localized spins, which may be significant, if the magnetic impurity concentration is large. In Mn-doped III-V and II-VI DMS the superexchange is usually antiferromagnetic and suppresses the long-range ferromagnetic order. The direct exchange in the magnetic oxides is perhaps predominantly ferromagnetic,[7,17] and should, therefore, suppress the modulated state and stabilize the homogeneous ferromagnetic order. Thus, experimental investigation of the inhomogeneity of the ferromagnetic state can allow the discrimination between different exchange mechanisms. It would be interesting also to investigate the modulation effect in Mn-doped III-V DMS. It is clear that the modulation should happen only in DMS with free-carrier mediated ferromagnetism. Thus, observing the modulation may help to discriminate between the valence band and the impurity band mediated magnetism (see, e.g., the polaronic model[18]).

In conclusion, we showed that in a DMS, with ferromagnetism mediated by nondegenerate carriers, the homogeneous ferromagnetic state is only stable in the vicinity of the upper and lower transition temperatures. In the main part of the ferromagnetic phase a modulated state is stable. The modulation scale at the transition temperature into the modulated state is of order of the Debye-Hükel screening length.

This work was supported by the Israel Science Foundation (Grant No. 633/06).

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