A disordered RKKY lattice mean field theory for ferromagnetism in diluted magnetic semiconductors

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We develop a lattice mean field theory for ferromagnetic ordering in diluted magnetic semiconductors by taking into account the spatial fluctuations associated with random disorder in the magnetic impurity locations and the finite mean free path associated with low carrier mobilities. Assuming a carrier-mediated indirect RKKY exchange interaction among the magnetic impurities, we find substantial deviation from the extensively used continuum Zener model Weiss mean-field predictions. Our theory allows accurate analytic predictions for the magnetization curves, the suppressed low-temperature magnetization saturation, and the dependence of the magnetic properties on the carrier transport properties.

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Much of our current understanding of ferromagnetism in diluted magnetic semiconductors (DMS), notably in the extensively studied molecular beam epitaxy grown Mn-doped Ga$_{1-x}$Mn$_x$As (with the Mn doping level $x \approx 0.01 - 0.1$ system [1]), has been based on a very simple continuum Weiss mean field theory approximation [2] of the Zener model for the local (p-d) exchange coupling between the impurity magnetic moment $S = 5/2$ d-levels of Mn, and the itinerant carrier spin polarization, $s = 3/2$ holes of p-character in the valence band of GaAs. Spatial fluctuations associated with the random locations of Mn local moments are neglected in this continuum Weiss mean field theory (MFT). There is no theoretical rationale, except simplicity, underlying the neglect of the strong quenched spatial disorder due to the random magnetic impurity locations. Indeed, there have been several Monte Carlo simulations [3-7] attempting to include spatial disorder effects in the theory. These simulations as well as a recently developed percolation theory [8,9] have explicitly demonstrated the manifest importance of quenched disorder in DMS ferromagnetism, at least for the localized insulating DMS systems. There are also strong experimental signatures for the important interplay between disorder and magnetism in DMS materials.

In this Letter, we develop the first systematic theory for DMS ferromagnetism explicitly including spatial disorder effects in the successful Zener-RKKY mean field model [2,9]. Our theory is explicitly constructed for the metallic DMS systems with itinerant carriers (since we assume the carrier-mediated effective Mn-Mn indirect magnetic exchange interaction to be of free carrier mediated RKKY form), but the inclusion of a finite carrier mean free path in the theory (as we do in addition to the inclusion of Mn spatial disorder) allows us to make specific predictions about the dependence of the magnetic behavior of the system on the carrier transport properties. We also include a direct nearest-neighbor Mn-Mn antiferromagnetic exchange interaction in our disordered RKKY-mean field theory, which takes on significance for larger Mn concentrations. An associated essential feature of our model, which may be of crucial importance, is the fully discrete nature of our disordered mean field theory on a lattice. It is well-known that the discreteness associated with the specific lattice structure introduces unique features to the RKKY exchange interaction which are not captured in the corresponding continuum approximation. Our model, although conceptually simple, is actually quite rich as it depends on five independent physical parameters of the DMS system: the carrier-induced Mn-Mn RKKY coupling ($J_0$), the direct Mn-Mn (nearest-neighbor) antiferromagnetic exchange coupling ($J_{AF}^{AF}$), the local moment density ($n_i$), the free carrier density ($n_e$), and the carrier mean free path ($r_0$). In principle, the randomness in the Mn locations on the lattice could also be parameterized, particularly if clustering (or other spatial correlations) of Mn impurities is important during GaMnAs growth. We neglect at this stage any such correlation in Mn spatial locations (since no independent experimental information on the nature of quenched disorder is available), assuming the Mn atoms to be uniformly randomly distributed in the zinc-blende GaAs lattice at the Ga substitutional sites. (It would be easy to incorporate a more detailed description of disorder in the theory and to include Mn defects such as interstitials.) We also assume that the five parameters (i.e. $J_0$, $J_{AF}^{AF}$, $n_i$, $n_e$, $r_0$) of our theory are completely independent of each other, which is a theoretically and physically consistent approximation within our model since these parameters must be determined experimentally (or otherwise) and are independent parameters for our model.

Our effective Hamiltonian describes the Mn-Mn magnetic interaction between classical Heisenberg spins $S_i$ on a lattice:

$$
\mathcal{H} = \sum_{ij} J_{ij}^{RKKY} S_i \cdot S_j + \sum_{ij} J_{ij}^{AF} S_i \cdot S_j, \quad (1)
$$

where $S_i$ is the $i$th Mn local moment of spin $5/2$; $J_{ij}^{AF}$ is the direct antiferromagnetic exchange interaction between nearest-neighbor Mn spins, i.e. $J_{ij}^{AF} = 0$, unless
interaction: called p-d hybridization) between the holes and the Mn in GaMnAs). This indirect Mn-Mn exchange interaction arises from [1,2,9] the local Zener coupling (or the so-called p-d hybridization) between the holes and the Mn d-levels, which then leads to the effective Mn-Mn RKKY interaction:

\[ J_{ij}^{RKKY} \equiv J_0 r^{-4} \left[ \sin(2k_F r) - 2 k_F r \cos(2k_F r) \right] \tag{2} \]

where \( r = |\mathbf{r}_i - \mathbf{r}_j| \) is the spatial separation between the magnetically coupled Mn atoms; \( k_F \propto n_e^{1/3} \) is the Fermi wavevector corresponding to the carrier density \( n_e \), and the RKKY coupling strength \( J_0(>0) \), taken as a parameter in our theory, is related to the local Zener coupling \( J_{pd} \) between the Mn local moments and the hole spins, \( J_0 \propto m J_{pd} \), where \( m \) is the hole effective mass. Note that for low carrier densities the RKKY exchange interaction is mostly ferromagnetic except for large Mn-Mn separation \( (r \geq k_F^{-1}) \), and therefore, as long as \( n_c \ll n_e \), where \( n_c \) is the active density of Mn local moments (i.e. typical \( r \propto n_i^{-1/3} \)), the frustration effects associated with the oscillatory nature of RKKY exchange interaction are unimportant in the problem; distinguishing the DMS systems from random dilute metallic magnetic alloy spin glass systems (e.g. Cu-Mn), which are typically in the opposite limit of \( n_c \gg n_i \). It is important to emphasize that a continuum Weiss mean-field theory (which necessarily neglects all spatial fluctuation effects by averaging over the Mn positions), blindly applied to the Mn-Mn RKKY interaction, as was already done [10] a long time ago (and revived recently [2] in the context of DMS systems), always yields long-range ferromagnetic ordering of the Mn local moments for all values of \( n_c \) and \( n_i \), with a mean-field ferromagnetic transition temperature \( T_c^{MF} \propto J_0 n_i n_c^{1/3} \). This is obviously incorrect for larger values of \( n_c \) where ferromagnetism would eventually disappear [11] in a lattice model.

We include the collisional broadening due to the finite carrier mean free path \( (r_0) \) in the theory by incorporating an exponential suppression \( e^{-r/r_0} \) in Eq. (2), indicating that the RKKY interaction is cut off for \( r \gg r_0 \) due to resistive scattering effects. (This is important in view of the strong experimentally observed correlation between \( T_c \) and conductivity typically found in DMS systems.) This form for the collisional broadening induced suppression of the RKKY interaction is theoretically well-justified [12] for studying ferromagnetic ordering of the local moments. We have also considered thermal effects in the RKKY interaction by generalizing Eq. (2) to the corresponding finite temperature formula, but typically \( T_c \ll T_F \) for GaMnAs, where \( T_F \) (\( T_c \)) is the carrier Fermi temperature (Curie temperature), and therefore thermal broadening of the RKKY interaction is quantitatively and qualitatively unimportant in our calculations in contrast to the collisional broadening effects included in the \( e^{-r/r_0} \) suppression. Note that an important novel feature of our theory is the “integrating-out” of the free carrier variables as we consider an effective (disordered) local moment magnetic Hamiltonian defined by Eq. (1), with the free carrier information entering the theory only through the various physical parameters of the model, e.g. \( J_0, n_c, r_0, m, etc. \) This simplification is well-justified in studying DMS ferromagnetism since the long-range ferromagnetic ordering in the system arises entirely from the Mn local moments with the itinerant holes contributing little to ferromagnetism (by virtue of \( n_i \gg n_c \) and \( S_{Mn} \ll s_{hole} \)).

The Hamiltonian of Eq. (1) can be rewritten as a generalized Heisenberg model for Mn (classical) spins on a disordered GaAs lattice \( \mathcal{H} = \sum_{ij} J_{ij}(r) S_i \cdot S_j \) (where \( J_{ij} \equiv J_{ij}^{RKKY} + J_{ij}^{AF} \equiv J(r) \)) with the sum over \( i,j \) extending over (random) magnetic impurity locations in the fcc GaAs lattice. With no loss of generality we approximate the short-ranged antiferromagnetic interaction \( J_{ij}^{AF} \) by subsuming it in the definition of \( J_{ij}^{RKKY} \) or \( J(r) \) by multiplying it by a factor of 1/2 if the impurities occupy nearest-neighbor positions in the fcc lattice. This avoids introducing an additional unknown parameter \( J_{ij}^{AF} \), which could easily be introduced if such a need arises.

We use a lattice mean field theory and consider each impurity spin \( S_i \) to be immersed in an effective magnetic field, \( B_{eff}^{(i)} = \frac{g_\mu_B}{g_i} (J_i \langle \langle S_i^z \rangle \rangle) \), where \( J_i \equiv \sum_{ij} J_{ij} \) is the sum of all couplings to the impurities surrounding the site \( i \), and \( \langle \langle S_i^z \rangle \rangle \) is the thermally and site-averaged polarization, and \( g \) is the g-factor corresponding to the impurity. Clearly, \( J_i \) depends on the impurity configuration and \( B_{eff}^{(i)} \) is specific to the position \( R_i \). Given \( B_{eff}^{(i)} \), the thermally averaged spin polarization \( \langle S_i^z \rangle \) is \( (k_B = 1) \)

\[ \langle S_i^z \rangle = B_S \left( \mu_B B_{eff}^{(i)}/T \right) \]

where \( B_S \) is the usual mean field thermal Brillouin function. Although we calculate in mean field and use the average spin \( \langle \langle S_i^z \rangle \rangle \), we retain the site dependence in \( J_i \), thereby taking into account impurity disorder which subjects different Mn spins to different couplings depending on the local impurity configurations as determined by the random Mn locations on the GaAs lattice. \( \langle S_i^z \rangle \) is numerically calculated for a specific impurity distribution. To determine \( \langle \langle S_i^z \rangle \rangle \), the polarization averaged over all impurities, we integrate over all possible realizations of disorder, obtaining

\[ \langle \langle S_i^z \rangle \rangle = \int P(J) B_S \left( \langle \langle S_i^z \rangle \rangle J/T \right) dJ, \tag{3} \]

where \( P(J) \) is the probability distribution of \( J \). One then calculates \( \langle \langle S_i^z \rangle \rangle \) self-consistently from Eq. (3). \( P(J) \) is determined a priori via Monte Carlo sampling. We as-
sume that the Mn impurities occupy Ga lattice sites with uniform probability as determined by its concentration $x$ in Ga$_{1\_x}$Mn$_x$As.

In our lattice mean field theory the ferromagnetic transition temperature $T_c$ is determined by the site-averaged exchange coupling and is given by

$$T_c = \frac{35}{12} \sum_{i=1}^{\infty} N_i J(r_i),$$

(4)

where $N_i$ and $r_i$ are the numbers and distances of the $i$th nearest neighbors, respectively. We express $r_i$ and the remaining two length scales $r_0, (2k_f)^{-1}$ in units of the fcc unit cell length. The continuum limit is reached for $r_0, (2k_f)^{-1} \ll 1$ with

$$T_c^{\text{cont}} = (140\pi x/3) \int_0^\infty \pi^2 J(r) dr$$

(5)

$$= T_c^{\text{MFT}} \left[ 1 - \tan^{-1}(2k_F r_0)/(2k_F r_0) \right],$$

(6)

where $T_c^{\text{MFT}} \equiv (280\pi J_0 x/3)(3\pi^2 n_c/2)^{1/3} \propto J_0 x n_c^{1/3}$ is the continuum Weiss MFT value for $T_c$ which has been employed [2] extensively in the recent DMS literature (note that $S = 5/2$ here). The factor $f(2k_F r_0) \equiv [1 - \tan^{-1}(2k_F r_0)/(2k_F r_0)]$ takes into account the exponential cutoff imposed by the finite mean free path. We recover the continuum Weiss MFT result when $2k_F r_0 \gg 1$, i.e. in the strongly metallic regime. However, when $r_0$ becomes comparable to the length scale $(2k_f)^{-1}$; $f(x) \approx x^2 - \frac{1}{3} x^4 \cdots$, the RKKY interaction is effectively suppressed and $T_c$ is substantially lower than $T_c^{\text{MFT}}$ even in the continuum approximation. To obtain an accurate formula for $T_c$, it is necessary also to take into account the antiferromagnetic interaction between Mn impurities and to correct for the differences between the continuum approximation of Eq. (6) and the discrete lattice sum of Eq. (4). In the same way that an integral and a discrete approximation to that integral differ by a power series in the step size, the difference between the continuum and discrete formulae described above can be written as a series in $k_F$. With these improvements, one finds as a reasonable large $r_0$ approximation for $T_c$

$$T_c = T_c^{\text{MFT}} (1 + \alpha_2 n_c^{2/3} + \alpha_4 n_c^{4/3} + \alpha_6 n_c^2 + \cdots)
\times \left(1 - \beta_1 r_0^{-1}[n_c^{1/3}(1 - \beta_2 r_0^{-1})] + r_0^{-1} [\beta_3 - \beta_4 r_0^{-1}] \right)
\times f(2k_F r_0) - \frac{35}{2} \pi J(1/\sqrt{2});$$

(7)

$$\alpha_2 = -0.733256, \alpha_4 = 6.2594 \times 10^{-2},$$

$$\alpha_6 = 2.89 \times 10^{-3}, \alpha_8 = 3.7 \times 10^{-4};$$

$$\beta_1 = 0.1633, \beta_2 = 0.4284, \beta_3 = 0.5584, \beta_4 = 0.1176$$

(8)

where the first term in brackets is the exact nearest neighbor contribution while the second term contains the remaining couplings calculated in the continuum limit, and the optimal choice for $r_0$ is 0.953. For $r_0 \leq 1$ the relative error for $T_c$ from Eq. (9) is less than 2%. Evidently, the first term (accounting for the effect of impurities on neighboring sites) dominates as $r_0$ becomes comparable to the size of the GaAs unit cell. In this regime, the antiferromagnetic coupling removes half of $T_c$ (within our simplistic choice for $J^{\text{AF}}$) and thus plays a very important role for small mean free path. Obviously the result here depends strongly on the specific approximation one uses for $J^{\text{AF}}$ and would be stronger for stronger Mn-Mn AF coupling.

In Fig. 1(a) we show our direct numerical calculation of $T_c$ as a function of the carrier density $n_c$ (for fixed Mn doping level $x$) for four different values of the carrier mean free path varying from strongly metallic ($r_0 = 10$) to “almost” insulating ($r_0 = .5$). Clearly, for most choices of $n_c$ and $r_0$, $T_c^{\text{MFT}}$ is a poor approximation for $T_c$ in the disordered lattice system. We emphasize that the strong dependence of our calculated $T_c$ on the system conductivity (through $r_0$) is consistent with GaMnAs experimental results where increasing conductivity is found to enhance $T_c$. In Fig 1(b) we show our calculated spontaneous magnetization $M(T)$ results, which depend on the full exchange distribution $P(J)$. The $M(T)$ profile (convex, concave, or linear) depends on whether the system is in the insulating (small $r_0$, small $n_c$) or metallic (large $r_0$, large $n_c$) regime. Concavity in $M(T)$ is a signature of an insulating system, while convex profiles appear deep in the metallic regime [9]. For intermediate impurity densities and mean free paths, it is possible to obtain a linear magnetization curve. Within the framework of our model, we are able to span both the localized and metallic regimes by having very small and large values of $r_0$, respectively, and we find both types of behavior in the magnetization profiles as can be seen in Fig. 1(b).

There is a strong correlation between the degree of concavity in $M(T)$ and the extent to which the coupling probability distribution $P(J)$ has a strong weight near zero interaction strength and a multimodal profile. The multimodal $P(J)$ in general leads to a concave $M(T)$. A reasonable measure of the concavity is $\gamma \equiv \int_0^\infty M''(T) dT$, or the difference in the slopes of $M(T)$. The sign of $\gamma$ indicates whether $M(T)$ is convex (negative $\gamma$), concave (positive $\gamma$), or linear (if $\gamma \approx 0$). The temperatures $t_1$ and $t_2$ are chosen to capture an intermediate temperature range, neither very close to $T_c$ nor to zero. To go beyond the influence of the low temperature behavior, we set
FIG. 1. (a) $T_c$ relative to continuum $T_c^{MF}$ for carrier mean free path $r_0 = 10, 2, 0.9,$ and $0.5$ Solid curves are plotted with the antiferromagnetic interaction taken into account, while dashed curves depict $T_c$ with antiferromagnetism suppressed. (b) Magnetization profiles corresponding to $r_0 = 10$ ($x = 0.05$), $r_0 = 0.9$ ($x = 0.01$), $r_0 = 0.5$ ($x = 0.015$); $n_c/n_i = 0.3$ for all $r_0$ values

$t_1 = .1t_c$; to avoid the critical region, we use $t_2 = .7t_c$.

This result is a rough criterion for concavity, since the sign of $\gamma$ indicates whether the profile is convex, concave, or linear. In Fig. 2 we give results for our “magnetization phase diagram” where the regimes of convex/concave magnetization behavior are depicted on the $n_c/n_i$-$x$ two dimensional plots. Contour plots in Fig. 2 show in a concise way important trends; one sees very clearly the transition from some concavity in $M(T)$ to convex $M(T)$ behavior with increasing $r_0$. Raising impurity density also tends to make the magnetization profile more convex.

We have also calculated the saturation magnetization $M_0 \equiv M(T \to 0)$ using our theory, finding that consistent with experimental observations $M_0$ could indeed be less than unity particularly for larger values of relative carrier density ($n_c/n_i$) and/or for more metallic systems (i.e. larger values of $r_0$). The main suppression mechanisms are direct AF coupling between nearest-neighbor Mn-Mn interaction (increasing with $J^{AF}$ and $x$) and the oscillatory nature of the RKKY exchange coupling at large values of $kF$ (increasing with $n_c/n_i$).

FIG. 2. Magnetization phase diagram: contour plots of the concavity parameter $\gamma$ as a function of Mn concentration $x$ and $n_c/n_i$ for two values of carrier mean free path (a) $r_0 = .5$ and (b) $r_0 = 2$, where $n_c$ ($n_i$) are the carrier (local moment) densities, and $x$ is the Mn doping level. The $\gamma = 0$ contour divides the concave/convex phases in (a).

To summarize, we have developed a disordered lattice mean field theory for DMS ferromagnetism which incorporates spatial fluctuations associated with random lattice locations of the impurity moments, the finite carrier mean free path, and the Mn-Mn nearest-neighbor antiferromagnetic coupling. We calculate $T_c$ and magnetization curves for Ga$_{1-x}$Mn$_x$As ferromagnetic semiconductors, finding that all magnetic properties, including $T_c$, depart significantly from the predictions of the continuum Weiss mean-field theory. A particularly salient feature of our results is the strong theoretical correlation between $T_c$ and the metallicity of the system (i.e. $r_0$) as observed experimentally. It should be feasible to incorporate spin-orbit coupling and detailed valence band structure in the theory by generalizing our simple single valence band RKKY model. The most important essential approximation of our model is the assumption of the RKKY form for the carrier-mediated indirect exchange interaction between the impurity local moments. Recent first principles band structure calculations [13] show that the effective DMS interaction between impurity moments is indeed of the RKKY form. There have also been recent numerical calculations explicitly establishing [14] the validity of RKKY coupling in disordered semiconductors.
Our theory is computationally relatively efficient allowing us to obtain DMS magnetic properties as a function of five independent physical parameters \((J_0, J^AF, n_c, n_i, r_0)\). We believe that the lattice mean field approximation is fairly sound by virtue of \(n_i \gg n_c\) in the DMS systems (and because of the long range nature of the ferromagnetic RKKY interaction).

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