Why RKKY exchange integrals are inappropriate to describe ferromagnetism in diluted magnetic semiconductors.

Richard Bouzerar\textsuperscript{1§}, Georges Bouzerar\textsuperscript{2∗∗} and Timothy Ziman\textsuperscript{3¶}

\textsuperscript{1}Université de Picardie Jules Verne, 33 rue Saint-Leu, 80039 Amiens Cedex 01

\textsuperscript{2}Laboratoire Louis Néel 25 avenue des Martyrs, CNRS, B.P. 166 38042 Grenoble Cedex 09 France.

\textsuperscript{3}Institut Laue Langevin B.P. 156 38042 Grenoble France.

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\textsuperscript{§}email: richard.bouzerar@u-picardie.fr

\textsuperscript{∗∗}email: georges.bouzerar@grenoble.cnrs.fr

\textsuperscript{¶}and CNRS, email: ziman@ill.fr
Abstract

We calculate Curie temperatures and study the stability of ferromagnetism in diluted magnetic materials, taking as a model for the exchange between magnetic impurities a damped Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction and a short range term representing the effects of superexchange. To properly include effects of spin and thermal fluctuations as well as geometric disorder, we solve the effective Heisenberg Hamiltonian by means of a recently developed semi-analytical approach. This approach, “self-consistent local Random Phase Approximation (SC-LRPA)”, is explained. We show that previous mean-field treatments, which have been widely used in the literature, largely overestimate both the Curie temperatures and the stability of ferromagnetism as a function of carrier density. The discrepancy when compared to the current approach was that effects of frustration in RKKY oscillations had been strongly underestimated by such simple mean-field theories. We argue that the use, as is frequent, of a weakly-disordered RKKY exchange to model ferromagnetism in diluted III-V systems is inconsistent with the observation of ferromagnetism over a wide region of itinerant carrier densities. This may be puzzling when compared to the apparent success of calculations based on *ab-initio* estimates of the coupling; we propose a resolution to this issue by taking RKKY-like interactions between resonant states close to the Fermi level.
I. INTRODUCTION

After the discovery by Ohno et al. [1] that the doping by a small amount of magnetic impurities in GaAs could lead to relatively high Curie temperature in III-V semiconductors [2, 3], materials of this class have attracted considerable interest from both experimentalists and theoreticians. It is now accepted that the ferromagnetism is mediated by the “itinerant carriers” introduced after substitution of Ga, of nominal valence 3+, by Mn$^{2+}$. Among theories of the ferromagnetism in these materials, the simplest and most commonly used approach considers that the exchange between localized magnetic impurities is of standard RKKY type [4, 5]. The exchange integrals are, to second order in perturbation theory, quadratic with respect to the local coupling of the itinerant carrier to the localized magnetic impurity spin $J_{pd}$.

\[
J_{ij} = - \frac{J_{pd}^2}{\pi} \Im(\chi(R_{ij})) \tag{1}
\]

where $\chi$ is the magnetic susceptibility. The density of carriers enters in the magnetism via the dependence of the susceptibility but in the simplest approach the effects of disorder (multiple scattering of the itinerant carrier due to the magnetic impurities) are not taken into account. Once the effective magnetic Heisenberg Hamiltonian has been determined, it has commonly been solved by treating fluctuations within mean field theory, and disorder within a virtual crystal approximation (VCA), giving an approximation we shall refer to as (MF-VCA) [4, 5, 6, 7, 8] and is commonly referred to loosely as Zener Mean-Field theory. It has often been stated in the literature [9] that such an approach gives good account of the experimental situation in both III-V and II-VI doped semiconductors and that it can be relied on for quantitative prediction. This apparent consensus on the applicability of such a picture is, however, seriously in doubt, as we shall discuss.

From an experimental point of view, it is observed that the Curie temperatures are very sensitive to the method of preparation. Indeed the Curie temperatures often vary greatly when measured on samples as they are grown, and after they have been annealed. In the Zener Mean-Field Theory, the Curie temperature for fixed concentration of impurities varies with a simple power law ($T_C \propto \rho^{\frac{3}{2}}$) on the density of itinerant carriers. In reality, other aspects are important: such as configurational disorder, thermal fluctuations, and detailed compensating mechanisms [10, 11]. Calculations that treat the effects of band structure
realistically from first principles show that the RKKY description is inaccurate \cite{12, 13} in that the strength of interactions do not simply depend on distance, but also on lattice direction. Furthermore simple one-band model calculations of the exchange integrals treating the disorder by a Coherent Potential Approximation (CPA) has shown that the RKKY behaviour is in fact restricted to very small value of the ratio $J_{pd}/W$ \cite{14} where $W$ is the bandwidth of the carrier band. To higher order the oscillatory behaviour is strongly reduced. Calculations which, in addition, treat the effect of disorder on the itinerant carriers within CPA, show clearly that the exchange integrals (i) do not oscillate and (ii) are exponentially damped with distance \cite{15} as in the model calculations \cite{16}. A different approach was taken by Brey et al.\cite{17} who took a $k \cdot p$ description of the band structure with a non-local $J_{pd}$ coupling treated perturbatively. In this case the non-locality of the $J_{pd}$ coupling lead to suppression of oscillations. Another attempt \cite{18} to use a realistic (Slater-Koster) band structure with non-local couplings between p- and d-orbitals treated perturbatively, lead, however, to very different magnetic couplings which oscillated strongly. Both calculations included spin-orbit couplings but differed in their conclusions as to its importance. Recent work \cite{20} including Monte-Carlo simulations concluded that the effects of spin-orbit interactions on the Curie temperature are weak, unless the anisotropy induced is very large which seems unlikely for III-V semiconductors. Fiete et al. \cite{21} found effects of non-collinearity from spin-orbit coupling but they were significant at parameters corresponding to very low concentrations of dilute Ga(Mn)As.

Recent theoretical studies have shown that if the exchange integrals includes both the effects of disorder and realistic band structure, MF-VCA treatment of the Heisenberg model leads to larger Curie temperature than experimentally observed \cite{22}. In contrast, it was shown that using a self-consistent local Random Phase Approximation (SC-LRPA) it is possible to attain quantitatively accurate estimates of ferromagnetism in diluted magnetic semiconductors for annealed \cite{23} and partially annealed samples \cite{24}. This method will be made explicit in the next section, but the essential improvement over MF-VCA is that both disorder and transverse fluctuations are properly treated. For the disorder this is by avoiding any effective medium approximation and, instead, using sampling over randomly generated geometries. The tranverse fluctuations are included by preserving the rotational spin symmetry in the decoupling. The success of this semi-analytical approach is fully supported in the limit of large spin where classical Monte Carlo calculations can be performed \cite{25, 26}.
For finite spin we can compare only for the case of regular lattices, but by comparison to series expansions the Callen approach is known to give critical temperatures accurate to a few per cent even for couplings of short range. Note that, compared to Monte-Carlo simulations, the SC-LRPA has several advantages: it provides a direct expression for the Curie temperature and the computing cost is extremely low, allowing systematic examination of the space of couplings, and effects of cut-off and so forth. In addition it can include quantum fluctuations for finite spin $S$ for which Monte Carlo methods encounter problems of sign. The disadvantage of using “realistic” values of the exchange integrals taken from first principle calculations is that they are strongly material dependent, and do not provide a simple understanding of the parameters which control the exchange integrals. Thus, attempts to find simplified models where the exchange integrals between magnetic impurities would depend on a few physical parameters are still attractive and should be pursued.

The purpose of this paper is then to return to the simple RKKY approach whose validity has been argued on the basis of over-simplified calculation of the thermodynamics, but now to study the influence of disorder and transverse fluctuations using a more reliable calculation. Thus we shall examine the stability of ferromagnetism and Curie temperature for exchange integrals in the effective Heisenberg model of RKKY type. We will also, in the following, analyze both the effect of including a nearest-neighbour antiferromagnetic (AF) superexchange and damping of the exchange coupling. The damping mimics in a simple manner the effect of multiple scattering of the itinerant carriers over the magnetic impurities. As we have mentioned, while this corresponds to the average exchange in an approximation of weak coupling and weak disorder, it is not a completely general form for strong coupling.

The effective Heisenberg Hamiltonian reads,

$$H = -\sum_{ij} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j$$  \hspace{1cm} (2)

where $\mathbf{S}_i$ are quantum spins randomly distributed on a lattice, we denote $x$ the density of magnetic impurities. For comparison with Ga(Mn)As we take the lattice to be face centered cubic (fcc) corresponding to simple substitution of Mn on Ga sites. $J_{ij}$ is the exchange interaction coupling between two impurities located at site $i$ and $j$, it reads

$$J_{ij} = J_0 \exp\left(-\frac{r}{r_0}\right) \frac{(\sin(2k_F r) - 2k_F r \cos(2k_F r))}{(r/a)^4} + J_{ij}^{AF}$$  \hspace{1cm} (3)
where \( r = R_i - R_j \) is the distance between two impurities labeled \( i \) and \( j \). The first term is the damped RKKY exchange coupling, \( r_0 \) is the damping length. The second term is the direct antiferromagnetic superexchange interaction. For simplicity we restrict ourselves to short range SE term, \( J_{ij}^{AF} = J_{AF} \) if \( i \) and \( j \) are nearest neighbours (NN) and vanishes otherwise. We consider the diluted regime for magnetic impurities and itinerant carriers; thus we assume a spherical Fermi surface and \( k_F = (3\pi^2 n_c)^{1/3} \) where \( n_c \) is the density of carriers. Note that we shall restrict ourselves to the simplest RKKY form corresponding to the asymptotic form of a single parabolic band of carriers. Our model has four parameters: \( x, n_c, r_0 \), and \( J_{AF}/J_0 \). In the following calculations they are treated as being independent, although of course physically if we vary the doping \( x \) to model a series of compounds of different doping, the other parameters would be functions of \( x \). A derived parameter useful for comparison with experiment is the doping concentration per doping impurity \( \gamma = n_c/x \). In the simplest model of doping this would be 1, in practice it is usually much less and varies with sample history.

As we treat the disorder fully and thermal fluctuations accurately, we will show that previous treatments of the same Hamiltonian (Equation 2) by MF-VCA lead to wrong conclusions. Furthermore by understanding this simple model, we shall show that we can demonstrate the importance of qualitative aspects. In particular, the presence or absence of oscillations will be seen to be crucial to stabilize ferromagnetism at finite temperatures. In MF-VCA this was obscured by the simplified treatment. This may be significant in more complicated forms of model band structures, e.g. 6 band Kohn-Luttinger forms. In addition, in different \textit{ab-initio} treatments, for example local-density-approximation (LDA) or extensions such as LDA+U approaches, there are changes in the nature of the states close to the Fermi surface which will affect long-range oscillations of the exchange couplings and thus the region of stability of ferromagnetism.

One aspect we do not consider is the role of anisotropic couplings in spin-space coming from spin-orbit couplings. As we have noted above, there is some debate as to whether these make significant contributions to the exchange. We argue that for the Curie temperature this is not of primary importance especially as any spin anisotropy is random in direction. Of course spin-orbit effects must be considered for calculations of magnetic anisotropies. We will argue that for the Curie temperature, which is the subject of the present paper, what is much more significant is the bias in the sub-asymptotic form
of the isotropic RKKY-like couplings, coming from resonant effects. This will be explained in Section IV. The importance of such effects, rather than anisotropies, is supported by the success of the non-relativistic \textit{ab-initio} calculations which include the effects of hybridization and resonance.

II. METHOD

Let us now summarize the main steps of the approach we use. We define the following retarded Green’s function $G_{ij}(t)$ for localized spins at sites $i$ and $j$,

$$G_{ij}(t) = -i\theta(t) < [S_i^+(t); S_j^-(0)] >$$  \hspace{1cm} (4)

The important point is that we shall decouple in real space, and within a Random Phase Approximation, the equation of motion for the frequency-transformed Green’s functions $G_{ij}(\omega)$:

$$(\omega - h_i^{eff})G_{ij}(\omega) = 2\langle S_i^z \rangle \delta_{ij} - \left[ \langle S_i^z \rangle \sum_l J_{il} G_{lj}(\omega) \right]$$  \hspace{1cm} (5)

where $\langle S_i^z \rangle$ is the local magnetization at site $i$. For a given configuration of impurities and temperature, $\langle S_i^z \rangle$ should be determined self-consistently at each impurity site. $h_i^{eff}$ is the local effective field at site $i$,

$$h_i^{eff} = \sum_l J_{il} \langle S_l^z \rangle$$  \hspace{1cm} (6)

To determine $\langle S_i^z \rangle$ \textit{self-consistently} we use the Callen expression:

$$\langle S_i^z \rangle = \frac{(S - \Phi_i)(1 + \Phi_i)^{2S+1} + (S + 1 + \Phi_i)\Phi_i^{2S+1}}{(1 + \Phi_i)^{2S+1} - \Phi_i^{2S+1}}$$  \hspace{1cm} (7)

where $\Phi_i$ is a local effective boson occupation number.

$$\Phi_i = -\frac{1}{2\pi} \frac{1}{\langle S_i^z \rangle} \int_{-\infty}^{+\infty} \frac{ImG_{ii}(\omega)}{\exp(\omega/kT) - 1} d\omega$$  \hspace{1cm} (8)

At each temperature, (5),(7) and (8) form a closed set of equations. These may be solved to determine the temperature dependence of the local magnetization at each site and dynamical properties.
To determine the critical temperature $T_C$, we take the limit of vanishing $\langle S^z_i \rangle \to 0$ in the previous set of equations which leads to

$$\langle S^z_i \rangle = \frac{1}{3} S(S + 1) \frac{1}{\phi_i}$$

We obtain,

$$k_B T_C = \frac{1}{3 N_{\text{imp}}} S(S + 1) \sum_i \frac{1}{F_i}$$

The local quantity $F_i$ is

$$F_i \equiv \int_{-\infty}^{+\infty} \frac{A_{ii}(E)}{E} dE$$

where the reduced variable $E = \omega m$, $m$ is the averaged magnetization over impurity sites,

$$\lambda_i = \lim_{T \to T_C} \frac{\langle S^z_i \rangle}{m}$$

The local spectral function $A_{ii}(E)$ is,

$$A_{ii}(E) = -\frac{1}{2\pi} Im\left( \frac{G_{ii}(E)}{\lambda_i} \right)$$

We note that by the above argument, the dependence of $T_C$ on quantum spin $S$ is entirely in the factor $S(S + 1)$ in equation (10). For the magnetization in the ordered phase the dependence is more complex as equation (7) must be used at each stage of iteration.

In the following, to evaluate the effect of both disorder and thermal fluctuations beyond mean field theory we will compare our self-consistent local-RPA expression of the Curie temperature $T_C$ to the often-quoted MF-VCA expression.

$$T_C^{MF-VCA} = \frac{S(S + 1)}{3} x \sum_i N_i J(r_i)$$

$N_i$ (resp. $r_i$) is the number (resp. distance) of the $i^{th}$ nearest neighbour (summation over all the host crystal sites). We note that this expression is equivalent to equation 4 of ref.[5], and is the basis of the results of that paper. While in that paper sampling over different configurations of disorder was performed, the mean-field treatment of thermal fluctuations
Figure 1: (Color online) Curie temperature as a function of the carrier concentration (per impurity) \( \gamma \) for a fixed 5% concentration of impurities. The damping is \( r_0 = 2 \) lattice parameters and the superexchange contribution vanishes: \( J_{AF} = 0 \). The Curie temperature is averaged over 100 configurations of disorder for the self-consistent local RPA. Finite size corrections are seen to be small, by comparing points calculated for lattices of \( 16 \times 16 \times 16 \) (crosses) or \( 22 \times 22 \times 22 \) (squares). For comparison we show the results of MF-VCA. The points (a-d) are indicated for reference to the following Figure 2.

effectively replaced the complex geometry of random substitution by an homogeneous crystal lattice. In contrast, the SC-LRPA simultaneously treats the effects of random geometry explicitly and includes thermal fluctuations beyond molecular field theory. In this case the disordered medium can no longer be reduced to an effective crystal. In the following we will be able to see quantitatively how inaccurate such mean-field lattice approximations may be.
Figure 2: (Color online) Distribution of the local fields for $\gamma = 0.025$, 0.1, 0.2 and 0.3 and the same parameters shown in Figure 1. The distributions correspond respectively to point (a), (b), (c) and (d) in Figure 1.

III. NUMERICAL RESULTS

A. Results for Self-Consistent local RPA for the Curie Temperature as a function of carrier density

In this section we present self-consistent local-RPA calculations for the RKKY interaction for parameters, in particular concentration of interest for III-V semiconductors. We shall compare to the results of MF-VCA treatment and attempt to understand the differences when they appear. We first consider the RKKY interaction in the absence of AF superexchange term, i.e. we fix $J_{AF} = 0$, and assume strong damping, $r_0 = 2$, in units of the cubic lattice parameter. The effects of varying these two parameters will be considered in the following sections. In Figure 1 we have plotted the Curie temperature for fixed concentration $x = 5\%$ of Mn impurities as a function of $\gamma = n_c/x$ for both the self-consistent local-RPA and MF-VCA treatment.

Note that for the SC-LRPA an average is made over approximately 100 configurations of disorder for each set of parameters. For each configuration the magnetic impurities location
Figure 3: (Color online) Variation of the exchange constant with distance between impurities for $\gamma = 0.025$, 0.1, 0.2 and 0.3 and the same parameters shown in Figure 1. $r$ is in units of the lattice parameter of the fcc lattice.

have been randomly generated on a $L \times L \times L$ fcc lattice, where $L \geq 16$. In order to show that finite size effects are negligible we have plotted in the same figure the calculations corresponding respectively to 744 and 1987 magnetic impurities distributed randomly over the fcc lattice. By comparing results from the different sizes it is seen that finite size effects are negligible for the sizes considered here. It is seen that MF-VCA systematically overestimates the Curie temperature, and agreement between MF-VCA and SC-LRPA is only observed for very low carrier densities. Whilst the MF-VCA curve is monotonically increasing, the SC-LRPA curve exhibits a maximum at relatively low density of carriers $n_c \approx 0.1x$ and ferromagnetism disappears for $\gamma \geq 0.25$. Thus the better treatment of disorder and thermal fluctuations leads to a narrow region of stability for ferromagnetism. What is the reason for this? In the RKKY form of the exchange integrals for sufficiently large distances the exchange integrals become antiferromagnetic, leading to frustration of the ferromagnetism. The effects of this frustration are strongly underestimated within a VCA mean field treatment. This is illustrated in Fig. 2, where we plot the distribution $P(h^{\text{eff}})$ of the local fields at $T = 0 K$. For a given configuration of disorder the local field
at site $i$ is

$$h_i^{\text{eff}} = \sum_j J_{ij} \langle S_j^z \rangle$$  \hspace{1cm} (15)$$

In Figure 2, the distributions are plotted for $\gamma = 0.025, 0.1, 0.2, 0.3$, corresponding to (a), (b), (c) and (d) respectively, in Figure 1. In cases (a) and (b) we observe that the distribution of local fields is completely ferromagnetic: there is no antiferromagnetic part $P(h) = 0$ for $h \leq 0$. The distribution is very narrow in the case (a), this explains why in Figure MF-VCA and SC-LRPA are very close. The distribution broadens asymmetrically in case (b) and the average value $\langle h^{\text{eff}} \rangle = \int_{-\infty}^{+\infty} h P(h)$ increases, leading to higher $T_C$ in MF-VCA than in SC-LRPA. For higher density of carriers, (c) and (d), we observe a negative tail in the local field distribution. Note that, although this tail is very small, its effect on the Curie temperature is dramatic. Figures 1 and 2 show clearly that the ferromagnetism in diluted systems is very sensitive to frustration effects coming from the exchange couplings at large distances. In a VCA the effect of the tail is lost in the average over different sites of the lattice. In the SC-LRPA we see clearly an instability to bulk uniaxial ferromagnetism. It may seem surprising that a relatively small proportion of sites with local negative molecular fields can suppress the ferromagnetism; however by analogy to the response of a ferromagnet to random external fields we can make an Imry-Ma argument [31] that in three dimensions the rotational symmetry allows for complete break-up of ferromagnetism even for a small proportion of random fields. Thus it seems likely that the resultant state is either paramagnetic or of spin-glass type.

In Figure 3 we mark the variation of the Heisenberg exchanges (normalized to omit the $\frac{1}{r^3}$ decrease) as a function of distance for the same parameters $\gamma = 0.025, 0.1, 0.2, 0.3$, corresponding to (a), (b), (c) and (d) respectively, in Figure 1. Unlike the distribution of local fields, this gives no obvious clue as to the instability of ferromagnetism. This is perhaps surprising if we remark that the average distance between magnetic impurities at this concentration is only slightly larger ($r=1.07$ in the units of Fig. 3) than the lattice parameter of the fcc lattice.
B. Effect of the exponential cut-off.

We now discuss the effect of the exponential cut-off on the domain of ferromagnetic stability. The use of such a cut-off dates back to deGennes [32] who calculated the average coupling in weakly disordered systems. There has been much discussion of whether the difference between this average coupling and the undamped typical couplings that may determine the characteristic scale for spin-glass behaviour in the very dilute limit but for ferromagnetic transitions this has been assumed, without absolute proof to be appropriate. In Figure 4 we have plotted the Curie temperature as a function of the carrier density for different values of $r_0$: $r_0 = 2; 5; 10; \infty$. $J_{AF} = 0$ and the system size is $16 \times 16 \times 16$.

![Figure 4: (Color online) Curie temperature (self-consistent local-RPA) as a function of the carrier density for different values of $r_0$: $r_0 = 2; 5; 10; \infty$. $J_{AF} = 0$ and the system size is $16 \times 16 \times 16$.](image)

We defer the question of the influence of superexchange $J_{AF} = 0$ and consider a fixed density of magnetic impurities $x = 5\%$. The SC-LRPA shows that in the undamped RKKY limit the region of stability is extremely narrow. We observe that the ferromagnetic stability domain increases significantly when $r_0$ is relatively small, i.e., of the order of the average distance between impurities. For instance for $r_0 = 2$, the region of stability of ferromagnetism is five times broader than in the pure undamped case. This illustrates the strong influence...
Figure 5: (Color online) SC-LRPA Curie temperature as a function of $J_{AF}$ (normalized by the nearest-neighbour RKKY interaction $J_1$) for different carrier densities. The magnetic impurity concentration (5%) and RKKY damping are fixed ($r_0 = 2$). In inset (note that the axes are the same) we compare the Curie temperature calculated within MF-VCA.

of the oscillating tail on ferromagnetism. This figure also shows clearly that the undamped RKKY exchange integrals often used in model calculations can not explain the ferromagnetism often observed in III-V diluted materials for the carrier densities $n_c \approx x$ or $\gamma \approx 1$ for annealed samples which exhibit the highest $T_C$. Even for strong disorder ($r_0=2$), ferromagnetism is not possible for $n_c \geq 0.25x$. From this figure and Figure 11, we conclude that the use of RKKY exchange integrals often used in simplified theories is questionable and its often-cited “success” is due to the unrealistic MF-VCA approximation which overestimates the Curie temperature and misses the instability seen here. The difference with the agreement with first principle calculations is that there, as in model calculations with the disorder treated in CPA[16], not only are the exchange integrals are strongly damped but, in addition, the anti-ferromagnetic contributions are almost completely suppressed[22].
C. Influence of the superexchange.

Let us now turn to the influence on ferromagnetism of a short-range superexchange contribution. This is important for understanding materials as it is essentially independent of the longer range RKKY exchange and sensitive to local effects. In mean field theories a very large ferromagnetic coupling at small distances has been used to argue, for example in doped Ga(Mn)N, for potentially high Curie temperatures. For simplicity, and because superexchange are short range, we consider the case where superexchange modifies nearest-neighbour magnetic impurities only.

In Figure 5 we have plotted the dependence of the SC-LRPA Curie temperature as a function of $J_{AF}$ for different carrier density and fixed magnetic impurity concentration and $r_0=2$. First, in contrast to the mean-field treatment (see inset), we observe that in the region dominated by the RKKY term (nearest-neighbour ferromagnetic) that the Curie temperature is insensitive to $J_{AF}$. This can be understood as following: since the system is diluted, we might expect that $T_C$ should be controlled mainly by exchange integrals for typical distances between magnetic impurities. In fact, as we have discussed, it is not sufficient to consider only these distances: the long-range tail is also important. Our local approximation builds in all these features. In contrast the MF-VCA, in which the Curie temperature is linear in all couplings, could not capture these geometric effects. In our calculations, when the antiferromagnetic superexchange becomes dominant the Curie temperature vanishes abruptly. The reason for this instability is that the system becomes frustrated, but now from the short-range exchange, not the long range RKKY oscillations controlling the disappearance observed in Figures 1 and 4.

D. Variation of $T_C$ with concentration

Let us now discuss the influence of the impurity concentration on the Curie temperature. For simplicity, we consider only the case $J_{AF} = 0$. From the previous section, the results change little if we add a term of short-range superexchange, provided its strength is insufficient to give the instability of Figure 5. In Figure 6 we have plotted $T_C$ as a function of $x$ for different values of the carrier density per impurity $n_c = \gamma x$. For MF-VCA calculations we observe that $T_C$ increases monotonically with the density of both magnetic impurities.
Figure 6: (Color online) SC-LRPA calculations of the Curie temperature as a function of the magnetic impurity concentration and for different carrier density. $J_{AF} = 0$ and $r_0=2$. In the inset, which has the same axes, the MF-VCA Curie temperatures are plotted for the same parameters.

and itinerant carriers. The SC-LRPA, however, predicts much smaller Curie temperatures than the MF-VCA results and that $T_C$ systematically decreases with increasing the carrier density $\gamma$. In contrast to MF-VCA, we also observe that as we increase $\gamma$, a maximum in $T_C$ appears. The location of this maximum is shifted to lower impurity concentration as the carrier density per magnetic impurity increases. For sufficiently high densities ($\gamma \geq 0.25$) of carriers we observe complete suppression of ferromagnetism. We have already discussed previously the reason for this: the frustrating effect of the RKKY “tail”. Thus again, we see that better treatment of disorder and fluctuations, give very different trends from the predictions of simple effective medium theories. The immediate question raised is then, how can we understand, from a simple model point of view, the observed ferromagnetism? For example that observed at carrier densities where from Figure 6 we now predict $T_C = 0$ and we argue that that predictions from MF-VCA were invalid. This will be the subject of the next section.
IV. THE SOLUTION: GENERALIZATION OF RKKY TO TREAT RESONANCES

We have seen that simple RKKY forms of magnetic interaction lead to a region for stable ferromagnetic order that is very narrow. How can the experimental results be reconciled with this? One approach, which seems to be successful for the III-V is to abandon a simple model approach and use *ab-initio* results for the effective Hamiltonian. This works, as mentioned in the introduction, as the couplings do not display the changes of sign characteristic of the RKKY interaction. It is interesting to look more carefully at the physical reason behind this. While the non-spherical geometry of the Fermi surface of the host material affects the oscillations in exchange coupling it will not suppress the changes of sign. The significant point seems to be, in for example Ga(Mn)As, that the states at the top of the valence band
are strongly hybridized with the impurity states. This gives rise to resonant peaks in the
density of states close to the Fermi level. In consequence the states contributing to magnetic
exchange must be modified by the hybridization between impurity and host bands. In this
case the appropriate model is that of interacting resonances \[33\,34\]. As shown by Caroli and
others \[33\,35\,36\,37\], the simplest way to include resonant effects in order to generalize
the regular RKKY expression, is to replace the term \(J_{pd}\) by a frequency-dependent element
\(\frac{|V_{pd}|^2}{(\omega-\omega_+)}\). In perturbation theory, the interactions between two resonant impurities can be
separated into two contributions, the first an “RKKY-like” term generated by particle-hole
exchange between the two spins. While asymptotically the standard RKKY interaction
is recovered, there is an important “pre-asymptotic” regime where subleading corrections
to the exchange change the qualitative form of the interaction. In particular in the first
“RKKY-like” contribution there is a ferromagnetic “bias”, ie in the pre-asymptotic region,
the interaction oscillates with distance with period \(2k_f\) around a smoothly decaying value and
does not change sign. The second contribution is the antiferromagnetic superexchange term
coming from particle-particle excitations, ie with intermediate states where the moments
are unoccupied. For a perfect gas the superexchange term compensates the ferromagnetic
bias of the RKKY oscillation giving a a total exchange term that oscillates in sign \[36\]. With
the effects of disorder and interaction the compensation need not occur \[36\] and this may
provide a qualitative explanation of the form of the exchange interactions from \textit{ab-initio}
approaches. Because the \textit{ab-initio} calculations are non-perturbative, they do not allow an
easy separation into RKKY and superexchange contributions. As the \textit{ab-initio} calculations
include interactions and, to some extent, the disorder, the similarity between the forms seen
there ( see for example Fig. 3 of Ref. \[22\]) and the perturbative RKKY-like contribution
to interactions between resonances supports the idea that it makes sense to exclude the
superexchange contribution other than the short-range term. Thus an appropriate model
Hamiltonian may be to take the perturbative “RKKY-like” interaction between resonances.
While this will be more fully investigated in future work, we illustrate how this can restore
ferromagnetism in a range where, as we have noted above, standard RKKY interactions fails
to predict ferromagnetism. In Figure 7 we show results of calculation taking interactions
which include the resonant RKKY-like contribution \[36\,37\]. The resonances are defined by
a \(k\)-vector \(k_i\), i.e. \(E_{\text{imp}} = \hbar k_i^2 \frac{2m}{m}\) which can be varied continuously relative to a spherical Fermi
surface of fixed Fermi Energy \(E_f = \hbar k_f^2 \frac{2m}{m}\). Perturbative calculations between narrow resonant
levels interacting by free electrons give an RKKY-like contribution to the exchange, in momentum space,

\[ J_{RKKY}[q] = \frac{J_0}{4\pi q} \left[ \log \left( \frac{2k_f - k_i}{q} \right) - \log \left( \frac{2k_f + k_i}{q} \right) - \frac{k_i}{k_f^2 - k_i^2} \log \left( \frac{\left( \frac{k_f - k_i}{q} \right)^2}{\left( \frac{k_f + k_i}{q} \right)^2 - k_i^2} \right) \right] \tag{16} \]

Interactions in real space are obtained by Fourier transform of this analytical expression and an exponential damping term to make clear comparison with the results for standard RKKY (see Figure inset, for the couplings as a function of distance). We chose the value of doping \( \gamma = 0.3 \) and the same damping \( r_0 = 2 \), where after Figure (see point (d) of that figure) there was no ferromagnetism for well developed moments. It is seen that for resonances well below the Fermi level there is no ferromagnetism, but as the resonance level increases ferromagnetism appears, and the Curie temperature rises rapidly as the impurity approaches the Fermi level. This is clearly a simplified calculation, and the divergence for \( k_i \to k_f \) should be suppressed if superexchange is fully included. In addition we assume a moment which is fixed while this will, in fact, decrease as the resonance approaches the Fermi level. Nevertheless it illustrates the point that it is the resonant nature of the exchange that can resolve the apparent contradiction of observation, both from experiment and calculation based on \textit{ab-initio} approach, of ferromagnetism at doping levels approaching large values of \( \gamma \). We note that in the sense of the underlying model to take, this agrees with previous work emphasizing the influence of proximity to the Fermi level of the resonant states on Curie temperature. Our calculations of the ferromagnetism are different, however, and, we argue, more precise.

\section{V. CONCLUSIONS}

To conclude, we have studied the effects of both transverse fluctuations, and disorder, on ferromagnetism for diluted Heisenberg models assuming an RKKY type for the exchange integrals. We have shown that previous MF-VCA treatments are inappropriate to study ferromagnetism in diluted magnetic systems leading to strong over-estimates of Curie Temperatures. The apparent success of theories starting from RKKY couplings, which are often
cited in the literature as being “qualitatively correct”, is in fact due to the oversimplified approximations (MF-VCA) used to treat the effective Heisenberg model. We have seen from Figure 1 that there is agreement only in the limit of extremely small carrier density. The long-range tail of the RKKY interaction destabilizes ferromagnetism over all but very narrow ranges of parameter values. Even damping this tail is insufficient to explain ferromagnetism at the high doping densities of the materials showing the highest Curie temperature. For experimentally interesting densities, we have seen that the randomness is essential, as seen clearly if Fig. 2. Thus including the random geometry and transverse spin fluctuations, as we do, is much more significant than adding corrections to the continuum version of mean-field theory coming from the lattice version [5].

It is appealing to have a simple phenomenological picture of ferromagnetism of diluted magnetic semiconductors rather than having to rely on ab-initio calculations of exchange which must be performed material by material and where the underlying physics may be obscured by the fact that several aspects (hybridization, band structure, correlations, disorder...) are included but it is not easy to separate their effects individually. While we have shown that a picture of magnetic moments interacting with standard RKKY interactions is not compatible with experiments, we argue that the generalization to include the resonant nature of the exchange may be the correct phenomenology.

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