Phase Diagram of the Disordered RKKY Model in Dilute Magnetic Semiconductors

D. J. Priour, Jr. and S. Das Sarma

Condensed Matter Theory Center, Department of Physics, University of Maryland, College Park, Maryland 20742-4111, USA

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We consider ferromagnetism in spatially randomly located magnetic moments, as in a diluted magnetic semiconductor, coupled via the carrier-mediated indirect exchange RKKY interaction. We obtain, via Monte Carlo calculations, the magnetic phase diagram as a function of the impurity moment density n_i and the relative carrier concentration n_c/n_i. As evidenced by the diverging correlation length and magnetic susceptibility, the boundary between ferromagnetic and nonferromagnetic phases constitutes a line of zero temperature critical points which can be viewed as a magnetic percolation transition. In the dilute limit, we find that bulk ferromagnetism vanishes for $n_c/n_i > 0.1$. We also incorporate the local antiferromagnetic direct superexchange interaction between nearest neighbor impurities and examine the impact of a damping factor in the RKKY range function.

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There has been substantial recent interest in the old problem of long-range ferromagnetic ordering in localized "impurity" magnetic moments induced by indirect exchange ("RKKY") interaction mediated through (effectively) "free" carriers (either electrons or holes). This interest arises from the context of diluted magnetic semiconductors (DMS), such as $Ga_{1-x}Mn_xAs$ with $x \approx 0.1$, where the Mn dopants act both as the impurity magnetic moments and as acceptors producing the carriers (i.e., holes for GaMnAs) mediating the RKKY coupling. The standard model for DMS ferromagnetism has been the carrier-mediated RKKY interaction, and understanding the RKKY magnetic phase diagram, therefore, takes on particular significance. This is important in view of the diluted and the random nature of the spatial distribution of the Mn dopants which could lead to substantial frustration in the magnetic interaction between the impurity moments due to the oscillatory nature of the RKKY coupling. The latter yield substantial antiferromagnetic (AF) couplings which have the potential to disrupt ferromagnetism. Our goal in this Letter is to obtain the RKKY magnetic phase diagram in a disordered DMS system via direct Monte Carlo simulations.

The complex interplay of the long-ranged oscillatory behavior of the RKKY interaction and strong disorder makes simple theoretical statements difficult, and it is not obvious *a priori* whether in the dilute limit the ferromagnetic state is supported for any choice of system parameters even at $T = 0$. Mean field treatments such as the Curie-Weiss continuum mean field theory (MFT) are problematic, because they fail to take into account the discrete crystal lattice and thermal fluctuations and assume a ferromagnetic ground state without providing any means of assessing the validity of this assumption. In this Letter, we rigorously take into account positional disorder and we obtain the true ground state spin configurations, finding that the RKKY model *does* support a ferromagnetic phase, albeit only for a limited parameter range. In addition, we demonstrate via direct Monte Carlo calculations that the transition from the nonferromagnetic (NF) to the ferromagnetic (FM) phase is marked by the percolation of magnetic clusters which grow in size as AF couplings are reduced and ultimately coalesce to yield long-range ferromagnetic order; this constitutes zero temperature percolation critical behavior. We neglect all quantum fluctuations, but our interest being the interplay of disorder and magnetic interaction, our results should in general be valid with respect to the existence (or not) of the FM phase.

Although the specific calculations described in this work are motivated by possible carrier-mediated RKKY ferromagnetism in DMS systems, our results also apply more broadly to a variety of disordered magnets with competing interactions (e.g., the Edwards-Anderson model which we have also examined and found similar behavior) where the NF to FM transition at ($T = 0$) occurs via magnetic percolation. Note that we do not attempt to characterize the NF phase (which may be a simple paramagnet or a subtle glassy phase).

Our model physical system is $Ga_{1-x}Mn_xAs$ (by far the most studied DMS system) where $x_i \approx 0.01$–0.1, the concentration of Mn dopants, is in the dilute limit. We assume that Mn impurities only occupy Ga sites in the zinc-blende GaAs (fcc) lattice with a lattice constant a. The large spin ($S = 5/2$) of the Mn moments permits a classical treatment of the spins, which we regard as Heisenberg spins governed by the Hamiltonian $\mathcal{H} = \sum_{ij} J(r_{ij})\mathbf{S}_i \cdot \mathbf{S}_j + \sum_{\langle ij\rangle} J^{\mathrm{AF}}\mathbf{S}_i \cdot \mathbf{S}_j$, where r_{ij} is the separation between moments i and j. In $\mathcal{H}$, $J(r) = J_0 e^{-r/l} r^{-4}[\sin(2k_{\mathrm{F}}r) - 2k_{\mathrm{F}}r\cos(2k_{\mathrm{F}}r)]$ is the RKKY range function, where $k_{\mathrm{F}} = (\frac{3}{2}\pi^2 n_c)^{1/3}$ is the Fermi wave number and n_c is the hole density. $J_0(>0)$ is related to the local Zener coupling J_{pd} between the Mn local moments and the hole spins. In particular, we have $J_0 \propto mJ_{pd}^2$ with m being the hole effective mass and the $S^2 = (5/2)^2$ factor being absorbed into J_0. J^{AF} is the local antiferromagnetic superexchange coupling relevant only for neighboring impurities. With DMS materials being at best poor metals, we also introduce in the RKKY range function a damping factor $e^{-r/l}$ where the damping scale l is related to the carrier mean free path or perhaps a carrier localization length arising from Anderson localization of the free carriers; however, we initially set $l = \infty$ in order to study the long-ranged RKKY coupling. For the latter, an important subtle question is whether the full RKKY model, with its long-ranged oscillatory behavior, supports a ferromagnetic ground state for strongly disordered systems. We note that very large values of J_0 (which tend to localize the hole carriers and hamper indirect exchange [1]) are also deleterious to ferromagnetism and lower T_c, an effect not examined in this Letter. We also ignore all band structure effects, which should be adequate for qualitative purposes. Our goal here is to obtain explicitly the ideal RKKY phase diagram, rather than calculate results for a specific material or compare with experiments.

Salient length scales include the mean spacing between impurities, $l_s \equiv n_i^{-1/3}$, and the scale of the oscillations in the RKKY range function, k_F^{-1}. Hence $k_F l_s$ determines the relative importance of FM and AF couplings; for $k_F l_s \sim 1$ RKKY oscillations tend to disrupt ferromagnetic order, while one expects ferromagnetic interactions to dominate for $k_F l_s \ll 1$. Since $k_F l_s \propto (n_c/n_i)^{1/3}$, and due to its experimental relevance, n_c/n_i is a useful parameter of merit for gauging the importance of AF interactions. While we concentrate on FM ordering for the $n_c/n_i \ll 1$ regime, other work has examined the $n_c/n_i \gg 1$ limit, deep in the NF phase [2,3]. We will see that, although a stable ferromagnetic phase is supported by the pure RKKY model, it occurs only for a relatively narrow n_c/n_i domain. Previous Monte Carlo calculations [4–6] have examined on a qualitative basis the impact of competing interactions on the ferromagnetic state. We show explicitly that the transition of the FM to the NF phase at $T = 0$ is a disorder driven critical transition involving magnetic percolation.

In the presence of strong disorder, n_c/n_i has a role very similar to temperature ($k_B T/J_0$); just as thermal fluctuations at higher temperatures disrupt ferromagnetic order by flipping spins, a larger n_c/n_i is associated with strong AF interactions which prevent many pairs of spins from aligning. For large enough n_c/n_i, only spins in close proximity are ferromagnetically correlated. As n_c/n_i is decreased, these small correlated groups of spins increase in size. Ultimately, the magnetic clusters span the entire system and magnetic percolation occurs, signaling the appearance of long-range ferromagnetic order. This is the extended free-carrier analog of the bound magnetic polaron percolation ferromagnetic transition recently discussed [7] in the context of strongly localized DMS materials. To identify magnetic percolation and thereby locate the NF/FM phase boundary, we determine the typical size of the magnetic clusters using a standard technique to calculate the ferromagnetic correlation length ξ [8]. Magnetic percolation and concomitant long-range ferromagnetism occurs when ξ becomes comparable to the system size L.

In our Monte Carlo calculations, we average over at least 500 disorder realizations. We obtain ground state configurations via simulated annealing, and thermal fluctuations are provided by the heat bath technique [9]. To exploit finite size scaling, we examine the behavior of the normalized correlation length ξ/L as a function of L. In the NF phase, ξ/L diminishes as L is increased, while for ferromagnetic order, ξ/L increases with increasing L. One seeks the FM/NF phase boundary where ξ/L is constant in L for the critical value of n_c/n_i. To minimize finite size effects, we examine systems where the mean number of spins $\langle N \rangle$ is $\gtrsim 1000$. To calculate T_c, we also examine ξ/L, but we vary the temperature T instead of n_c/n_i and again we seek the critical ξ/L curve.

In Fig. 1, for a large system with $\langle N \rangle \sim 1000$, we display calculated characteristics which would be accessible in experiment. Panel (a) of Fig. 1 is a graph of the ferromagnetic order parameter $m = [\langle |\mathbf{S}| \rangle]$ (square brackets indicate

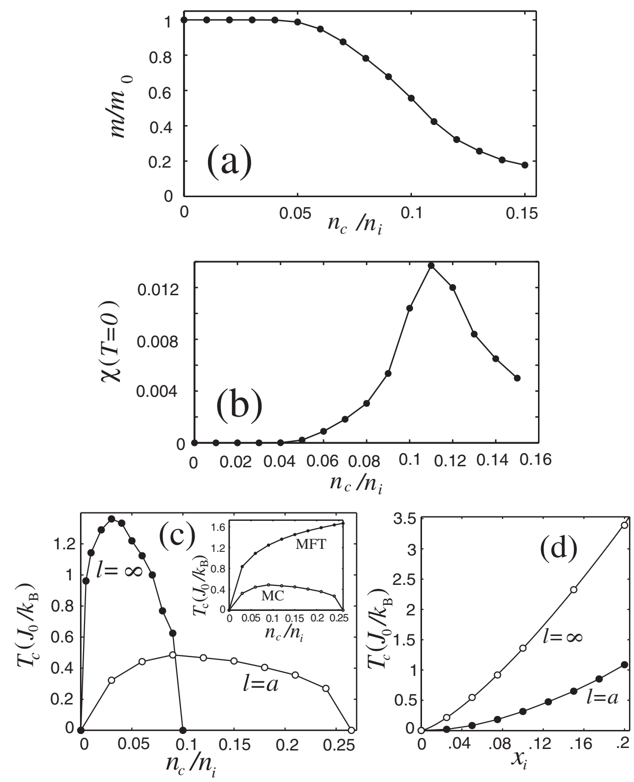

FIG. 1. $T = 0$ magnetization and susceptibility, and T_c plots vs n_c/n_i for $x_i = 0.1$; results are shown for the undamped RKKY with no AF superexchange coupling. Panels (a) and (b) display the zero temperature magnetization and susceptibility, respectively, for $\langle N \rangle = 1000$. Panel (c) depicts Curie temperatures vs n_c/n_i. The solid circles correspond to the undamped ($l = \infty$) case and the open circles to the damped ($l = a$) RKKY model. The inset of (c) shows MFT and MC T_c's for $l = a$. Panel (d) is a graph of T_c vs x_i with $n_c/n_i = 0.03$ for $l = \infty$ and $l = a$. For $l = \infty$, open circles are Monte Carlo results, while the solid line in (d) is a theoretical curve, given by $29.2x_i^{4/3}$; for $l = a$, closed circles are Monte Carlo T_c's, and the solid line is a guide to the eye.

disorder averaging) obtained for the undamped ($l = \infty$) case. For small n_c/n_i, one sees an apparent plateau where spins are essentially perfectly collinear; a reduction in the polarization begins for $n_c/n_i \gtrsim 0.05$. It is tempting to regard the "plateau" and noncollinear regimes as distinct phases, but this notion can be seen to be illusory if one considers that for any spin there is always a finite probability of having a void large enough that the distance from the spin to its nearest neighbor exceeds the distance to the first zero of the RKKY interaction. Since the moment would not interact ferromagnetically with the closest spin, it is reasonable to assume that there is always some noncollinearity for any finite value of n_c/n_i.

On the other hand, for larger n_c/n_i it is also not evident in Fig. 1(a) where the polarization drops to zero (as would happen in the thermodynamic limit), and one has instead a long tail for $n_c/n_i \gtrsim 0.1$. This slow decay of the magnetization for even fairly large systems (e.g., $\langle N \rangle = 1000$), due to the presence of large magnetic clusters in the NF phase near the phase boundary, discourages the simple approach of locating the FM/NF transition by seeking where m vanishes, and we instead seek a sharper signal for the phase transition, magnetic percolation. In graph (b), the $T = 0$ magnetic susceptibility $\chi = [\langle \mathbf{S}^2 \rangle] - [\langle |\mathbf{S}| \rangle]^2$ (with a peak at $n_c/n_i \sim 0.1$) is shown. Similar singular behavior is also a feature of the finite temperature second order FM to NF transition (where the temperature is varied with all other parameters held fixed), and the $\chi(T = 0)$ peak suggests that our FM to NF phase transition represents a zero temperature critical point where the critical behavior is driven by disorder rather than by thermal fluctuations, a hallmark of a percolation transition.

Panel (c) of Fig. 1 depicts the Curie temperatures T_c vs n_c/n_i; for the undamped case (solid symbols) T_c peaks for $n_c/n_i \sim 0.03$ and then declines, vanishing for $n_c/n_i \approx 0.1$ in sharp contrast with Curie-Weiss continuum MFT, which yields the monotonically increasing $T_c \propto n_i n_c^{1/3}$. For the damped RKKY model (open symbols) with $l = a$, T_c is maximized for $n_c/n_i \sim 0.12$, and the Curie temperature is nonzero over a broader range than for $l = \infty$. However, the peak T_c for the damped model is considerably suppressed relative to that of the $l = \infty$ case. Panel (d) of Fig. 1 also displays T_c, but with $n_c/n_i = 0.03$ fixed and x_i allowed to vary. The open circles are the Monte Carlo results for $l = \infty$, while the solid curve is a theoretical curve given by $T' = 29.2 x_i^{4/3}$. It can be shown that the good agreement of the $x_i^{4/3}$ law with the calculated Curie temperatures implies that even for x_i as large as 0.2 the system is in the dilute limit (i.e., thermodynamic quantities are insensitive to the details of the lattice structure). However, this does not mean that the dependence of T_c is correctly given by continuum MFT, and one actually has $T_c = x_i^{4/3} g(n_c/n_i)$, where g is constant in continuum Curie-Weiss theory, but in our case has a nontrivial dependence as highlighted in Fig. 1(c) and in the inset.

By finding the critical n_c/n_i values for various Mn concentrations x_i, we construct a phase diagram for the RKKY model for DMS systems, indicated for the undamped ($l = \infty$) case by the solid line in Fig. 2; a substantial range of x_i values is included, certainly encompassing the experimentally relevant range. Nonetheless, the phase boundary deviates very little from the low (but finite) $n_c/n_i = 0.1$. This is consistent with the notion that the dilute limit has been reached even for Mn doping levels as high as 20%.

As ferromagnetism in $Ga_{1-x}Mn_xAs$ is found in experiment to be robust for $n_c/n_i > 0.1$, for a more realistic treatment we consider a damping scale equal to the crystal lattice constant, $l = a$. Since the factor $e^{-r/l}$ attenuates the more distant AF couplings more sharply than the ferromagnetic interactions at closer range, one expects RKKY damping to extend the FM/NF phase boundary to greater n_c/n_i values, and the dashed line in Fig. 2, which displays the FM/NF phase boundary for $n_c/n_i \geq 0.25$, is consistent with this intuition. It is important to note that although damping expands the domain of the ferromagnetic phase by suppressing AF couplings more severely than ferromagnetic interactions, the fact that the latter are reduced leaves the ferromagnetic state more readily disrupted by thermal fluctuations as can be seen in Fig. 1 where T_c values for the ($l = \infty$) and the ($l = a$) cases are shown together. Note that an alternate possibility for an extended FM regime in the phase diagram could be the Fermi surface warping [4] due to band structure effects in GaMnAs.

For $Ga_{1-x}Mn_xAs$, where the relevant parameter regime of interest is $n_c/n_i < 0.5$, $x < 0.1$, and $l \sim a$, a salient early experimental finding is that T_c is maximized for $x_i = x_i^{\mathrm{opt}} \sim 0.07$ (though this is dependent on sample preparation). We consider strong local superexchange AF couplings (i.e., between nearest neighbors on the fcc lattice) as a major contribution to the weakening of the ferromagnetic phase for greater x_i, where neighboring Mn impurities are more common. These adjacent moments would presumably form spin singlets and hence not contribute to the ferromagnetic phase. Since the value of the local Mn-Mn coupling J^{AF} is not precisely known, we examine a representative set of values, and we work in terms of the rescaled superexchange coupling $j^{\mathrm{AF}} \equiv J^{\mathrm{AF}}/J^{\mathrm{RKKY}}_{\mathrm{max}}$, where $J^{\mathrm{RKKY}}_{\mathrm{max}} = 4\pi J_0$ is the maximum FM coupling possible (for

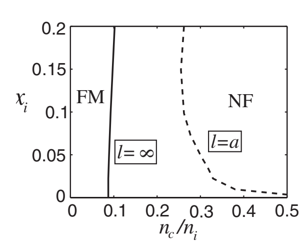

FIG. 2. Phase diagrams for the RKKY model; the solid line corresponds to the undamped, full RKKY model, while the dashed line is for the damped case where $l = a$.

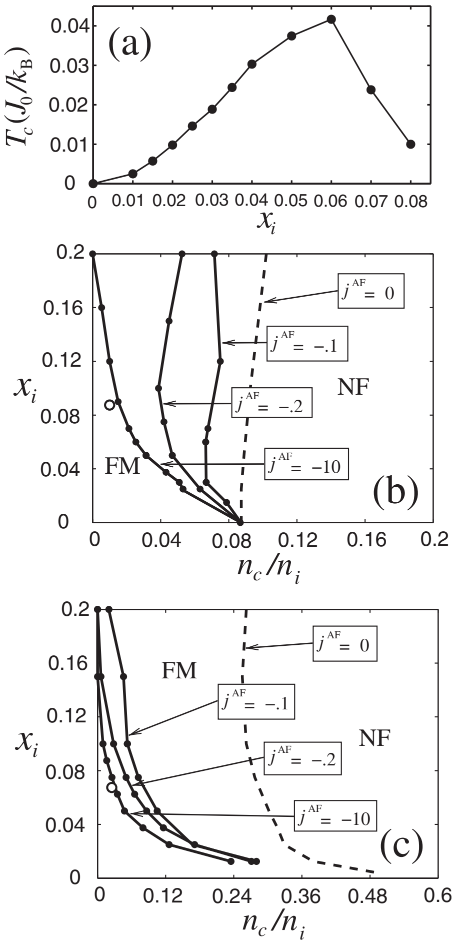

FIG. 3. The graph in panel (a) displays T_c vs x for $n_c/n_i = 0.05$, $l = a$, and $j^{AF} = -10$. Panels (b) and (c) are phase diagrams for the RKKY model with a local AF coupling incorporated for $l = \infty$ (b) and $l = a$ (c); the open circles at the left denote locations in the diagrams where T_c is optimized.

$l = \infty$) for two nearest neighbor impurities. Figure 3(a) shows a graph of T_c versus x_i for a large AF superexchange ($j^{AF} = -10$), with $n_c/n_i = 0.05$ and $l = a$ held fixed; T_c peaks for $x_i^{opt} = .06$, in reasonable accord with experiment. In general for any finite value of J^{AF}, larger x_i values will strongly suppress the FM phase due to the direct AF coupling between the Mn moments.

As was done for $J^{AF} = 0$, we obtain the $T = 0$ phase diagrams with results displayed in panels (b) ($l = \infty$) and (c) ($l = a$) of Fig. 3 for several j^{AF} values. In both the damped and undamped cases, it is evident that a strong to moderate j^{AF} sharply restricts the domain of the ferromagnetic phase; in (b) and (c), the ferromagnetic region quickly narrows as the impurity concentration rises beyond a few percent, effectively cutting off ferromagnetism for $x_i \gtrsim 0.1$. Only in the dilute limit, where neighboring impurity pairs are less abundant do the FM/NF phase boundaries in (b) and (c) revert to their position in the $j^{AF} = 0$ case. For the case of a very strong local AF interaction (with $j^{AF} = -10$), we have determined the values of x_i and n_c/n_i which maximize T_c, indicated in the phase diagrams of Figs. 3(b) and 3(c) with open circles.

Finally, we comment on the formation of magnetic clusters above T_c, for the strongly damped case where the length scale l of $J(r)$ is much smaller than the typical separation l_s between spins. It has been suggested [10] that in this limit, one can define a clustering temperature T^* above T_c where substantially sized magnetic clusters form. In fact, one can argue that $\xi \propto l_s(l_s/l)^{2/3}[\ln(T/T_c)]^{-2/3}$, indicating that the cluster size ξ is only weakly dependent on T/T_c, with much greater sensitivity to l_s/l. One can also seek a T^* below which typical magnetic clusters are at least $\xi = \alpha l_s$ in size, where α is a dimensionless factor, and one finds $T^* = \exp[\alpha^{3/2}(l_s/l)]T_c$; it is clear that T^* rapidly becomes large relative to T_c in the $l \ll l_s$ limit.

In conclusion, we have worked out the $T = 0$ phase diagram of the full RKKY model in the dilute limit (i.e., in the DMS context) at $T = 0$, finding that a ferromagnetic ground state is indeed supported, albeit over a very small region of the phase diagram, while continuum mean field theory erroneously assumes ferromagnetic order as the stable $T = 0$ phase leading to $T_c^{MFT} \propto n_i n_c^{1/3}$, quite distinct from our nonmonotonic T_c. We have found that the $T = 0$ phase diagram of the RKKY model consists of ferromagnetic and nonferromagnetic phases separated by a line of $T = 0$ critical points in which the NF phase gives way to ferromagnetic order via the percolation of magnetic clusters. We note that a zero temperature transition of a NF to a FM phase signaled by magnetic percolation is not special to the RKKY model in the DMS context, but is of broad relevance to magnetic systems where there is strong disorder and circumstances which allow for substantial AF couplings. We have found that introducing a cutoff in $J(r)$ (arising, for example, from disorder or carrier localization effects) and including a local AF superexchange term leads to a $T = 0$ phase diagram and T_c behavior in reasonable agreement with experiment. The FM phase of the RKKY DMS model is fragile, existing over a restricted parameter space ($l = \infty$) with moderate T_c's or on a broader domain (finite l) of parameter space with reduced T_c's.

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