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 T_c and provides simple explanations for a number of observed anomalies, including the non-Brillouin function magnetization curves, the suppressed low-temperature magnetization saturation, and the dependence of T_c on conductivity.

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Much of our current understanding of ferromagnetism in diluted magnetic semiconductors (DMS), most notably in the extensively studied molecular beam epitaxy grown Mn-doped $Ga_{1-x}Mn_xAs$ (with the Mn doping level $x \approx 0.01$–0.1 system), has been based on a simple continuum Weiss mean field theory (MFT) approximation [1] 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. 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 [2–8] attempting to include spatial disorder effects in the theory. These simulations as well as a recently developed percolation theory [9,10] 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 [11].

In this Letter, we develop the first systematic theory for DMS ferromagnetism explicitly including spatial disorder effects in the Zener-RKKY mean field model [1,10] appropriate for the metallic DMS systems. Our framework is a thermal mean field theory, a reasonable approximation to make due to the large coordination number (12) of the zinc-blende lattice and the long-range of the indirect exchange interaction in the metallic situation. Our theory is explicitly constructed for metallic DMS systems with itinerant carriers (since we assume the carrier-mediated effective Mn-Mn indirect magnetic exchange interaction to be of the RKKY form), but the inclusion of a finite carrier mean free path in the theory allows us to make specific predictions about the dependence of the magnetic behavior of the system on the carrier transport properties. An alternative (and more accurate statement) is that we include an exponential cutoff in the range of the RKKY interaction, allowing us to obtain results while smoothly interpolating between long-range and short-range magnetic interactions simply by varying the cutoff parameter r_0, which should be related to the "localization length" of the DMS carriers. We also include a direct nearest-neighbor Mn-Mn antiferromagnetic exchange interaction in our theory, which takes on significance for larger Mn concentrations or in the presence of Mn interstitial defects. An essential feature of our model is the fully discrete nature of our disordered mean field theory on a lattice; the discreteness associated with the specific lattice structure introduces unique features to the RKKY exchange interaction which are not caught 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^{AFM}), the effective local moment density (n_i), the free carrier density (n_c), and the exponential cutoff length scale (r_0) directly related to the carrier mean free path (localization length). 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.

Our effective Hamiltonian describes the Mn-Mn magnetic interaction between classical Heisenberg spins $\mathbf{S}_i$ on a lattice:

$$\mathcal{H} = \sum_{ij} J_{ij}^{\mathrm{RKKY}} \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{ij} J_{ij}^{\mathrm{AFM}} \mathbf{S}_i \cdot \mathbf{S}_j, \qquad (1)$$

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where $\mathbf{S}_i$ is the ith Mn local moment of spin 5/2; J_{ij}^{AFM} is the direct antiferromagnetic exchange interaction between nearest-neighbor Mn spins, i.e., $J_{ij}^{\mathrm{AFM}} = 0$, unless i and j are nearest neighbors. The first term in the effective Hamiltonian, the carrier-mediated RKKY indirect exchange interaction responsible for producing DMS ferromagnetism, describes the magnetic interaction between the Mn local moments induced by the free carrier spin polarization. This indirect Mn-Mn exchange interaction arises from [1,10] 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:

$$J_{ij}^{\mathrm{RKKY}} \equiv J_0 r^{-4}[\sin(2k_F r) - 2k_F r\cos(2k_F r)] \qquad (2)$$

where $r = |\mathbf{R}_i - \mathbf{R}_j|$ is the spatial separation between the magnetically coupled Mn atoms; $k_F \propto n_c^{1/3}$ is the Fermi wave vector corresponding to the carrier density n_c, 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, $J_0 \propto mJ_{pd}^2$, 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 $n_c \ll n_i$, where n_i 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 [12] a long time ago (and revived recently [1] 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 $T_c^{\mathrm{MFT}} \propto J_0 n_i n_c^{1/3}$. This is obviously incorrect for larger values of n_c where ferromagnetism would eventually disappear [13] in a lattice model.

The Hamiltonian of Eq. (1) can be rewritten as a generalized random exchange Heisenberg model for random Mn spins on a disordered GaAs lattice $\mathcal{H} = \sum_{ij} J_{ij}(r)\mathbf{S}_i \cdot \mathbf{S}_j$ [where $J_{ij} \equiv J_{ij}^{\mathrm{RKKY}} + J_{ij}^{\mathrm{AFM}} \equiv J(r)$] with the sum over i,j extending over (random) magnetic impurity locations in the GaAs lattice. For notational brevity, we subsume the short-ranged antiferromagnetic interaction J_{ij}^{AFM} in the definition of $J(r)$.

We use a lattice mean field theory and consider each impurity spin S_i to be immersed in an effective magnetic field, $B_{\mathrm{eff}}^{(i)} = \frac{1}{g\mu_B}(J_i\langle\langle S^z\rangle\rangle)$, where $J_i \equiv \sum_j J_{ij}$ is the sum of all couplings to the impurities surrounding the site i, $\langle\langle S^z\rangle\rangle$ is the thermal and site-averaged polarization, and g is the g factor corresponding to the impurity. Given $B_{\mathrm{eff}}^{(i)}$, the thermally averaged spin polarization $\langle S_i^z\rangle$ is ($k_B = 1$) $\langle S_i^z\rangle = SB_S[g\mu_B B_{\mathrm{eff}}^{(i)}/T]$, where B_S is the usual mean field thermal Brillouin function. We retain the exact 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 $\langle\langle S^z\rangle\rangle$, the polarization averaged over all impurities, we integrate over all possible realizations of disorder, obtaining

$$\langle\langle S^z\rangle\rangle = \int P(J)SB_S(\langle\langle S^z\rangle\rangle J/T)dJ, \qquad (3)$$

where $P(J)$ is the probability distribution of J. One then calculates $\langle\langle S^z\rangle\rangle$ self-consistently from Eq. (3). $P(J)$ is determined *a priori* via Monte Carlo sampling.

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

$$T_c = \frac{35}{12}x\sum_{i=1}^{\infty} N_i J(r_i), \qquad (4)$$

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

$$T_c^{\mathrm{cont}} = \frac{140\pi x}{3}\int_0^{\infty} r^2 J(r) = T_c^{\mathrm{MFT}} f(2k_F r_0), \qquad (5)$$

where $T_c^{\mathrm{MFT}} \equiv (280\pi J_0 x/3)(3\pi^2 n_c/2)^{1/3} \propto J_0 x n_c^{1/3}$ is the continuum MFT value for T_c, which has been employed [1] 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 on the magnetic interaction. We recover the continuum Weiss MFT result in Eq. (5) when $2k_F r_0 \gg 1$, i.e., in the strongly metallic regime. However, when r_0 becomes comparable to the length scale $(2k_F)^{-1}$; $f(x) \approx \frac{1}{3}x^2 - \frac{1}{5}x^4 \cdots$, the RKKY interaction is effectively suppressed and T_c is substantially lower than T_c^{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. (5) 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 formula 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

$$T_c = T_c^{\mathrm{MFT}}(1 + \alpha_2 n_c^{2/3} + \alpha_4 n_c^{4/3} + \alpha_6 n_c^2 + \cdots) \times \left\{1 - \beta_1 r_0^{-1}[n_c^{1/3}(1 - \beta_2 r_0^{-1})] + r_0^{-1}[\beta_3 - \beta_4 r_0^{-1}]\right\} f(2k_F r_0) - \frac{35}{2} x J(1/\sqrt{2}), \quad (6)$$

where $\alpha_2 = -0.733\,256$, $\alpha_4 = 6.2594 \times 10^{-2}$, $\alpha_6 = 2.89 \times 10^{-3}$, $\alpha_8 = 3.7 \times 10^{-4}$, $\beta_1 = 0.1633$, $\beta_2 = 0.4284$, $\beta_3 = 0.5584$, $\beta_4 = 0.1176$. T_c in Eq. (6) is correct to better than one part in 10^6 for large r_0 (as compared with our numerical calculations). Accuracy diminishes as r_0 is made smaller, though the formula given in Eq. (6) is still correct to within 1% for $r_0 = 1$. One can also arrive at a formula to cover the small r_0 regime. In this case, T_c can be expressed as the sum of two terms

$$T_c = \frac{35}{12 k_B} x \left[6J(1/\sqrt{2}) + 16\pi \int_{r_a}^{\infty} J(r) r^2 dr \right], \quad (7)$$

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_a is 0.953. For $r_0 \leq 1$ the relative error for T_c from Eq. (7) 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 plays an important role for small r_0, suppressing T_c substantially depending on the precise value of J^{AFM}.

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 ($r_0 = 0.5$). Clearly, for most choices of n_c and r_0, T_c^{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 [14]. In Fig. 1(b) we show our calculated spontaneous magnetization $M(T)$ results, which depend on the full exchange distribution $P(J)$ shown in the lower part of Fig. 1(b). 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 [10]. 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, 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 $\gamma \equiv \int_{t1}^{t2} M''(T) dT$, or the difference in the slopes of $M(T)$. The sign of γ indicates whether $M(T)$ is convex (negative γ), concave (positive γ), 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. We choose $t_1 = 0.1 t_c$ and $t_2 = 0.7 t_c$. 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

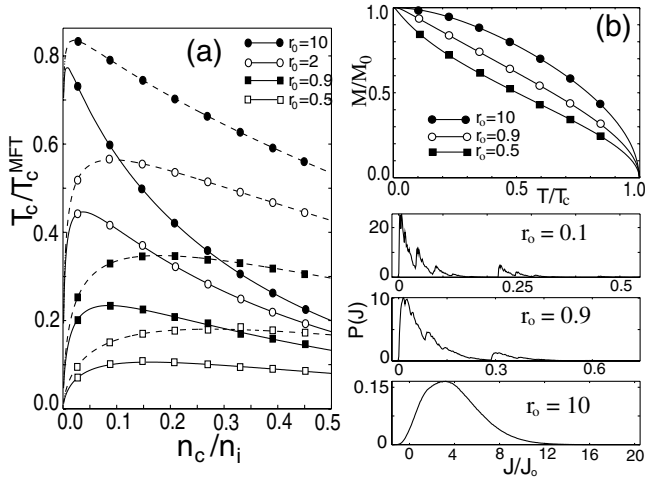

FIG. 1. (a) T_c relative to continuum Weiss T_c^{MFT} for carrier mean free path $r_0 = 10$, 2, 0.9, and 0.5. Solid curves are plotted with the antiferromagnetic interaction taken into account, while dashed curves depict T_c with antiferromagnetism suppressed. (b) Magnetization profiles corresponding 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. Below, we show the exchange coupling distributions $P(J)$ corresponding to the magnetization curves.

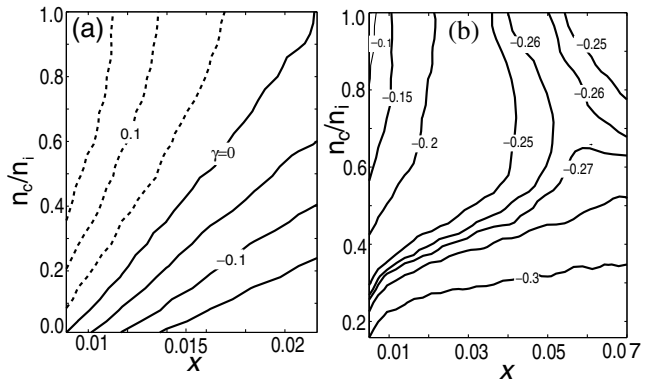

FIG. 2. Magnetization phase diagram: contour plots of the concavity parameter γ as a function of Mn concentration x and n_c/n_i for two values of carrier mean free path (a) $r_0 = 0.5$ and (b) $r_0 = 2$, where n_c (n_i) are the carrier (local moment) densities, and x is the Mn doping level. The $\gamma = 0$ contour divides the concave/convex phases in (a).

very clearly the transition from some concavity in $M(T)$ to convex $M(T)$ behavior with increasing r_0. Raising the magnetic impurity density also tends to make the magnetization profile more convex.

We have also calculated the saturation magnetization $M_S \equiv M(T \to 0)$ using our theory, finding that consistent with experimental observations [14] M_S/M_0 (where M_0 is the magnetization with all of the Mn s pins fully aligned) 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 M coupling between nearest-neighbor Mn-Mn interaction (increasing with J^{AFM} and x) and the oscillatory nature of the RKKY exchange coupling at large values of $k_F r$ (increasing with r_0 and n_c/n_i). The effect of the antiferromagnetic part of the RKKY coupling can be seen in Fig. 3. The broken line, corresponding to $r_0 = 1$ and $x = 0.01$, shows less suppression than the solid $r_0 = 5$, $x = 0.05$ curve, since larger r_0 correspond to a longer ranged RKKY coupling, thereby increasing the number of spins interacting antiferromagnetically; a higher doping fraction, $x = 0.05$, means a greater number of impurities involved in the AFM coupling. Both of the M_S curves decrease monotonically with n_c/n_i. Inset (a) of Fig. 3 shows the effect of increasing the fraction of Mn atoms in interstitial sites, y with n_c/n_i fixed. In addition to interacting with the Mn in Ga sites via a strong direct AFM exchange, the interstitial impurities do not contribute to the overall magnetization, adding to the suppression of M_S. The contribution of the spin-3/2 holes (not included in the results of Fig. 3), whose polarization is opposite that of the impurities, makes the suppression of M_S even more significant; the total saturation magnetization is $M_S^{\mathrm{total}} = M_S^{\mathrm{Mn}}[1 - (g_h/g_{\mathrm{Mn}})(n_c/n_i)(\langle s\rangle/\langle S\rangle)]$, where g_h and g_{Mn} are the g factors for the holes and Mn ions, respectively. In inset (b), we take into account the fact that the Mn interstitial dopants, being electron donors, will reduce the hole density. In this simple model for compensation, we assume that each Ga-substituted Mn absorbs exactly one electron, contributing a hole, while each interstitial Mn donates exactly two electrons, eliminating two carriers. Since interstitial Mn each neutralize two carriers, one has for the carrier density $n_c/n_i = (1 - 3y)$. Our explanation for the lack of full saturation is manifestly extrinsic, i.e., subject to variation from experiment to experiment depending on the Mn interstitial density in the sample; one should be able to improve M_S by appropriately annealing away the Mn interstitial density.

We have developed a disordered lattice mean field theory for DMS ferromagnetism that 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

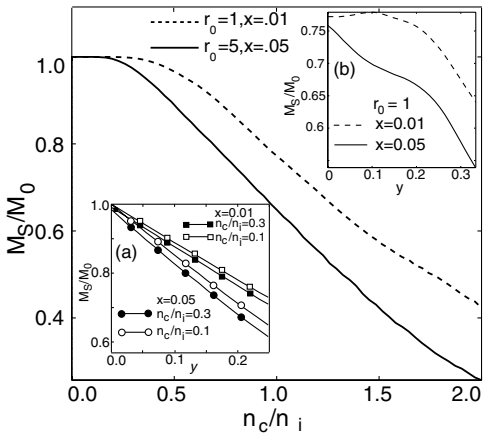

FIG. 3. Suppression of the saturation magnetization M_S due to the oscillatory nature of the RKKY exchange interaction. Inset (a) shows the suppression by direct AFM coupling to Mn dopants in interstitial sites as a function of the fraction of interstitial impurities (y) with n_c/n_i held fixed. In inset (b), M_S is given as a function of y with the carrier density modeled as $n_c/n_i = (1 - 3y)$.

$Ga_{1-x}Mn_xAs$ ferromagnetic semiconductors, finding that all magnetic properties, including T_c, depart significantly from the predictions of the extensively used 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. We establish that the observed lack of DMS saturation magnetization may be entirely extrinsic, arising from the Mn interstitial defects. The most important, essential approximation of our model is the assumption of an effective RKKY form for the carrier-mediated indirect exchange interaction between the impurity local moments, which is supported by recent numerical calculations explicitly establishing [15] the validity of RKKY coupling in disordered semiconductors.

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