Long Paper Draft

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We critically discuss the shortcomings of the continuum Weiss-Dietl Virtual Crystal Approximation (VCA) for diluted magnetic semiconductors such as $Ga_{1-x}Mn_xAs$, and we discuss in this context *lattice mean field theory*, which takes into account the discrete nature of the crystal lattice. Mean field theories are discussed in light of results from Monte Carlo calculations, and it is emphasized that the DMS parameter space corresponds to a strongly disordered regime in a sense opposite to the realm amenable to mean field theoretic approaches. Nevertheless, we obtain the critical exponents at the 1% level, showing little deviation from those of the simple cubic (non-disordered) Heisenberg model. We obtain $T = 0$ phase diagrams for the RKKY model for DMS, the *nn* FM; *nnn* Af model examined by Binder *et al*, and models with strong bond disorder, arguing that the $T = 0$ phase transitions in each case constitute zero temperature magnetic percolation critical transitions. We also speculate briefly about the nature of the non-ferromagnetic phase.

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I. INTRODUCTION

II. A CRITICAL EXAMINATION OF DMS MEAN FIELD THEORIES

The Virtual Crystal Approximation (VCA)

The VCA, a continuum mean field theory, neglects the crystal lattice, failing to predict even the qualitative behavior of T_c. Indeed, the VCA formula fails on a qualitative level, since it predicts a T_c which rises monotonically in the carrier concentration n_c for fixed impurity doping fraction x_i, in stark contrast to the non-monotonic dependence which must inevitably occur.

Lattice Mean Field Theory

By taking into account the discrete structure of the GaAs crystal lattice, we capture salient qualitative T_c trends as well as the shape (concave or convex) of the $m(T)$ curve.

strongly disordered DMS systems with primarily FM couplings

Site Disorder

the strong disorder limit The parameter regime appropriate to magnetic semiconductors is characterized by an interaction scale l small in comparison with the average separation $l_s = n_i^{-1/3}$ between Mn impurities, a condition where simple mean field theoretic approaches fare poorly. In fact, the $l \ll l_s$ regime is better understood in terms of random sphere percolation, in a sense opposite to the $l_s \ll l$ "mean field" limit. With the random sphere percolation picture and a real space renormalization group argument, we obtain a theoretical formula for T_c given by $T_c = \eta / k_B J(2 r_c l_s)$, where k_B is the Boltzmann constant, η is a dimensionless constant of order unity, and $r_c \approx 0.43$ is the critical radius for the percolation of random spheres where the volume density of the spheres is assumed to be unity. For couplings such as the damped RKKY with an exponential cutoff $e^{-r/l}$, T_c decreases very sharply as the localization length is decreased.

We discuss below results from Monte Carlo calculations checking the accuracy of the T_c forumula given above, thereby verifying that the typical DMS parameter range (with a finite interaction range $l \sim a$ and dilute impurity concentrations $x_i \lesssim 0.1$) corresponds to the strongly disordered regime mentioned above where simple mean theoretical techniques are manifestly untenable.

Magnetic clustering above Tc It has been suggested that for strongly disordered systems with $l \ll l_s$, there is a temperature T^* above the Curie temperature where there is nonetheless substantial local ferromagnetic ordering. However, we will argue that there is no definite T^* where ferromagnetic clusters suddenly appear, though there is a significant temperature regime above T_c where the ferromagnetic correlation length ξ is considerably larger than the inter-moment spacing l_s. The clustering phenomenon is illustrated in 1 which depicts Swendson-Wang snapshots of a disordered 2D Ising model with a short-ranged exponential coupling $J(r)$ where sizable clusters of aligned spins can be seen in panel (b) even for temperatures considerably higher than T_c (i.e. for $T = 2T_c$). We will obtain via theoretical arguments a rough estimate of ξ^* characterizing the T^* phenomenon, and we will find that the size of the magnetic clusters is a disorder mediated phenomenon, with a strong dependence on the index l_s/l and a comparatively weak temperature dependence.

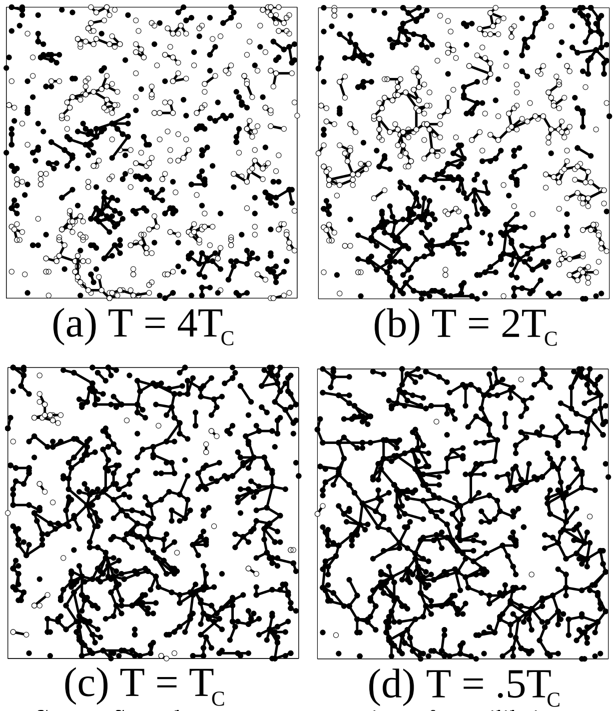

FIG. 1. Swendson-Wang portraits of equilibrium spin configurations for a strongly disordered 2D Ising model for $x = 0.05$, $J(r) = e^{-r/l}$, and $l/a = 1.0$. Each panel corresponds to a different temperature with $T/T_c = \{4.0, 2.0, 1.0, 0.5\}$ for (a), (b), (c), and (d) respectively. Dark (open) circles represent "up" ("down") spins.

To find a typical length scale ξ^* for the pocket of ferromagnetic order which form above T_c, we consider the effect of disorder fluctuations on local FM order and for simplicity model the ferromagnetic regions as spheres of radius ξ^*; we also neglect the specific crystal lattice structure, operating under the assumption that $l_s \gg a$. The mean number of impurity moments in a particular volume $V = 4/3\pi\xi^{*3}$ is $\langle N\rangle = \rho_i V$, and typical fluctuations in the particle number are given by $\delta N = \langle N\rangle$. Hence, the relative shift in particle density is $\delta N/\langle N\rangle = \langle N\rangle^{-1/2}$ with "compressions" (fluctuations which increase N) increasing the local ρ_i in V and "rarefactions" (disorder induced decreases in N) diminishing ρ_i. One expects local ferromagnetic ordering in V to arise from the former since fluctuations which increase N lead to a locally enhanced density ρ_i' and diminished separation between moments, $l_s' = \rho_i^{-1/3}$. As a result of the smaller l_s', the typical coupling between magnetic moments is enhanced leading to a local T_c^{loc} larger than the bulk value.

To obtain an idea of the typical cluster size, we will seek the largest spherical volume V for which typical number fluctuations (when positive) increase the proximity of magnetic impurities enough to raise the local T_c to T. We assume a coupling $J(r) = e^{-r/l}$ with a strong exponential cutoff (to facilitate analysis, a purely exponential coupling is assumed, though the results would be qualitatively similar for a more complicated $J(r)$ such as the damped RKKY function). Setting $T_c^{\text{loc}} = T$, and using the previously given bulk T_c formula, we have $T_c^{\text{loc}} = T = \eta/k_B J(2r_c l_s')$. Hence, the ratio of the enhanced local transition temperature to the bulk T_c is

$$T/T_c = J(2r_c l_s')/J(2r_c l_s) = e^{[2r_c/l(l_s - l_s')]} \tag{1}$$

$$= \exp\left[2r_c/l\left(\rho_i^{-1/3} - \rho_i'^{-1/3}\right)\right] \tag{2}$$

Seeking ξ^* (which can be computed from ρ_i' from $\rho_i' = (4\pi/3)\xi^{*3}$), we form the logarithm of both sides of Eq. 2 and multiply the result by $l/(2r_c)$, yielding

$$[l/(2r_c)]\ln T/T_c = \rho_i^{-1/3}\left[1 - \left(\rho_i^{1/3}/\rho_i'^{1/3}\right)\right] \tag{3}$$

Via $\rho_i' = (\langle N\rangle + \delta N)/V$ and $\delta N = \langle N\rangle^{1/2}$, we may rewrite $(\rho_i/\rho_i')^{1/3}$ as

$$(\rho_i/\rho_i')^{1/3} = [1 + \langle N\rangle^{-1/2}]^{-1/3} \approx 1 - \frac{2}{3}(\pi\rho_i\xi^{*3})^{-1/2} \tag{4}$$

where we have expanded the expression in brackets via the Binomial Theorem, noting that the accuracy of this approximation improves as ξ^* becomes larger. Substituting this expression for ρ_i/ρ_i' into the right side of Eq. 3 permits us to solve for the cluster size ξ^*. Solving for ξ^*/l_s to find the ratio of the magnetic cluster length scale to the spacing between magnetic moments yields

$$\xi^*/l_s = 2\,(\pi r_c/\rho_i)^{1/3}\,(l_s/l)^{2/3}\,[\ln(T/T_c)]^{-2/3} \tag{5}$$

In terms of the range of $J(r)$, ξ^* varies as $l^{-2/3}$, but only logarithmically in the temperature ratio T/T_c, and thus a very sharp cutoff in the interaction between spins could lead to appreciably sized ferromagnetic clusters even for temperatures substantially above T_c. This comparative insensitivity to temperature indicates that the T^* magnetic cluster phenomenon appears quite gradually as the temperature is lowered toward the Curie Temperature.

By inverting Eq 5, one can also determine the temperature ratios T/T_c for which spin clusters of a particular size αl_s appear. Solving Eq. 5 for T/T_c yields

$$T/T_c = \exp\left[(4/3)(l_s/l)r_c\pi^{-1/2}\alpha^{-3/2}\right] \tag{6}$$

It is evident from Eq. 6 that the temperature below which pockets of ferromagnetic order exceed l_s by a fixed (though possibly quite large) factor of α rises very sharply as l is reduced; in fact, there is an essential singularity in l/l_s.

Via rigorous Monte Carlo, we numerically check the formulae for the correlation length ξ^* given in Eq. 5.

Bond Disorder

In addition to models with very strong site disorder, we also examine situations in which there is comparatively little site disorder (e.g. in a regular cubic lattice), but very strong bond disorder. For sufficiently broad bond strength distributions, we develop a limiting theory analogous to the random sphere percolation theory pertinent in the case of very strong site disorder. Again, we validate our formula for T_c via Monte Carlo evidence. Via the latter, we also assess the validity of the Handrich result [K. Handrich, Phys. Status Solidi B **32**, K55 (1969)] which we find (as intuition would predict) to be quantitatively correct only when the width of the bond strength distribution is small in comparison with the mean coupling between impurity moments.

strong disorder in combination with appreciable AF couplings

Simple mean field theories tend to assume a ferromagnetic ground state, and this is a significant liability when antiferromagnetic couplings (e.g. from RKKY oscillations) become strong enough to partially or completely disrupt ferromagnetism even at very low temperatures. In the DMS context, one expects theories which presume a ferromagnetic ground state, as is true for Curie-Weiss continuum mean field theory, to encounter serious difficulties when n_c/n_i is large enough to eliminate ferromagnetism at the energetically favored ($T = 0$) phase. Graphs of juxtaposed VCA and Monte Carlo T_c results are discussed to highlight the manner in which the improper handling of AF couplings by mean field theoretic treatments contribute significantly for appreciable (i.e. experimentally relevant) values of n_c/n_i. Ultimately, the Monte Carlo T_c's fall to zero while the corresponding VCA results continue to rise, even deep in the "spin glass" regime with $n_c/n_i \sim 1$ (where a non-zero ferromagnetic T_c would be nonsensical). It is observed that the lattice MFT fares somewhat better than its continuum counterpart, in that it does eventually predict a non-monotonic Curie Temperature.

III. SIMPLE (RANDOM) DISORDER VERSUS SPECIAL TYPES OF DISORDER

We examine the effect on T_c of situations more complicated than simple random doping. In the simplest doping scheme, the occupancy of a site by an impurity is not influenced by the status of surrounding sites (occupied or unoccupied), and the probability of occupancy is given by the overall impurity fraction x_i. However, one can consider more complicated situations such as impurity superlattices. The impact of introducing correlations among magnetic impurities or arranging Mn dopants in regular superlattices can be a difficult question to address within the framework of mean field theory, so emphasis will be given to Monte Carlo calculations. However, some lattice MFT results will be discussed as well.

Relaxing the discretization condition

A very simple way to modify the manner in which Mn dopants are distributed is to remove the requirement that impurities reside only on specific sites in the fcc crystal lattice in favor of a continuous distribution in which impurities are randomly assigned to arbitrary positions.

Neglegible AF couplings ($n_c/n_i \ll 1$)

Detailed Monte Carlo data indicates very close agreement (generally to within 1% or better) between Curie temperatures obtained for magnetic impurities confined to a lattice and systems comprised of continuously distributed Mn dopants. This close agreement exists for essentially the entire x_i and l regime appropriate to DMS, with the proviso that AF couplings (and hence n_c/n_i are kept small.

We provide analytical arguments to account for the surprisingly close agreement of the continuum and discrete T_c's as well as other thermodynamic quantities (e.g. the magnetization $m(T)$ calculated for $T \leq T_c$). However, we emphasize that the similarities of the thermodynamic behaviors of impurities confined to the fcc lattice and magnetic dopants not bound to lattice positions does *not* imply that one can accept VCA formulae for T_c.

Appreciable Af couplings for larger n_c/n_i

We compare thermodynamic variables (particularly T_c) for discrete and continuous impurity distributions for situations (in the DMS context) where antiferromagnetic couplings are significant. The goal is to examine T_c for the two types of distributions (with $x_i \sim 0.1$ and $l \sim a$) while steadily increasing n_c/n_i to see where (or if) the agreement breaks down.

Impurity superlattice

In both Monte Carlo and MFT calculations, we study magnetic semiconductors in which the Mn dopants are organized in regular arrays commensurate with the Ga sites in the GaAs lattice.

IV. $T = 0$ PHASE DIAGRAMS OF MODELS WITH COMPETING INTERACTIONS

We consider qualitatively distinct models which allow for significant interplay of AF couplings and strong disorder, and we obtain the low temperature phase diagram in each case. As a common feature, we find a phase boundary marked by zero temperature magnetic percolation critical behavior as a NF phase gives way to a FM phase via the growth and coalescence of magnetic clusters culminating in full long range ferromagnetic order.

RKKY models in the DMS context

As in our recent PRL, we present $T = 0$ phase diagrams both for the damped and undamped RKKY models gleaned from Monte Carlo calculations. We assess the robustness of the ferromagnetic phase (when it exists), by calculating T_c as a function of n_c/n_i; non-monotonic behavior in the Curie Temperature is readily evident. One can also see that, though incorporating a damping factor $l = a$ increases the extent of the FM state on the phase diagram, while simultaneously reducing the peak T_c.

By calculating a few quantities which are invariant for a particular universality class (e.g. β/ν and the Binder cumulant computed at T_c), we find little difference (i.e. no more than a few percent) from the corresponding values for simple cubic (non-disordered) Heisenberg model.

The *nn* FM and *nnn* AF model on an fcc lattice

In addition to obtaining the low temperature phase diagram, we also calculate β/ν and the Binder cumulant evaluated at T_c as for the damped RKKY DMS models. It is quite possilbe that our results will be very close to the corresponding values for the simple cubic Heisenberg model, though Binder *et al* found much larger ($\sim 10\%$) deviations in 1995.

The Edwards-Anderson Model

The Edwards-Anderson model is an example of a model with bond disorder (in contrast to the previously discussed site disordered models) with substantial competing AF interactions set up by the random placement of antiferromagnetic bonds. The prevalence of the AF couplings is varied by adjusting the total fraction f_{A} of Af couplings; for simplicity, the latter are assigned the same magnitude as the FM couplings. We find that decreasing f_{A} from the standard 50% level leads to the same magnetic percolation phenomena seen in the site disordered models discussed previously. We find a transition to long-ranged FM order for $f_{\mathrm{A}} = 0.1$.

Again, as for the models with site disorder, we calculate β/ν and the Binder cumulant at T_c to within 1% accuracy. Our goal is to examine specifically the impact of increasingly prevalent AF couplings (but no site disorder) on the universality class. Based on preliminary results, any shifts in the exponents relative to the $f_{\mathrm{A}} = 0$ (standard simple cubic Heisenberg model) are expected to be slight.

Speculation as to the nature of the NF phase

We do not directly consider the phase very deep in the NF phase where $n_c/n_i \sim 1$ spin "glassy" behavior is expected, but we study the nature of the phase near the boundary with the ferromagnetic phase.

Short-ranged coupling between impurities

Intuition would suggest that in the very dilute limit, where $l \ll l_s$, the NF phase would behave more like a standard paramagnet and have relatively little glassy character. Moreover, the vicinity of the highly dilute limit, a good approximation to the ground state spin configuration can be achieved via a technique akin to "real space" renormalization group calculation, sketched below.

In seeking the lowest energy state in the $l \ll l_s$ regime, one first forms small spin clusters by aligning individual magnetic moments with the closest neighboring spin, as dictated by the coupling between them. These small groups of spins (usually spin pairs which either collinear or singlets, depending on the sign of the coupling between them) are then regarded as a new set of individual spins, and the effective coupling between these "spins" is calculated. The RG-like procedure is iterated until it terminates with all of the spins in the system are assigned to a single large cluster.

Longer-ranged $J(r)$, $l \geq l_s$

In regime where the range of $J(r)$ is at least comparable to l_s, complicated glassy behavior is a much stronger possibility in the NF phase. A strong local AF interaction between neighboring spins can also contribute glassy behavior for systems with higher impurity concentrations ($x_i \sim 0.1$).

Monte Carlo studies of "glassy" behavior in the FM phase

In the DMS context, we consider the possibility of glassy behavior even fairly deep in the ferromagnetic phase. We concentrate on n_c/n_i values which are moderate, though certainly not strong enough to disrupt the ferromagnetic order of the ground state. To assess the capacity of incipient glassiness to hamper the appearance of ferromagnetic order, we begin with a high temperature (purely random) spin configuration, we use the Heat-Bath Monte Carlo technique and abruptly cool the system to $T = 0$ (this is identical to the spin relaxation procedure use by Walker and Walstedt [PRB **22** 3816 (1980)] deep in the spin glass regime where $n_c/n_i \sim 1$).

Waiting for systems to relax to local minima is less computationally demanding than the annealing procedure one employs to find global minima in the energy landscape, and we should hence be able to access reasonably large systems.

Preliminary findings suggest that in the vicinity of the FM/NF phase boundary (though on the FM side), with increasing L, the local minima one finds when starting with the $T = \infty$ state are less and less likely to be ferromagnetically ordered. Ultimately, for very large L, glassy behavior can prevent altogether the relaxation of the high temperature state to a spin configuration with long-range frromagnetism. To examine this phenomenon in a quantitative manner, we calculate the normalized correlation length ξ/L, our goal being to determine the trend for very large L. Depending on whether ξ/L ultimately increases or decreases with the system size L, we determine whether in the thermodynamic limit competing AF couplings prevent the high temperature paramagnetic phase from attaining FM order under rapid cooling.

V. CONCLUSIONS

Salient points and major results are reiterated.

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[1] D. J. Priour, Jr., E. H. Hwang, and S. Das Sarma, Phys. Rev. Lett. **92**, 117201 (2004).