

Superexchange induced canted ferromagnetism in dilute magnets

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We argue, in contrast to recent studies, that the antiferromagnetic superexchange coupling between nearest neighbor 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 dilute magnetic semiconductors, such as GaMnAs, using *ab initio* couplings allows us to predict the existence of a canted phase and provides an explanation for the apparent contradictions observed in experimental measurements. Finally, we compared with previous studies that used Ruderman-Kittel-Kasuya-Yosida couplings and reported nonferromagnetic states when the superexchange is too strong. Even in this case the ferromagnetism should remain essentially stable in the form of a canted phase.

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I. INTRODUCTION

The physics of disordered and/or dilute magnetic systems has attracted considerable interest and attention from both theoreticians and experimentalists. Among these materials one finds, for instance, manganites (LaSrMnO₃, LaCaMnO₃),¹⁻⁴ diluted magnetic semiconductors like GaMnAs,⁵ which have been widely studied, the so-called *d*⁰ materials (HfO₂, CaO),^{6,7} the Heusler alloys like Ni₂MnSn,^{8,9} and the double perovskites like Sr₂FeMoO₆.^{10,11} In these materials, one key issue is the understanding of the influence of the carrier (hole or 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 superexchange¹² 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 (nondilute) the superexchange coupling competes with the double-exchange coupling and leads to canted ferromagnetic phases.¹³⁻¹⁵ 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 neighbor 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 nondilute materials and double-exchange systems, only spins involved in nearest neighbor pairs get canted (Fig. 1). This is particularly rel-

evant for diluted magnetic semiconductors (DMSs). In DMSs, 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 superconducting quantum interference device (SQUID) measurements and the density of spins extracted from x-ray diffraction (XRD) is often observed.¹⁸⁻²² We shall see that the existence of a canted phase provides a natural explanation for the observed disagreement. We also solve a conflict between recent Monte Carlo simulations,²³ which found ferromagnetism in a region where the self-consistent local random phase approximation (RPA) predicted an instability.²⁴ This instability actually signals a new phase with unsaturated 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 magnitude of the Curie temperature only.

The paper is organized as follows. In the first part, we analyze the effect of the superexchange coupling assuming a

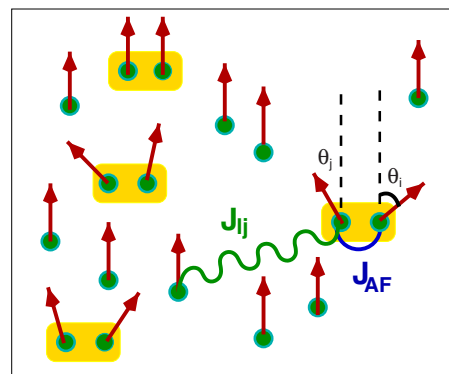


FIG. 1. (Color online) Schematic representation of the canted ground state resulting from the competition between the long-range ferromagnetic couplings J_{ij} and the superexchange coupling J_{AF} . The spins involved in pairs become canted and the angles θ_i vary from spin to spin.

simple model for the extended exchange integrals. In the second part we 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 *ab initio* calculations using the tight-binding (TB) linear muffin-tin orbital (LMTO) method.²⁵ Note that couplings calculated within other *ab initio* approaches should lead to similar results.^{26,27} In the third and last part, we discuss the case where the couplings are of the Ruderman-Kittel-Kasuya-Yosida (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.²⁸

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

$$H = - \sum_{ij} x_i x_j J_{ij} \vec{S}_i \cdot \vec{S}_j + \sum_{(ij)} x_i x_j J_{AF} \vec{S}_i \cdot \vec{S}_j, \quad (1)$$

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 \vec{S}_i at site i is classical ($|\vec{S}_i|=1$). The first term corresponds to the long-range exchange couplings and the second term is the nearest neighbor 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 $J_{ij} = J_0 e^{-r_{ij}/\lambda}$. The parameter λ controls the range of the couplings; but since there is no abrupt cutoff, there is no strict percolation threshold for the ferromagnetism in this problem. The tail of the couplings always induces a finite transition temperature. In fact, this model is not so far from the exchange couplings in III-V diluted magnetic semiconductors such as $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ or $\text{Ga}_{1-x}\text{Mn}_x\text{N}$ as calculated from first principles. More realistic couplings will be used in the next section. While 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 with the Hamiltonian (1) with respect to the angles (θ_i, ϕ_i) . 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 neighbor spins (\vec{S}_i, \vec{S}_j) starts to become 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 neighbor remain 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 neighbor spin. Each spin experiences 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, θ_i is close to $-\theta_j$, so that their resulting transverse field is small. On the other hand, a given unpaired spin \vec{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 $E = J_{AF} \cos(\theta_i - \theta_j) - h_i \cos \theta_i - h_j \cos \theta_j$, where $h_i = \sum_l J_{il}$ 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 $\theta_i = -\theta_j \equiv \theta$). The minimization gives a canting angle

$$\cos \theta = \frac{h}{2J_{AF}} \quad (2)$$

for $J_{AF} \geq h/2$ (and $\theta=0$ otherwise). For $J_{AF} \rightarrow \infty$, the two spins are antialigned 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 which 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 (SCLRPA) method^{24,29,30} is reliable. When directly applied to models in the presence of superexchange couplings, it has been argued that ferromagnetism disappears when the nearest neighbor coupling is dominated by the antiferromagnetic superexchange contribution.^{24,30} 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 the 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 SCLRPA to calculate the critical temperature of the canted state. In principle, as in the original SCLRPA, one has to start with the equation of motion of the retarded Green's function $G_{ij}^{\mu\nu}(\omega)$,

$$G_{ij}^{\mu\nu}(\omega) = -i \int_{-\infty}^{+\infty} dt e^{i\omega t} \theta(t) \langle [S_i^\mu(t), S_j^\nu(0)] \rangle, \quad (3)$$

where μ, ν are the spin components. Because of the canted ground state, the decoupling of the equation of motion involves both the longitudinal $\langle S_i^z \rangle$ and transverse magnetizations $\langle S_i^x \rangle$. As a consequence, the transverse Green's function $G_{ij}^{\pm-}$ is now coupled to both $G_{ij}^{\bar{-}}$ and $G_{ij}^{\bar{z}}$. Since solving these

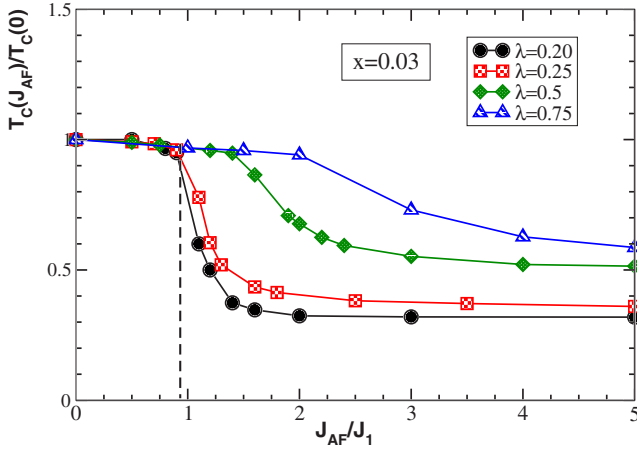


FIG. 2. (Color online) Curie temperature as a function of the superexchange coupling strength. The density of magnetic impurities is set to $x=0.03$ and the coupling range λ varies from 0.2 to 0.75. $J_1=J_0e^{-a/\sqrt{2}\lambda}$ denotes the nearest neighbor coupling in the absence of superexchange. The dashed line shows the instability threshold of the simple (noncanted) SCLRPA calculation.

coupled equations is more involved,³¹ we propose a simplified ansatz: after the determination of the canting angles $\{\theta_i\}$ (for a given configuration of disorder) we map the canted problem to an effective fully ferromagnetic one with reduced spin amplitude, $S_i \rightarrow S \cos(\theta_i)$. Note that, this is equivalent to replacing the couplings by $J_{ij} \rightarrow J_{ij} \cos(\theta_i) \cos(\theta_j)$. The advantage of this mapping is that we can use the standard SCLRPA to calculate the Curie temperature T_C , although the ground state is canted. Note also that this mapping is exact in two limiting cases: (i) small superexchange coupling and (ii) the large- J_{AF} limit where the canted pairs become disconnected from the system. As will be discussed in the following, a comparison with Monte Carlo results supports this procedure in the intermediate coupling regime as well.

We now come back to the simple model where the long-ranged couplings are defined by $J_{ij}=J_0e^{-r_{ij}/\lambda}$. In Fig. 2, we have plotted the Curie temperature as a function 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, ($J_1=J_0e^{-a/\sqrt{2}\lambda}$), T_C is almost insensitive to J_{AF} . This is in agreement with previous observations 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 increased, there is a critical value above which the ground state gets canted (the critical value increases with λ). 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 neighbor 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 lattice, for $x=0.05$ the concentration of spins involved in pairs is 0.0225, or $x_{eff}=0.55x$. The new characteristic distance between remaining impurities is $x_{eff}^{-1/3}$ and has to be

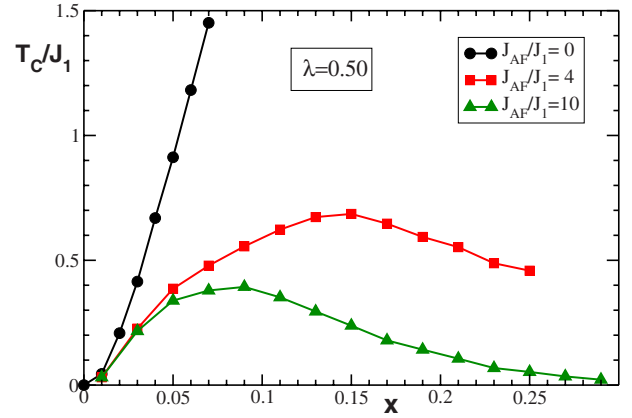


FIG. 3. (Color online) Curie temperature (in units of J_1) as a function of the magnetic impurity concentration x for different values of the superexchange strength. The range of the couplings, λ , is fixed to 0.50.

compared to the coupling range λ , which controls whether the impurities “percolate.” When λ is reduced, the ratio of the saturating value over $T_C(0)$ gets quite large (see Fig. 2) because we approach the regime of nonpercolation, 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 removing all spins that are coupled by J_{AF} . This is true for a small enough concentration of impurities and we now investigate the effect of varying the concentration.

In Fig. 3, we have plotted the Curie temperature as a function of the impurity concentration x for a fixed value of the parameter λ and different values of J_{AF} . In the absence of superexchange coupling, we observe a strong increase of T_C with x . For $J_{AF}=4J_1$, the Curie temperature is strongly reduced and exhibits a maximum at $x \approx 0.15$. The maximum is reduced 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 become canted, the local fields they create on the other spins are 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 neighbor 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 $\text{Ga}_{1-x}\text{Mn}_x\text{As}$. First, we recall

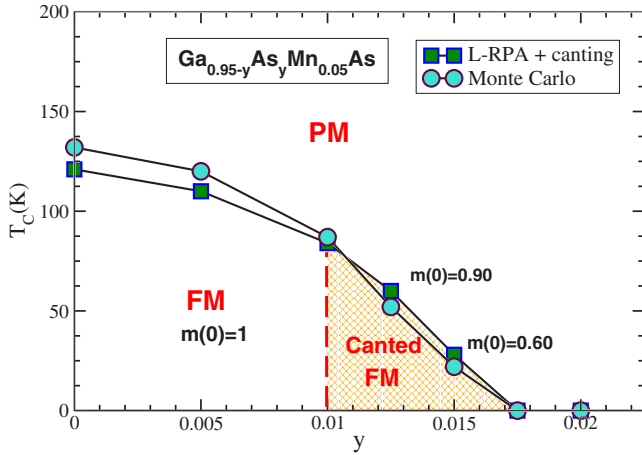


FIG. 4. (Color online) Calculated phase diagram (temperature-antisite concentration) for $\text{Ga}_{1-y}\text{As}_y\text{Mn}_{0.05}\text{As}$ indicating the predicted canted phase. The antisite concentration y allows tuning of the carrier concentration. The density of magnetic impurity x is fixed to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

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 growth of the samples, there are additional defects which appear, namely, As antisites (As_{Ga}) (which substitute for the Ga sites) or Mn interstitials Mn_I . The existence of these defects is the main source of 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. Note that, in contrast to As_{Ga} , Mn_I also affects the density of magnetically active moments. Thus, to simplify the study, we will focus on the effect of tuning the density of carrier by maintaining constant the density of Mn^{2+} by varying the density of As_{Ga} . We would like to stress that the results which will be presented have a general character and are independent of the precise nature of the compensating defects. 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 tuning of the superexchange coupling,^{32,33} 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 neighbor coupling (see Fig. 5 in Ref. 23). This is clear from *ab initio* studies, where the couplings have been calculated without any adjustable parameters (tight-binding linear muffin-tin orbital calculations), and subsequently used in several publications.^{23,24,29}

In Fig. 4, using the SCLRPA in the same way as described in Sec. II, and using *ab initio* couplings, we calculate the predicted phase diagram for $\text{Ga}_{1-y}\text{As}_y\text{Mn}_{0.05}\text{As}$ (temperature-density of As antisites). By energy minimization, we have found a wide region of the phase diagram ($0.01 < y$

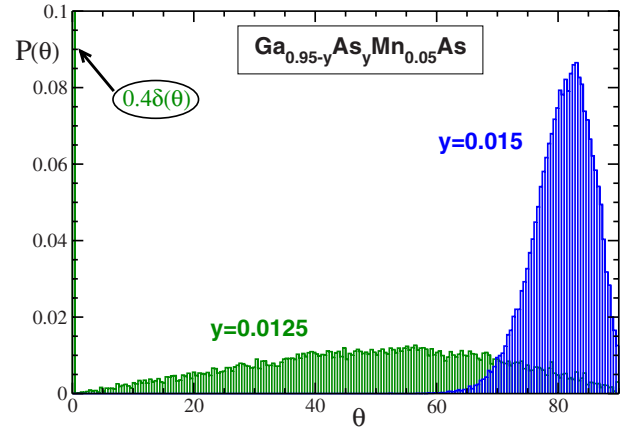


FIG. 5. (Color online) Distribution $P(\theta)$ of the canting angle of the nearest neighbor pairs in $\text{Ga}_{1-y}\text{As}_y\text{Mn}_{0.05}\text{As}$ for two different values of the concentration of As antisites, y . The density of Mn^{2+} is $x=0.05$.

< 0.0175) where the ground state is not fully ferromagnetic but canted. This is a consequence of the superexchange coupling J_{AF} , which 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 those of Monte Carlo simulations.²³ Although the nature of the phase was not discussed in that study, this validates the simplified treatment of the canting of the ground state. Note that, both Monte Carlo simulations and SCLRPA calculations 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. 29 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. 29). This instability in fact signals the occurrence of a new phase, which 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 nonsaturated. 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 $m(0)=1$ (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, $m(0) \geq x_{\text{eff}}/x=0.55$ 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 (δ 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 $\theta \approx 80^\circ$ is narrower 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 bulk magnetization (with a SQUID) is different from the magnetization expected from the determination of the Mn density from XRD measurements, assuming a fully polarized ferromagnet.^{18–22} 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 except if the temperature is too high.³⁴ In a recent study,²⁹ it was shown that one could explain the effect of different annealing treatments³⁵ by the existence and 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.^{36,37} 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.^{18,38} 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 reduced 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, which 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. 38 at small fields ($H=500$ Oe), we observe that the magnetization is about twice smaller for the 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, which must be at play once the coupling between nearest neighbors 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 reanalyzing the experimental data on the basis of the present work.

We should now stress that we have neglected three additional effects which may play an important role in specific samples. (i) Hydrogenation changes the couplings locally by forming local bridges near the H atom. This was argued to enhance the ferromagnetic coupling for neighbor spins in ZnCoO.³⁹ This effect was not taken into account here, and would reduce the effect of the superexchange coupling. (ii) The couplings used here are calculated *ab initio* within the coherent potential approximation (CPA) approach. Thus the

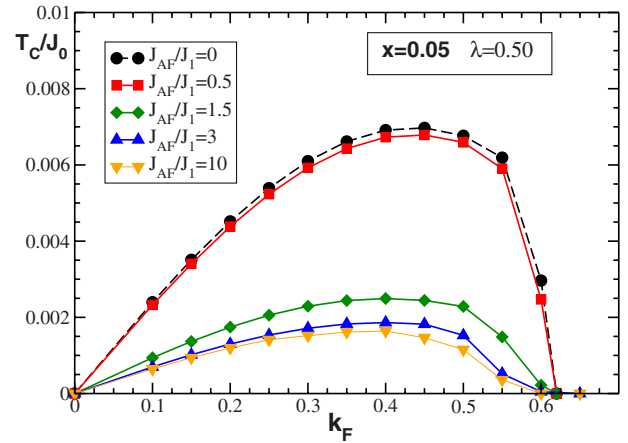


FIG. 6. (Color online) Curie temperature for the RKKY model as a function of k_F for different values of the superexchange coupling J_{AF} . The density of impurities is set to $x=0.05$ and the parameter $\lambda=0.50$. The calculations are performed on the fcc lattice.

effects of short-range correlations and cluster formation are not properly included. It has been shown that the formation of small clusters like trimers, quadrupers, etc., changes the magnetic couplings.⁴⁰ We note that these effects should be relevant at high enough concentrations and should lead to reorientations of the spins in these clusters. (iii) Here, the impurities were placed randomly with no correlations so that the number of dimers is fixed by the geometry of the lattice. In fact, forces between impurities have been reported⁴⁰ and may also lead to correlations in the impurity positions, or even formation of larger clusters.

IV. MODEL WITH RKKY OSCILLATIONS

In the last section we discuss another interesting case, the interplay between frustration resulting from RKKY couplings and the superexchange. In order to compare with previous studies, we assume the long-range couplings to be given by $J_{ij}=J_0 e^{-r/\lambda} F(k_F r)$, where $F(k_F r) = (k_F r) \cos(k_F r) / (r/r_0)^3$ and $r=|r_i-r_j|$, r_0 being the nearest neighbor distance. The parameter is an effective Fermi vector k_F which is determined by the density of carriers, $k_F = (3\pi^2 n_h)^{1/3}$ (in units of the inverse of the lattice spacing). Note that this study is also motivated by the fact that RKKY couplings are often used to study the ferromagnetism in diluted magnetic semiconductors,^{41,42} although it was shown that they are inappropriate.³⁰ In previous studies, it was argued, in particular, that the stability region for ferromagnetism was very narrow upon increasing J_{AF} ,³⁰ a point that was reaffirmed later using Monte Carlo simulations.²⁸ We now argue that the stability region is in fact wider because of unsaturated phases.

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. 30. 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. 30) 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 an almost 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. 28). 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 Carlo²⁸ and SCLRPA calculations³⁰ give the same results [see Figs. 1 and 4 of Ref. 30 and Fig. 1(c) of Ref. 28]. The effects of frustration are therefore properly handled by the SCLRPA, and thus it cannot be the source of the disagreement. In addition, the comparison between the Monte Carlo simulations of Ref. 23 and the modified SCLRPA results 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 which was missed before. It would be of interest to clarify this issue.

V. CONCLUSION

In conclusion, we have shown that the competition between nearest neighbor antiferromagnetic superexchange

coupling and long-range ferromagnetic couplings gives rise to a canted ferromagnetic phase in dilute magnets. We emphasize that short- and long-range competing interactions play quite different roles. In the latter 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 interimpurity 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 *ab initio* couplings. The existence of this phase provides a simple explanation for 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.

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