Superexchange induced canted ferromagnetism in dilute magnets

G. Bouzerar,1 R. Bouzerar,2 and O. Cépas3
1. Institut Néel, département MCBT, 25 avenue des Martyrs,
C.N.R.S., B.P. 166 38042 Grenoble Cedex 09, France
2. Université de Picardie Jules Verne (LPMC), 33 rue Saint-Leu, 80039 Amiens Cedex 01, France.
3. Laboratoire de physique théorique de la matière condensée,
C.N.R.S. UMR 7600, Université Pierre-et-Marie-Curie, Paris, France.
(Dated: February 1, 2008)

We argue, in contrast to recent studies, that the antiferromagnetic superexchange coupling between nearest neighbour spins does not fully destroy the ferromagnetism in dilute magnets with long-ranged ferromagnetic couplings. Above a critical coupling, we find a canted ferromagnetic phase with unsaturated moment. We have calculated the transition temperature using a simplified local Random Phase Approximation procedure which accounts for the canting. For the dilute magnetic semiconductors, such as GaMnAs, using ab-initio couplings allows us to predict the existence of a canted phase and provide an explanation to the apparent contradictions observed in experimental measurements. Finally, we have compared with previous studies that used RKKY couplings and reported non-ferromagnetic state when the superexchange is too strong. Even in this case the ferromagnetism should remain essentially stable in the form of a canted phase.

I. INTRODUCTION

The physics of disordered/dilute magnetic systems has attracted a considerable interest and attention from both theoreticians and experimentalists. Among these materials one finds for instance manganites (LaSrMnO3, LaCaMnO3...),12,13,14,15 diluted magnetic semiconductors as GaMnAs,16 which were widely studied, the so-called d0 materials (HfO2, CaO,...)5,6,7 the Heusler alloys as Ni2MnSn,8,9 or the double perovkites as Sr2FeMoO6.9,10,11 In these materials one of the key issue is the understanding of the influence of the carrier (hole/electron) concentration on both magnetic and transport properties. Indeed, the variation of the carrier concentration often leads to drastic changes and gives rise to interesting physics. In particular a competition arises between direct or superexchange interaction of the localized magnetic moments and indirect couplings via the itinerant carriers. In general, the superexchange coupling dominates at low carrier concentration but is overtaken by the ferromagnetic contribution at higher concentration. For example, in manganites (non dilute) the superexchange coupling competes with the double exchange coupling and leads to canted ferromagnetic phases.13,14,15 However, as soon as disorder is introduced into the system, new magnetic phases may appear such as ferromagnetic droplets in a canted antiferromagnetic matrix as observed in manganites.16,17 In dilute systems, where the probability to have nearest neighbour pairs is small it is not clear whether the superexchange coupling has the same effects. In particular, it is not obvious that superexchange alone can eventually completely destroy the ferromagnetic phase or induce new phases. The aim of the present study is to focus on this issue.

In this paper we show that in a dilute system of classical spins the superexchange competes with the long-ranged ferromagnetic couplings and favors a canted ferromagnetic phase in part of the phase diagram (temperature-concentration). However, in contrast to non dilute materials and double exchange systems, only spins involved in nearest neighbour pairs get canted (Fig. 1). This is particularly relevant for diluted magnetic semiconductors (DMS). In DMS, the magnetic couplings are extended and the superexchange dominates at sufficiently low carrier concentration. In these materials, a conflict between the measured low temperature total magnetic moment obtained by SQUID measurements and the density of spins extracted from X-ray diffraction (XRD) is often observed.18,19,20,21,22 We shall see that the existence of a canted phase provides a natural explanation to the observed disagreement. We also solve a conflict between recent Monte Carlo simulations23 which found ferromagnetism in a region where the Self-Consistent-Local RPA predicted an instability.24 This instability actually signals a new phase with unsaturated

FIG. 1: (Color online.) Schematic representation of the canted ground-state resulting from the competition between the long range ferromagnetic couplings Jij and the superexchange coupling JAF. The spins involved in pairs get canted and the angles θ, vary from spin to spin.
ferromagnetism, as we shall see. Because the nature of the ground state was not analyzed, this conclusion was missed in the Monte Carlo studies which focused on the amplitude of the Curie temperature only.

The manuscript is organized as follows. In the first part, we will analyze the effect of the superexchange coupling assuming a simple model for the extended exchange integrals. In the second part we will discuss the specific case of GaMnAs where it is known that superexchange coupling dominates over the indirect ferromagnetic contribution for sufficiently low hole density. In this part, for a quantitative study, realistic couplings will be taken from \textit{ab-initio} calculations (TB-LMTO). In the third and last part, we will discuss the case where the couplings are of the RKKY form, in order to study the competition between superexchange and frustration effects induced by the oscillating tail. It will be shown that, in the presence of the superexchange coupling, the stability region is significantly larger than found in previous studies.

In the following, we will consider the diluted Heisenberg Hamiltonian which reads,

\[ H = -\sum_{ij} x_i x_j J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{ij} x_i x_j J_{AF} \mathbf{S}_i \cdot \mathbf{S}_j \]

(1)

where the random variable \( x_i \) is 1 if the site is occupied by a magnetic impurity (otherwise 0). The total concentration of magnetic impurities is \( x \). The localized spin \( \mathbf{S}_i \) at site \( i \) is classical (\(|\mathbf{S}_i| = 1\)). The first term corresponds to the long-range exchange couplings and the second term is the nearest neighbour antiferromagnetic superexchange contribution. Because we will discuss the particular case of GaMnAs, for convenience we have performed all the calculations for a fcc lattice.

II. A SIMPLE UNFRUSTRATED MODEL

In this section we consider a simple model where the couplings are relatively extended but all ferromagnetic \( J_{ij} = J_0 e^{-\lambda r_{ij}} \). The parameter \( \lambda \) controls the range of the couplings; but since there is no abrupt cut-off, there is no strict percolation threshold for the ferromagnetism in this problem. The tail of the couplings always induce a finite transition temperature. In fact, this model is not so far from the exchange couplings in III-V diluted magnetic semiconductors such as Ga\(_{1-x}\)Mn\(_x\)As or Ga\(_{1-x}\)Mn\(_x\)N as calculated from first principles. More realistic couplings will be used in the next section. Whilst for \( J_{AF} = 0 \) and at \( T = 0 \) K, the ground state is ferromagnetic and fully saturated (no frustration), we discuss its nature in the presence of \( J_{AF} \). For this purpose, for a given configuration of disorder (position of the magnetic impurities), we minimize numerically the total energy associated to the Hamiltonian (1) with respect to the angles \((\phi_i, \phi_j)\). For simplicity, we consider only the case where the spins are coplanar \((\phi_i = 0)\). The calculations were performed for systems containing typically 1000 diluted spins. We have found that beyond a critical value of \( J_{AF} \), one pair of nearest neighbour spins \((\mathbf{S}_i, \mathbf{S}_j)\) starts to get canted (see Fig. 1) whereas all the other spins remain aligned along the magnetization axis. As we increase \( J_{AF} \) more pairs get canted but the spins that have no nearest neighbour remains almost parallel to the z axis. The canting results from the competition between the local field resulting from the long range couplings and the superexchange contribution of the nearest neighbour spin. Each spin sees a different environment and therefore the canting does not occur simultaneously for all the pairs as we increase \( J_{AF} \). For the same reason, the canting angles are different from spin to spin. For the unpaired spins, however, the canting angle is very small. This results from two combined effects. On one hand, for each pair, \( \phi_i \) is close to \(-\phi_j\), so that their resulting transverse field is small. On the other hand, a given unpaired spin \( \mathbf{S}_k \) experiences the sum of the transverse fields due to all canted pairs. Because of their random sign, the sum averages out to a small value. We can therefore neglect the small canting angles of the unpaired spins, as shown in Fig. 1.

For simple illustration, we recall what happens to a single pair in the effective field of the other spins. The energy of this pair is \( E = J_{AF} \cos(\phi_i - \phi_j) - h_i \cos \phi_i - h_j \cos \phi_j \), where \( h_i = \sum_j J_{ij} \) is the local field on spin \( i \) (for simplicity, we assume \( h_i = h_j = h \) to be the same for both sites, so that \( \phi_i = -\phi_j = \theta \)). The minimization gives a canting angle,

\[ \cos \theta = \frac{h}{2J_{AF}} \]

for \( J_{AF} \geq h/2 \) (and \( \theta = 0 \) otherwise). For \( J_{AF} \to \infty \), the two spins are anti-aligned and orthogonal to the other spins: they are effectively decoupled from them. We emphasize that, in our calculations, we have kept the real local fields \( h_i \) that differ from site to site. In addition we also have a finite probability to have trimers of spins, quadrimers, etc. so that the real canting angles are not given by (2) but are determined self-consistently.

Now, in order to calculate the critical temperature, we include thermal fluctuations about the ground state. In the case where the ground state is fully polarized, it has been shown that the self-consistent local random phase approximation method (SC-LRPA) is reliable. When directly applied to models in presence of superexchange couplings, it has been argued that ferromagnetism disappears when the nearest neighbour coupling is dominated by the antiferromagnetic superexchange contribution. In fact, the reported instability turns out to occur at the same critical value at which the ground-state becomes canted, as found above. The instability therefore directly reflects the change of ground state and should not be interpreted as the result of frustration of the long-range couplings. In order to correct for this, we extend the SC-LRPA to calculate the critical temperature of a canted state.
original SC-LRPA one has to start with the equation of
motion of the retarded Green’s function, $G^{\mu \nu}_{ij}(\omega)$, where,

$$G^{\mu \nu}_{ij}(\omega) = -i \int_{-\infty}^{+\infty} dt e^{i\omega t} \langle [S^\mu_i(t), S^\nu_j(0)] \rangle \tag{3}$$

where $\mu, \nu$ are the spin components. Because of the
canted ground-state, the decoupling of the equation of
motion involves both the longitudinal $\langle S^z_i \rangle$ and trans-
verse magnetizations $\langle S^x_i \rangle$. As a consequence, the trans-
verse Green’s function, $G^{x x}_{ij}$, is now coupled to both
$G^{z z}_{ij}$ and $G^{x x}_{ij}$. Since solving these coupled equations
is more involved we propose a simplified ansatz: af-
after the determination of the canting angles $\{\theta_i\}$ (for a
given configuration of disorder) we map the canted prob-
to an effective fully ferromagnetic one with reduced
spin amplitude, $S_i \rightarrow S \cos(\theta_i)$. Note that, this is equiva-
 lent to replace the couplings by $J_{ij} \rightarrow J_{ij} \cos(\theta_i) \cos(\omega_j)$. The
advantage of this mapping is that we can use the stan-
don SC-LRPA to calculate the Curie temperature $T_C$, although the ground state is canted. Note also that,
this mapping is exact in the two limiting cases: (i) small
superexchange coupling and (ii) large $J_{AF}$ limit where
the canted pairs become deconnected from the system.
As will be discussed in the following, a comparison with
Monte Carlo results supports this procedure in the inter-
mediate coupling regime as well.

We now come back to the simple model where the
long ranged couplings are defined by $J_{ij} = J_0 e^{-x_{ij}}$. In
Fig. 2 we have plotted the Curie temperature as a func-
tion of $J_{AF}$ for a fixed density of magnetic impurities
and various values of the coupling range. First, as long
as the superexchange coupling is smaller than typically
the ferromagnetic coupling between nearest neighbors,
$J_1 = J_0 e^{-x_{ij}}$, $T_C$ is almost insensitive to $J_{AF}$. This
is in agreement with previous observation that in the
diluted regime, the Curie temperature is controlled by
couplings corresponding roughly to the average distance
between the magnetic impurities, $x^{-1/3}$. When $J_{AF}$ is in-
creased, there is a critical value above which the ground
state gets canted (the critical value increases with $\lambda$).
When this happens, we observe a reduction of $T_C$, and then
a saturation to a finite value for strong $J_{AF}$. Let
us discuss the limit of strong $J_{AF}$. The saturation of
$T_C$ corresponds to the regime where nearest neighbour
spins are orthogonal to the other spins. The saturated
value can be viewed as that of a system of $x_{eff}$ spins (interacting with ferromagnetic interactions) in which all
pairs have been removed. For example, on the fcc lat-
tice, for $x = 0.05$ the concentration of spins involved in
pairs is 0.0225, or $x_{eff} = 0.55 \times T$. The new character-
istic distance between remaining impurities is $x_{eff}^{1/3}$ and
has to be compared to the coupling range $\lambda$, which con-
trols whether the impurities “percolate”. When reducing
$\lambda$, the ratio of the saturating value over $T_C(0)$ gets quite
large (see Fig. 2) because we approach the regime of non-
“percolation”, where the remaining impurities get more
weakly coupled. To conclude this paragraph, we observe
a smooth crossover in the Curie temperature between a
weak-coupling regime where $J_{AF}$ has no effect and a
strong-coupling regime where it is equivalent to remove
all spins which are coupled by $J_{AF}$. This is true for a
concentration of impurities small enough and we now in-
vestigate the effect of varying the concentration.

![Fig. 2](image-url)

**Fig. 2:** (Color online.) Curie temperature as a function of the superexchange coupling strength. The density of magnetic impurities is set to $x = 0.03$ and the coupling range $\lambda$ varies from 0.2 to 0.75. $J_1 = J_0 e^{-a_{ij}}$ denotes the nearest neighbour coupling in the absence of superexchange. The dashed line shows the instability threshold of the simple (non-canted) SC-
LRPA calculation.

![Fig. 3](image-url)

**Fig. 3:** (Color online.) Curie temperature (in units of $J_1$) as a function of the magnetic impurity concentration $x$ for different values of the superexchange strength. The range of the couplings, $\lambda$, is fixed to 0.50.
a function of the impurity concentration, \( x \), for a fixed value of the parameter \( \lambda \) and different values of \( J_{AF} \). In the absence of superexchange coupling, we observe a strong increase of \( T_C \) with \( x \). For \( J_{AF} = 4J_3 \), the Curie temperature is strongly reduced and exhibits a maximum at \( x \approx 0.15 \). The maximum reduces to \( x \approx 0.08 \) and is more pronounced for \( J_{AF} = 10J_1 \). Above this value the Curie temperature decreases strongly to eventually vanish at \( x \approx 0.30 \). The presence of a maximum can be understood as resulting from the competition between two effects. As we increase the density of magnetic impurities, \( x \), the local fields increase (the impurities interact more strongly because they get closer). At the same time the probability of nearest neighbor spins increases also. Since nearest neighbor spins get canted the local fields they create on the other spins is reduced (and eventually vanish in the limit of infinite \( J_{AF} \)), so that the number of magnetically-active spins (as far as ferromagnetism is concerned) is effectively reduced. Note that for strong \( J_{AF} \), one expects a site percolation which for nearest neighbour coupling on fcc lattice occurs at \( x_C \approx 0.20 \). Beyond this critical value, the phase should be of Néel type. One would need to include all fluctuations in order to calculate its critical Néel temperature, a problem beyond the scope of the present paper, since we focus on the ferromagnetic phase only.

III. REALISTIC COUPLINGS: THE CASE OF GaMnAs

Let us now discuss the case of the widely studied diluted III-V magnetic semiconductor Ga\(_{1-x}\)Mn\(_x\)As. First, we remind that the substitution of Ga\(^{3+}\) by Mn\(^{2+}\) introduces a localized spin \( S=5/2 \) and a hole in the valence-band (more precisely in the impurity band).

During the molecular beam epitaxy (MBE) growth of the samples, there are additional defects which appear, namely As anti-sites (As\(_{Ga}\)) which substitute the Ga sites. Arsenic antisites formation is one of the main mechanisms for compensation in diluted magnetic semiconductors. They lead to the reduction of the density of carriers which in turn reduces the strength of the magnetic couplings and eventually the Curie temperature. In term of holes, As\(_{Ga}\) is a double acceptor (double donor of electrons). If \( y \) denotes the density of As antisites and \( x \) the density of Mn\(^{2+}\) then the density of holes is \( n_h = x - 2y \). The reduction of the carrier density via As\(_{Ga}\) not only affects the long range couplings but also allows to ”tune” the superexchange coupling\(^{30,31}\), and thus provides a way to test the ideas developed above. Indeed, beyond a certain concentration of As\(_{Ga}\), the superexchange mechanism dominates the nearest neighbour coupling (see Figure 5 in \([23]\)). This is clear from \textit{ab initio} studies, where the couplings have been calculated without any adjustable parameters (Tight-Binding Linear Muffin Tin Orbitals), and subsequently used in several publications\(^{23,24,27}\).

![Figure 4](image-url)  
**FIG. 4:** (Color online.) Calculated phase diagram (temperature-antisite concentration) for Ga\(_{1-x}\)\(_y\)As\(_x\)Mn\(_y\)As indicating the predicted canted phase. The antisite concentration \( y \) allows to tune the carrier concentration. The density of magnetic impurity \( x \) is fixed to 0.05. The transition temperature is calculated with modified LRPA, and compared with Monte Carlo (from ref. \([23]\)), using the same set of \textit{ab-initio} couplings. \( m(0) \) is the total magnetization per spin at zero temperature.

In Fig.\([4]\) using SC-LRPA in the same way as described in section II, and using \textit{ab-initio} couplings, we calculate the predicted phase diagram for Ga\(_{1-x}\)\(_y\)As\(_x\)Mn\(_y\)As (temperature - density of As antisites). By energy minimization, we have found a wide region of the phase diagram \((0.01 < y < 0.0175)\) where the ground state is not fully ferromagnetic but canted. This is the consequence of the superexchange coupling \( J_{AF} \) that increases when the antisite concentration gets larger. As far as the transition temperature is concerned, we stress that the values obtained are in very good agreement with that of Monte Carlo simulations\(^{23}\). Although the nature of the phase was not discussed in their study, this validates the simplified treatment of the canting of the ground-state. Note that, both Monte Carlo simulations and SC-LRPA were performed (i) with the same exchange couplings and (ii) with the same number of shells (approximately 20). Note also that the values of the Curie temperature in the fully polarized ferromagnetic phase \((y \leq 0.01)\) are a little smaller here than those published in ref. \([27]\) because the number of shells is smaller. More importantly, in this study, the Curie temperature vanished abruptly beyond \( y = 0.01 \) (see Fig. 1 of ref. \([27]\)). This instability in fact signals the occurrence of a new phase, that we identify as a canted phase. Thus, in contrast to what was claimed before, the ferromagnetism survives down to much smaller concentrations of carriers than anticipated (up to \( y \approx 0.0175 \)), but is non-saturated. Beyond this concentration, the canted phase disappears because the long-range couplings also become antiferromagnetic and
thus introduce real frustration into the system. Because of that, for $y \geq 0.0175$ the ground-state is expected to be a spin-glass.

To characterize the ground state, we have also calculated the total magnetization per spin $m(0)$ at $T = 0$ K. For $y \leq 0.01$ the magnetization is $m(0) \approx 1$ (by definition). As we enter the canted ferromagnetic phase the total magnetization starts to reduce significantly. For $y = 0.0125$ the magnetization is already reduced by 10% and for $y = 0.015$ it is only $m(0) \approx 0.60$ which is very close to the lower bound obtained by removing all pairs, $m(0) \geq x_{eff}/x = 0.55$ for $x = 5\%$. In order to get an idea of how the magnetization changes from site to site, we have plotted the distribution of canting angles in Fig. 5. Note that the distribution is given for spins having at least one nearest neighbor. We observe for $y = 0.0125$ that approximately 40\% of the spins are still not canted (delta peak at $\theta = 0$). The distribution of angles is very broad with a maximum at about 50°. On the other hand, for $y = 0.015$ we observe a strong change in the distribution. In the latter case, all spins are canted, and the distribution peaked at about $\theta \approx 80°$ is more narrow than that of $y = 0.0125$.

Let us now discuss the relation between our calculations and experimental data. It is often seen in the literature that the measurement of the bulk magnetization (by SQUID) is different from the magnetization expected from the determination of the Mn density from XRD measurement, assuming a fully polarized ferromagnet. The direct measurement often leads to much smaller values. Furthermore, it is also seen that the magnetization strongly changes after annealing of as grown samples. During annealing, the magnetic impurities are redistributed in the sample, which becomes more homogeneous. In a recent study, it was shown that one could explain the effect of different annealing treatments by the existence and the rearrangement of interstitial Mn defects (Mn$_I$). Indeed Mn$_I$ is a defect that preferentially sits near a Mn ion (which substitutes Ga) and is coupled antiferromagnetically to it. This leads to the formation of a local singlet state for the dimer of Mn and therefore reduces the number of magnetically active Mn, and hence the total magnetization. However, it is now possible to control the density of carriers by chemical hydrogenation of the samples. In this process, it is believed that the density of Mn and the density of defects Mn$_I$ do not change. Therefore, if interstitial Mn$_I$ defects were the main source of reducing the bulk magnetization $m(0)$, one would expect $m(0)$ to remain the same for all these hydrogenated samples. This is in contradiction with the measurements that indicate that the samples with lowest carrier density (insulating or very dirty metallic behavior) have a much smaller $m(0)$. For instance, in Fig. 3 of ref. [36] at small fields ($H = 500$ Oe), we observe that the magnetization is about two times smaller for hydrogenated sample with the lowest $T_C$, compared with the reference sample with no hydrogenation. This is very hard to reconcile with the presence of Mn$_I$ because this would require a large number of such defects. Our study points out a different compensation mechanism, that must be at play once the coupling between nearest neighbours is antiferromagnetic (as evidenced from ab initio studies). As we said before, the number of pairs of nearest neighbors is large at $x = 5\%$, so the reduction of $m(0)$ is already large without having to invoke a large number of Mn$_I$. We therefore argue that the reduction of the total magnetization is due to the canting of the pairs which occurs when the density of carriers becomes small enough, and we suggest to re-analyze the experimental data on the basis of the present work.

IV. MODEL WITH RKKY OSCILLATIONS

In the last paragraph we discuss another interesting case, the interplay between frustration resulting from the RKKY couplings and the superexchange. In order to compare with previous studies, we assume the long range couplings to be given by $J_{ij} = J_0 e^{-r_{ij}/r_0} F(k_{FR})$, where $F(k_{FR}) = (k_{FR}) \cos(k_{FR})/(r/r_0)^3$ and $r = |r_i - r_j|$, $r_0$ is the nearest neighbour distance. The parameter is an effective Fermi vector $k_F$ which is determined by the density of carriers. Note that this study is also motivated by the fact that RKKY couplings are often used to study the ferromagnetism in diluted magnetic semiconductors, although it was shown that they are inappropriate. In previous studies, it was argued, in particular, that the stability region for ferromagnetism was very narrow, upon increasing $J_{AF}$ a point that was reaffirmed later using Monte Carlo simulations. We now argue that the
FIG. 6: (Color online.) Curie temperature for the “RKKY” model as a function of \( k_F \) for different values of the superexchange coupling \( J_{AF} \). The density of impurity is set to \( x = 0.05 \) and the parameter \( \lambda = 0.50 \). The calculations are performed on the fcc lattice.

The stability region is in fact wider thanks to unsaturated long-range couplings. In Fig. 6 we have plotted the Curie temperature as a function of \( k_F \) for the RKKY-like model defined above. In the absence of superexchange coupling we observe that the Curie temperature (solid circles) exhibits a maximum and vanishes above \( k_F \approx 0.60 \) because of the oscillations in the couplings. This is in agreement with ref. \(^{28}\). However, when we switch on the superexchange coupling, we observe that the Curie temperature is reduced but does not vanish. This is in contrast to what was published previously where the ferromagnetism was apparently suppressed by the superexchange coupling. The reason for this discrepancy is as discussed previously, the occurrence of a new phase. Indeed previous calculations were done without including the canting. Again, the disappearance of \( T_C \) (Fig. 5 of ref. \(^{28}\)) reflects only the change in the nature of the ground state. As we increase \( J_{AF} \) further, we observe a saturation in the Curie temperature and almost an unchanged region of stability of the ferromagnetic phase. These results are also in contradiction with those of ref. \(^{28}\). Indeed, it is shown in this paper that the ferromagnetic phase is similarly suppressed (see Fig. 3 of ref. \(^{26}\)). The drastic reduction of the ferromagnetic phase occurs also at low carrier concentration, although the frustration effects due to the long range couplings are very small in this region. Let us discuss the origin of the discrepancy. In the absence of \( J_{AF} \), Monte Carlo \(^{26}\) and SC-LRPA \(^{28}\) give the same results (see Fig. 1 and 4 of ref. \(^{28}\) and Fig. 1c of ref. \(^{26}\)). The effects of frustration are therefore properly handled by SC-LRPA, and thus it cannot be the source of the disagreement. In addition, the comparison between the Monte Carlo simulations of \(^{26}\) with the modified SC-LRPA clearly shows that the effects of the superexchange are also properly treated. We suggest that the discrepancy comes from the existence of the canted phase that was missed before. It would be of interest to clarify this issue.

V. CONCLUSION

In conclusion, we have shown that the competition between nearest-neighbour antiferromagnetic superexchange coupling and long-range ferromagnetic couplings gives rise to a canted ferromagnetic phase in dilute magnets. We emphasize that short-range and long-range competing interactions play quite different roles. In the later case, the oscillating tail of the RKKY interaction, for instance, introduces frustration at long distance and consequently reduces the stability region of the ferromagnetic phase. In the first case, however, when superexchange is added, pairs of spins get canted but the stability region remains weakly affected, even at strong coupling. The Curie temperature of the canted phase is reduced simply because the canting weakens the internal fields. More generally, in random dilute systems, competing couplings of a range much shorter than the typical inter-impurity distance should lead only to local spin reorientations, and will not affect the long-range properties. Applying these ideas to GaMnAs, we have predicted the existence of a canted phase in the phase diagram. We have calculated its critical temperature using a modified local RPA approach that was found to be reliable by comparison with Monte Carlo simulations using the same \textit{ab initio} couplings. The existence of this phase provides a simple explanation to recent experiments in diluted magnetic semiconductors, where the bulk magnetization was found to be smaller than the saturation value; without having to invoke a large number of compensating defects in the samples. It would be of great interest to check by local probes whether the ground state is indeed canted in the range of concentrations we have predicted.

Acknowledgments

We would like thank E. Kats for the hospitality at ILL.

\(^1\) S. Jin, T. H. Tiefel, M. McCormack, R. A. Fastnacht, R. Ramesh, and L. H. Chen, Science 264, 413 (1994).

\(^2\) For a review, see A. P. Ramirez, J. Phys. Cond. Matt. 9, 8171 (1997).
