Carrier-induced ferromagnetism in concentrated and diluted local-moment systems

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For modeling the magnetic properties of concentrated and diluted magnetic semiconductors, we use the Kondo-lattice model. The magnetic phase diagram is derived by inspecting the static susceptibility of itinerant band electrons, which are exchange coupled to localized magnetic moments. It turns out that rather low band occupations favor a ferromagnetic ordering of the local moment systems due to an indirect coupling mediated by a spin polarization of the itinerant charge carriers. The disorder in diluted systems is treated by adding a CPA-type concept to the theory. For almost all moment concentrations x, ferromagnetism is possible, however, only for carrier concentrations n distinctly smaller than x. The charge carrier compensation in real magnetic semiconductors (in $Ga_{1-x}Mn_xAs$ by, e.g., antisites) seems to be a necessary condition for getting carrier induced ferromagnetism.

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I. INTRODUCTION

The exciting research field "*spintronics*" refers to new phenomena of electronic transport, for which the electron spin plays a decisive role, in contrast to conventional electronics for which the electron spin is practically irrelevant. For a full exploitation of spintronics, one should have materials that are simultaneously semiconducting and ferromagnetic. That is the reason for the intensive effort that has been focused on the search for magnetic semiconductors with high Curie temperatures. It is to the merit of Ohno and co-workers[1,2] to reach a T_C of up to 110 K in $Ga_{1-x}Mn_xAs$ and to demonstrate the electric control of T_C by means of a gate voltage.[3] (Even larger T_C values have been observed for annealed multilayers.[4]) Intense experimental as well as theoretical research on the outstanding phenomena associated with the interplay between ferromagnetic cooperative features and semiconducting properties is currently going on.[5] It is the important challenge of materials science to understand the ferromagnetism in compounds such as $Ga_{1-x}Mn_xAs$, and to find out the conditions for Curie temperatures T_C sufficiently exceeding room temperature. This paper shall contribute to the fundamentals of ferromagnetism in diluted local-moment systems.

It is commonly accepted[5] that the *(ferromagnetic) Kondo-lattice model (KLM)*, certainly better denoted as *s-f* or *s-d model* or, in its strong-coupling regime, as *double exchange model*, represents a good starting point for the description of the so-called local-moment magnetism. To this class of magnetic materials belong the classical magnetic semiconductors (insulators) such as the Eu chalcogenides EuO, EuS, EuTe,[6] which today are classified as "*concentrated*" magnetic semiconductors. Other representatives are the local-moment metals Gd, Dy, Tb, etc., as well as $Eu_{1-x}Gd_xS$, etc., for which magnetic and electrical properties are provoked by two different electronic subsystems. Strictly localized $4f$ electrons of the rare earth ion provide the magnetic moment while itinerant $5d/6s$ electrons take care of the electrical conductivity. These local-moment systems reveal an exceptionally rich variety of physical properties with basic ingredients being the electronic correlations and spin ordering. Thereby, an interband exchange between the local moments and the itinerant conduction electrons appears to play a dominant role, in particular, as far as the magnetic and magneto-optic properties are concerned.

The same holds for the already mentioned *diluted magnetic semiconductors (DMS)*. The implantation of Mn^{2+} ions in the prototypical semiconductor GaAs provides local moments $\left(S=\frac{5}{2}\right)$ which decisively influence the electronic GaAs states giving them, e.g., an extraordinary temperature dependence. Furthermore, each divalent Mn ion creates in principle one valence band hole. The temperature dependence of the band states induced by exchange coupling to the local-moment system is a well-known feature of the "*concentrated*" ferromagnetic semiconductors. Striking consequences of this special temperature dependence are the "*redshift*" of the optical absorption edge[6] and the metal-insulator transition in Eu-rich EuO.[7,8] The responsible exchange interaction appears to be decisive for the physics of the DMS, too. It creates the ferromagnetism in these materials. An important question is whether and how the disorder of the localized magnetic (Mn^{2+}) moments influences the magnetic stability. With respect to the main goal, namely, reaching room temperature ferromagnetism, the disorder aspect must be considered as a central point to clarify.

The natural precondition for an understanding of the "*diluted*" ferromagnetic semiconductors is to have understood the "*concentrated*" counterparts. From a theoretical point of view, that means to find a convincing (approximate) solution of the (ferromagnetic) KLM.[9–12] The general solution of the sophisticated many-body problem provoked by KLM is not yet available. The model describes the mutual influence of two well-defined electronic subsystems, localized magnetic moments and itinerant band electrons. It turns out to be a nontrivial challenge to treat both subsystems simultaneously on the same theoretical level. To our information, such a theory does not yet exist. It is the aim of this paper to propose a new way to approach this problem.

The second step is to introduce disorder of the localized magnetic moments by dilution and to inspect its influence on the magnetic stability.[13–15] Does the disorder weaken or even strengthen the ferromagnetism? How can we understand the fact that surprisingly low moment concentrations and carrier densities are able to mediate a ferromagnetic ordering in diluted magnetic semiconductors $Ga_{1-x}Mn_xAs$. The final goal is to work out the prerequisites for room temperature ferromagnetism in diluted magnetic semiconductors. We therefore, derive the magnetic phase diagram of a diluted Kondo-lattice (concentration x) in terms of model parameters such as x, the carrier concentration $n \leqslant x$, and the exchange coupling J. For this purpose, we introduce in the next section the KLM and a proposal for its electronic self-energy. The concept of disorder is developed in Sec. III, while the magnetic phase diagram [Curie temperature $T_C = T_C(x,n,J)$] is read off from the singularities of the paramagnetic susceptibility (Sec. IV). The results are discussed in Sec. V.

II. KONDO-LATTICE MODEL

The (ferromagnetic) Kondo-lattice model is today certainly one of the most frequently applied models in solid state theory, because of its great variety of potential applications to technologically promising topics in the wide field of collective magnetism. It refers to magnetic materials that get their magnetic properties from a system of localized magnetic moments being indirectly coupled via interband exchange to itinerant conduction electrons. Many characteristic features of such materials can be traced back to this interband exchange. The respective model Hamiltonian,[9,10]

$$H = H_s + H_{sf}, \tag{1}$$

describes the interaction of itinerant band electrons in a homogeneous magnetic field B (μ_B is the Bohr magneton),

$$H_s = \sum_{ij\sigma} (T_{ij} - z_\sigma \mu_B B \delta_{ij}) c_{i\sigma}^\dagger c_{j\sigma} \tag{2}$$

and localized magnetic moments (spins $\mathbf{S}_i$) via an intraatomic exchange,

$$H_{sf} = -J \sum_j \sigma_j \cdot \mathbf{S}_j = -\frac{1}{2} J \sum_{j\sigma} (z_\sigma S_j^z n_{j\sigma} + S_j^{-\sigma} c_{j\sigma}^\dagger c_{j-\sigma}) \tag{3}$$

without any direct exchange interaction between the localized spins. $c_{j\sigma}^\dagger$ ($c_{j\sigma}$) is the creation (annihilation) operator for a Wannier electron with spin σ ($\sigma = \uparrow, \downarrow$) at site $\mathbf{R}_j$ ($n_{j\sigma} = c_{j\sigma}^\dagger c_{j\sigma}$; $z_\sigma = \delta_{\sigma\uparrow} - \delta_{\sigma\downarrow}$; $S_j^\sigma = S_j^x + \mathrm{i} z_\sigma S_j^y$). J is the exchange coupling and T_{ij} the hopping integral. The latter is connected by Fourier transformation to the Bloch energy $\epsilon(\mathbf{k})$:

$$T_{ij} = \frac{1}{N} \sum_{\mathbf{k}} \epsilon(\mathbf{k}) e^{\mathrm{i}\mathbf{k}\cdot(\mathbf{R}_i - \mathbf{R}_j)}. \tag{4}$$

In spite of its simple structure, the model Hamiltonian (1) provokes a rather sophisticated many-body problem, which, at least for the general case, could not be solved exactly up to now. One of the main challenging questions is whether or not and under what conditions the interband exchange J may cause a collective (ferromagnetic) ordering of the coupled local-moment/itinerant electron system. Conventional second-order perturbation theory predicts an indirect Heisenberg exchange [Rudermann-Kittel-Kasuya-Yoshida (RKKY)] between the local moments. Approximate statistical mechanics of the resulting Heisenberg model, e.g., in the framework of the Tyablikov method,[16] indeed predicts ferromagnetism, but only for very low band occupations $n = (1/N)\Sigma_{j\sigma}\langle n_{j\sigma}\rangle$ (Ref. 10). A *modified* RKKY theory presented in Ref. 10, which takes into account higher order terms of the induced conduction electron spin polarization by a mapping of the *s-f* interaction (3) on an effective Heisenberg-Hamiltonian, results in a magnetic phase diagram with respect to the coupling strength J and the band occupation n. To our information, however, there does not exist a complete theory that treats the electronic part and the magnetic moment part of the KLM on the same level and in the same theoretical framework. Admittedly, this indeed appears to be a rather involved task. Very often, only the electronic problem is investigated while the local moment magnetization is phenomenologically simulated by a Brillouin function.[9,17,18] Such procedure presumes ferromagnetism, that by no means is always valid, without deriving it self-consistently within the KLM.

The electronic part of the many-body problem is solved as soon as the single-electron Green function $G_{\mathbf{k}\sigma}(E)$ is available or, equivalently, the electronic self-energy $M_\sigma(E)$,

$$G_{\mathbf{k}\sigma}(E) = \frac{\hbar}{E - \epsilon(\mathbf{k}) + z_\sigma \mu_B B - M_\sigma(E)}. \tag{5}$$

For simplicity, we assume from the very beginning a wave-vector independent self-energy. A $\mathbf{k}$ dependence of the self-energy would be mainly due to magnon energies $\hbar\omega(\mathbf{k})$ appearing as a consequence of magnon emission and absorption processes by the band electron.[12] However, the neglect of a direct Heisenberg exchange between the localized spins in the KLM (1) can be interpreted as the $\hbar\omega(\mathbf{k}) \to 0$ limit. In a previous paper,[17] we have developed a theory for the electronic self-energy, which fulfills, in the low carrier-density limit ($n \to 0$), all the known exact limiting cases:

$$M_\sigma(E) = -\frac{1}{2} J z_\sigma \langle S^z\rangle + \frac{1}{4} J^2 \frac{a_\sigma G_0\left(E - \frac{1}{2} J z_\sigma \langle S^z\rangle - z_\sigma \mu_B B\right)}{1 - b_\sigma G_0\left(E - \frac{1}{2} J z_\sigma \langle S^z\rangle - z_\sigma \mu_B B\right)}. \tag{6}$$

An extensive discussion of the reliability of this self-energy can be found in the above mentioned paper.[17] a_σ, b_σ are parameters which are fixed by rigorous high-energy expansions to fulfill the first four spectral moments,

$$a_\sigma = S(S+1) - z_\sigma \langle S^z\rangle (z_\sigma \langle S^z\rangle + 1), \quad b_\sigma = b_{-\sigma} = \frac{J}{2}, \tag{7}$$

$G_0(E)$ is the "*free*" propagator,

$$G_0(E) = \frac{1}{N}\sum_{\mathbf{k}} \frac{1}{E - \epsilon(\mathbf{k})}. \tag{8}$$

Since Eq. (6) is exact for a maximum number of special cases in the low-density limit, it should represent a reasonable starting point for the description of ferromagnetic semiconductors, which, by definition, is restricted to low densities of itinerant charge carriers. $M_\sigma(E)$ is the electronic self-energy for the "*concentrated*" (periodic) Kondo lattice. In the next section, we propose how to model the disorder of the magnetic moments in diluted ferromagnetic semiconductors.

III. ELECTRONIC SELF-ENERGY OF THE DILUTED SYSTEM

We consider a binary alloy of constituents α (concentration $1-x$) and β (concentration x). α symbolizes nonmagnetic sites (Ga^{3+}), while site β carries a magnetic moment (Mn^{2+} ion) being exchange coupled via (3) to the itinerant charge carriers. The atomic level of α sites is in the presence of a magnetic field B,

$$\epsilon_{\alpha\sigma} = T_0 - z_\sigma \mu_B B. \tag{9}$$

On β sites, however, the local interband exchange H_{sf} (3) acts on the charge carriers. That is accounted for by a "*dynamic*" atomic energy level incorporating the self-energy $M_\sigma(E)$ (6),

$$\epsilon_{\beta\sigma} = T_0 + M_\sigma(E) - z_\sigma \mu_B B. \tag{10}$$

We consider the charge carriers in the "*diluted*" Kondo lattice as a system of particles propagating in the above-defined fictitious binary $\alpha\beta$ alloy, thereby neglecting a Coulomb disorder potential which might be important in some circumstances[13] (e.g., metal-insulator transition). The single-particle properties can then be derived from the propagator

$$R_\sigma(E) = \int_{-\infty}^{+\infty} \mathrm{d}\omega \frac{\rho_0(\omega)}{E - \omega - \Sigma_\sigma(E)}, \tag{11}$$

where $\Sigma_\sigma(E)$ is now the electronic self-energy in the diluted system and $\rho_0(x)$ the Bloch-density of states of the noninteracting carriers. For the determination of the decisive self-energy we use a standard CPA formalism,[19] i.e., this quantity is determined by the CPA equation,

$$0 = (1-x)\frac{-z_\sigma \mu_B B - \Sigma_\sigma(E)}{1 - R_\sigma(E)(-z_\sigma \mu_B B - \Sigma_\sigma(E))} + x\frac{M_\sigma(E) - z_\sigma \mu_B B - \Sigma_\sigma(E)}{1 - R_\sigma(E)(M_\sigma(E) - z_\sigma \mu_B B - \Sigma_\sigma(E))}. \tag{12}$$

The limiting cases $x=0$ [$\Sigma_\sigma(E) = -z_\sigma \mu_B B$] and $x=1$ ["*concentrated*" KLM with $\Sigma_\sigma(E) = M_\sigma(E) - z_\sigma \mu_B B$] are obviously fulfilled.

The configurational averaging, inherent in CPA, takes care for translational symmetry and therewith for site-independent average spin-dependent occupation numbers,

$$\langle n_\sigma \rangle = \int_{-\infty}^{+\infty} \mathrm{d}E \frac{\rho_\sigma(E)}{e^{\beta(E-\mu)} + 1} \equiv \int_{-\infty}^{+\infty} \mathrm{d}E\, f_-(E)\rho_\sigma(E), \tag{13}$$

$f_-(E)$ is the Fermi function, μ is the chemical potential, and $\rho_\sigma(E)$ is the quasiparticle density of states of the *interacting* particle system,

$$\rho_\sigma(E) = -\frac{1}{\pi}\mathrm{Im}\, R_\sigma(E). \tag{14}$$

In the special case of a paramagnetic system and $B \to 0^+$ ("pm") the density of states reads

$$\rho_{\mathrm{pm}}(E) = -\frac{1}{\pi}\mathrm{Im}\, R_{\mathrm{pm}}(E) \tag{15}$$

with

$$R_{\mathrm{pm}}(E) = \int_{-\infty}^{+\infty} \mathrm{d}\omega \frac{\rho_0(\omega)}{E - \omega - \Sigma_{\mathrm{pm}}(E)}. \tag{16}$$

The paramagnetic self-energy obeys the CPA equation (12) in the following form:

$$0 = (1-x)\frac{-\Sigma_{\mathrm{pm}}(E)}{1 + R_{\mathrm{pm}}(E)\Sigma_{\mathrm{pm}}(E)} + x\frac{M_{\mathrm{pm}}(E) - \Sigma_{\mathrm{pm}}(E)}{1 - R_{\mathrm{pm}}(E)(M_{\mathrm{pm}}(E) - \Sigma_{\mathrm{pm}}(E))}. \tag{17}$$

The self-energy for the paramagnetic phase of the "*concentrated*" KLM (6) becomes in view of Eq. (7) especially simple,

$$M_{\mathrm{pm}}(E) = \frac{1}{4}J^2 \frac{S(S+1)G_0(E)}{1 - \frac{1}{2}JG_0(E)}. \tag{18}$$

We need these expressions when calculating, in the next section, the paramagnetic susceptibility of the itinerant charge carriers.

IV. STATIC MAGNETIC SUSCEPTIBILITY

In the theory of Ref. 17, the local moment magnetization $\langle S^z \rangle$ is left as a parameter which was represented by a Brillouin function. However, for a given parameter constellation, it is by no means predetermined that the system will indeed be ferromagnetic, i.e., a full theory would require a self-consistent treatment of $\langle S^z \rangle$ within the ("*concentrated*" or "*diluted*") KLM. This turns out to be a rather nontrivial goal. For our purpose, to derive the magnetic phase diagram of the KLM, we circumvent this problem by exploiting the static susceptibility of the itinerant electron subsystem,

$$\chi(T) = \sum_\sigma z_\sigma \left(\frac{\partial}{\partial B}\langle n_\sigma \rangle\right)_{T>T_C}^{B\to 0}. \tag{19}$$

We inspect exclusively the possibility of ferromagnetism, the average occupation number $\langle n_{i\sigma} \rangle$ is therefore site independent [Eq. (13)].

The spontaneous magnetization $\langle S^z \rangle$ of the local moment system and the conduction electron spin polarization $\langle n_\uparrow - n_\downarrow \rangle$ are mutually conditional. Therefore, they become critical for the same parameters, in particular, at the same temperature. In the critical region, we can therefore assume

$$\left(\frac{\partial}{\partial B} \langle S^z \rangle \right)_{T>T_C}^{B \to 0} = \eta \cdot \chi(T). \tag{20}$$

The proportionality of the response functions can be traced back to a proportionality of the expectation values $\langle S^z \rangle$ and $\langle n_\uparrow - n_\downarrow \rangle$, which is in terms of a Taylor expansion certainly fulfilled. In order to concentrate on the effects of dilution, we made a simple ansatz for the proportionality factor η, which neglects the dependence on model parameters and temperature. Instead we assume a equivalence of the reduced quantities

$$\frac{\langle S^z \rangle}{S} \Leftrightarrow \frac{\langle n_\uparrow - n_\downarrow \rangle}{n} \tag{21}$$

and take $\eta = S/n$. This ansatz, plausible as it is, can probably be replaced by more profound theories in an improved approach.

A straightforward derivation of the itinerant-electron susceptibility χ according to Eqs. (11), (6), (19), and (20) eventually ends up with the following expression:

$$\chi(T) = -2\mu_B \frac{Q_x(T) + K_x(T)}{1 + \eta J K_x(T)}. \tag{22}$$

For clarity, the lengthy derivation of $Q_x(T)$ and $K_x(T)$ is shifted to the appendix.

From the singularities of the paramagnetic susceptibility χ, we find the Curie temperature T_C as a function of model parameters such as lattice structure, spin value S, moment concentration x, band occupation $n \leqslant x$, and exchange coupling J. The singularities are the solutions of the following equation:

$$0 = 1 + \eta J K_x(T = T_C). \tag{23}$$

The instabilities of the paramagnetic phase towards ferromagnetism are thus given by the solutions of this equation.

V. MAGNETIC PHASE DIAGRAM

We have evaluated the criterion for ferromagnetism (23) for a simple cubic (sc) lattice where the width W of the Bloch band has been chosen to be 1 eV. The goal is to find out for which parameter constellations (moment concentration x, band occupation $n \leqslant x$, exchange coupling J) the system becomes ferromagnetic and what are the values for the Curie temperature $T_C = T_C(x, n, J)$. We start the analysis of the results with a discussion of the "*concentrated*" systems, where (having substances like EuO and Gd in mind) the exchange coupling constant J is ferromagnetic. To be consistent, we have restricted ourselves even in the case of "*diluted*" systems to a ferromagnetic exchange coupling $J > 0$, although the most topical diluted magnetic semiconductors seem to have an antiferromagnetic coupling. Furthermore, our model study considers the coupling of electrons to localized moments, the case of holes instead of electrons will not essentially change the important statements.

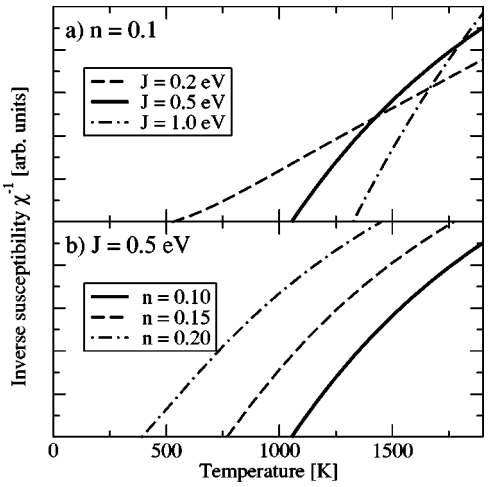

FIG. 1. Paramagnetic inverse susceptibility of the "*concentrated*" ($x=1$) Kondo lattice as function of the temperature, in (a) for a fixed band occupation $n=0.1$ and different exchange couplings, in (b) for a fixed exchange coupling $J=0.5$ eV and different carrier concentrations n.

Let us first inspect the case of the "*concentrated*" Kondo lattice ($x=1$). Figure 1 shows the paramagnetic inverse susceptibility of the band electrons as a function of the temperature for various parameter constellations (n, J). For sufficiently high temperatures and almost all parameter constellations, a Curie-Weiß behavior can be recognized. From the zeros of χ^{-1} we can read off the respective Curie temperature. In some cases two zeros are found (not shown in the figure). The requirement that χ must be positive in the paramagnetic phase ($T > T_C$) makes the choice of the physically relevant solution unique.

The band occupation n enters the susceptibility (19) and therefore the calculated T_C via the chemical potential μ, which is accordingly determined with the help of Eq. (13). Additionally n is included in the choice of η. Figure 2 demonstrates that ferromagnetism does exist with a distinct band occupation dependence of the Curie temperature. The most

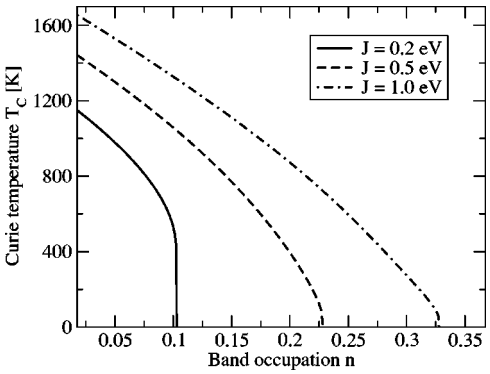

FIG. 2. Curie temperature as a function of the band occupation n for various exchange couplings J in the "*concentrated*" ($x=1$) Kondo-lattice model. Parameters: sc lattice, $W=1$ eV, $S=\frac{5}{2}$.

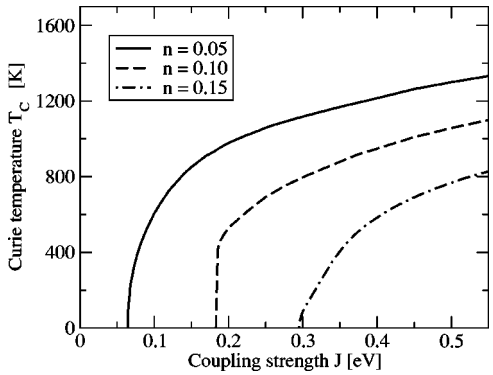

FIG. 3. Curie temperature as a function of the exchange coupling strength J for three different band occupations n in the "*concentrated*" ($x=1$) Kondo-lattice model. Parameters: sc lattice, $S=\frac{5}{2}$, $W=1$ eV.

remarkable feature is the restriction of ferromagnetism to surprisingly low carrier concentrations n. Arbitrarily small band occupations are sufficient to create a ferromagnetic order. In any case, the Curie temperature is zero for $n=0$. It was, however, numerically not possible to decide whether or not there is a steep but continuous increase to finite values. Note that the KLM does not consider a direct exchange between the localized moments. So the collective ordering is fully mediated by the interband exchange, i.e., by the conduction electron spin polarization. The width of the ferromagnetic phase on the n axis increases with the exchange coupling strength J, being restricted, however, even for strong couplings to low itinerant electron concentrations. The maximum value of the Curie temperature also increases with J. Typical J values for ("concentrated") ferromagnetic semiconductors such as EuO and EuS are of the order of some tenth of eV (Refs. 20 and 21).

Similar results are found with the "modified" RKKY of Refs. 10 and 12 where an effective Heisenberg model is solved by the Tyablikov approximation.[16] The model theory in Ref. 22 yields also qualitatively the same T_C behavior, namely a steep increase of T_C for very weak band occupations, a rather distinct maximum and then also a very rapid decrease to zero. The new feature of our theory (Fig. 2) is the T_C behavior for $n \to 0$.

The general J dependence of T_C is shown in Fig. 3. Two features are worth mentioning. First, T_C appears to run into a saturation in the strong coupling region. This is similar to what is reported in Ref. 10. In the present theory, however, the saturation needs a substantially stronger exchange coupling. Second, a critical $J=J_c(n)$ is needed to switch on ferromagnetism, which, at least in the low concentration regime, increases with increasing n.

We now inspect the influence of the dilution of the moments ($x<1$). We assume that each magnetic ion can in principle donate one electron to the conduction band. However, not all these charge carriers can be considered as really itinerant, so that $n \leqslant x$. Therewith we simulate the situation in the diluted ferromagnetic semiconductors. In the case of Mn^{2+} in $Ga^{3+}As^{3-}$, e.g., holes are created in the GaAs valence band which are partly compensated by antisites.[5] The inspection of the paramagnetic susceptibility as a function of

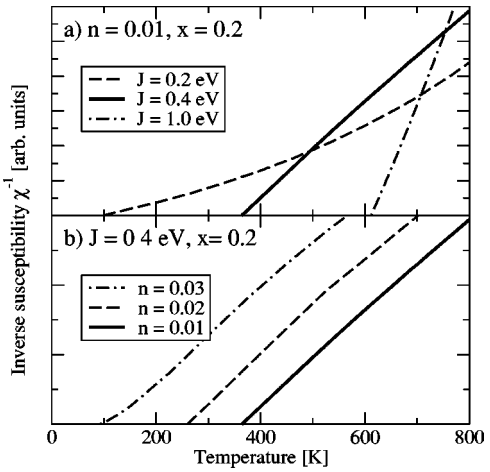

FIG. 4. Paramagnetic inverse susceptibility of the "*diluted*" Kondo lattice as a function of the temperature, in (a) for a fixed band occupation $n=0.01$ and different exchange couplings, in (b) for a fixed exchange coupling $J=0.4$ eV and different carrier concentrations n.

temperature for a given parameter constellation (Fig. 4) makes it clear that ferromagnetism does exist in the diluted moment system, too. The resulting Curie temperature is plotted in Fig. 5 as a function of the carrier concentration n for various moment concentrations x. As in the case of the "*concentrated*" system, ferromagnetism is restricted to the very low concentration region. Also the J dependence of the Curie

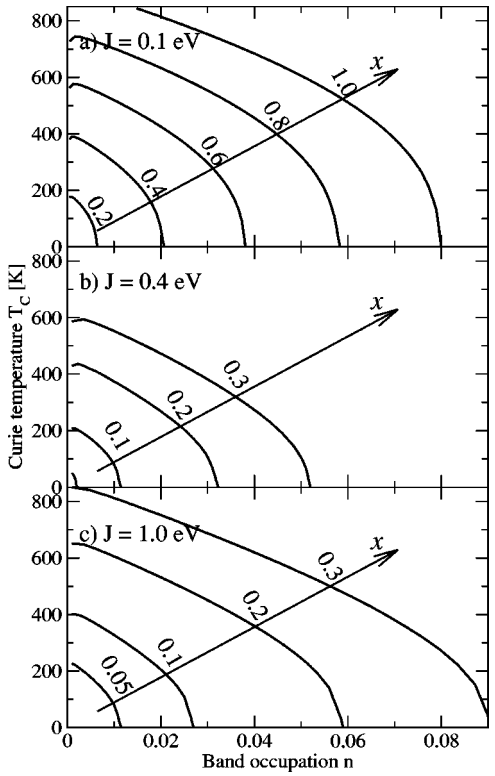

FIG. 5. Curie temperature as a function of the band occupation n for various concentrations x of magnetic moments in the "*diluted*" ($x<1$) Kondo-lattice model. (a) $J=0.1$ eV, (b) $J=0.4$ eV, (c) $J=1.0$ eV. Parameters: sc lattice, $W=1$ eV, $S=\frac{5}{2}$.

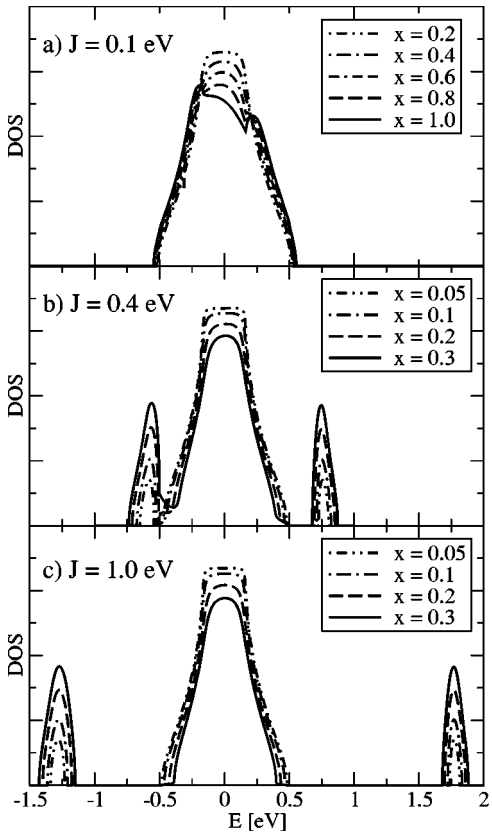

FIG. 6. Paramagnetic quasiparticle density of states of the "*diluted*" Kondo-lattice model in the paramagnetic phase as a function of energy for different values of the moment concentration x and three different exchange couplings J. Parameters: sc lattice, $W=1$ eV, $S=\frac{5}{2}$.

temperature for a given (x,n) pair is very similar to that for the "*concentrated*" systems plotted in Fig. 3. What is remarkable, however, is the fact that the concentration n must be very much smaller than the concentration x in order to allow ferromagnetic ordering. The compensation effects observed in diluted magnetic semiconductors (antisites, etc.) seem to be a necessary precondition for ferromagnetism in the diluted system. For $n=x$ ferromagnetism is excluded. An explanation for this is given by the quasiparticle density of states.

For sufficiently high values of J, the (paramagnetic) quasiparticle density of states (Fig. 6) consists of three parts. The low-energy and the high-energy subbands are built up by states from the correlated β-sites, while the middle structure is due to the uncorrelated α sites. The correlated subbands, which are exclusively responsible for a possible magnetic order, are exchange-split by about $\frac{1}{2}J(2S+1)$. When the three structures are well separated, then, the area under the two correlated peaks amounts to x while that of the uncorrelated middle band is $1-x$. With increasing x, i.e., higher moment density, more and more spectral weight is shifted into the correlated quasiparticle subbands. In simple terms, the two correlated bands can be understood as follows: An electron propagating in the low-energy subband hops mainly over lattice sites where it can orient its spin parallel to the local-moment (Mn^{2+}) spin $\left[\sim-\frac{1}{2}JS\right]$. In the high-energy subband, the spin orientation is predominantly antiparallel $\left[\sim+\frac{1}{2}J(S+1)\right]$. Since we have used for the self-energy $M_\sigma(E)$ the low-density approach of Ref. 17, the QDOS does not exhibit a noteworthy band occupation dependence.

The first precondition for ferromagnetism is that the Fermi edge lies in one of the correlated subbands. We observe in principle the same general structure as in the concentrated case ($x=1$) exhibited in Fig. 2. Extremely low carrier concentrations are already sufficient to induce ferromagnetism. Roughly estimated, we find ferromagnetic ordering for band occupations $0<n<n_c(J)\cdot x$, where $n_c(J)$ is the critical band occupation for $x=1$ at a given J.

It is indeed observed for diluted magnetic semiconductors that the number of itinerant carriers is substantially smaller than the number of local moments.[5] In $Ga_{1-x}Mn_xAs$, e.g., each Mn^{2+} ion in principle provides one hole in the valence band. However, only a certain percentage of them are really itinerant, the others are compensated, e.g., by antisites or interstitial Mn atoms, that act as donors. Erwin and Petukhov[23] were the first to suggest that such compensation effects might be in favor of a collective order. In the limit $J\rightarrow\infty$ they mapped the Hamiltonian (1) on an effective Heisenberg model and evaluated the latter using classical percolation theory. With our treatment of the Kondo-lattice model, which is valid for quantum spins and finite J, we can confirm that compensation is necessary for the existence of ferromagnetism. The reason is the complete filling of the lower correlated subband in Fig. 6 for $n=x$. This corresponds in the "*concentrated*" local-moment systems (Fig. 2) to a half-filling of the correlated spectrum, which is known to prevent a magnetic order.[10,12] In contrast to Erwin *et al.* the $n_c(J)$ determined from our results is substantially smaller than x. More recently a similar behavior was found by Bouzerar *et al.*[15] and Brey *et al.*[24]

Our findings are in particular interesting, because they seem to be in disagreement with some *ab initio* calculations.[25,26] These papers mostly refer to compensation effects of As antisites. Since interstitial Mn atoms have a different magnetic behavior, its compensation might have a different effect on T_C, too. Nevertheless, this point is apparently still an exciting open question, both for experimentalists and theoreticians.

VI. SUMMARY

In conclusion, it can be stated that the basic theory for the self-energy (6) is undoubtedly justifiable for the low-concentration limit of the KLM. Fortunately, this is obviously just the most relevant region for stable ferromagnetism.[10,12,22] The assumption of equivalent criticality (20) of the two subsystems of the KLM is certainly acceptable, while the choice of the parameter η [see Eq. (21)] seems to be plausible. Nevertheless, the latter surely needs stronger confirmation. Interesting remarks about this fact can be found in Ref. 27. A change of η, however, does not qualitatively alter the findings of the theory. The absolute values of the Curie temperatures depend of course sensitively on η.

We have shown by a CPA-type treatment of the disordered KLM how the magnetic disorder in diluted local-

moment systems influences the existence of a ferromagnetic phase and the respective Curie temperature. The model study gives a qualitative explanation of the ferromagnetism in diluted magnetic semiconductors. A main consequence of our model study is that a substantial compensation of the itinerant charge carriers ($n<x$) by antisites or other mechanisms appears to be a necessary condition for the existence of a ferromagnetic ordering. It is intended for the future to apply our theory to real diluted magnetic semiconductors (negative J!).

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APPENDIX

We give here the full analytical solution for the paramagnetic susceptibility (22). By definition (19) it is determined by the electron polarization. Substituting (11) and (14) into the spectral theorem (13) yields

$$\frac{\partial\langle n_\sigma\rangle}{\partial B}=\int_{-\infty}^{+\infty}\mathrm{d}E\, f_-(E)\left(-\frac{1}{\pi}\,\mathrm{Im}\int_{-\infty}^{+\infty}\mathrm{d}\omega\frac{\rho_0(\omega)\cdot\left(\frac{\partial}{\partial B}\Sigma_\sigma(E)\right)}{[E-\omega-\Sigma_\sigma(E)]^2}\right). \tag{A1}$$

According to the chain rule the derivative is reduced to that of $(\partial/\partial B)\Sigma_\sigma(E)$. It is derived from the application of $\partial/\partial B$ to Eq. (12). Afterwards the limit $B\to 0$ is taken. Those terms which are proportional to $\partial\langle S^z\rangle/\partial B=\eta\cdot\chi(T)$ give rise to $K_x(T)$, the most important term of Eq. (22),

$$K_x(T)=\int_{-\infty}^{+\infty}\mathrm{d}E f_-(E)\left(-\frac{1}{\pi}\,\mathrm{Im}\left[D_{\mathrm{pm}}(E)\frac{B_x(E)H(E)}{N_x(E)}\right]\right) \tag{A2}$$

The remaining terms are summed to $Q_x(T)+K_x(T)$ with

$$Q_x(T)=\int_{-\infty}^{+\infty}\mathrm{d}E\, f_-(E)\times\left(-\frac{1}{\pi}\,\mathrm{Im}\left[\frac{D_{\mathrm{pm}}(E)}{N_x(E)}\left(A_x(E)-\frac{B_x(E)}{1-\frac{1}{2}JG_0(E)}\right)\right]\right). \tag{A3}$$

Equation (22) is a consequence of the result

$$\chi(T)=-J\eta\chi(T)K_x(T)-2\mu_B[Q_x(T)+K_x(T)]. \tag{A4}$$

In these expressions we have used further abbreviations, which are chosen according to mathematical simplicity. Hence, the individual terms do not carry a particular physical meaning. The ω integrations in (A1) and (A6) are denoted as

$$D_{\mathrm{pm}}(E)=\int_{-\infty}^{+\infty}\mathrm{d}\omega\frac{\rho_0(\omega)}{(E-\omega-\Sigma_{\mathrm{pm}}(E))^2}, \tag{A5}$$

$$G_k(E)=\int_{-\infty}^{+\infty}\mathrm{d}\omega\frac{\rho_0(\omega)}{(E-\omega)^{k+1}}. \tag{A6}$$

From the variety of terms emerging after differentiating Eq. (12) an x-independent factor

$$H(E)=\frac{1-\frac{1}{2}JG_0(E)-\frac{1}{4}J^2S(S+1)G_1(E)}{\left(1-\frac{1}{2}JG_0(E)\right)^2} \tag{A7}$$

can be separated. The remaining terms are

$$N_x(E)=(1-x)\frac{1-D_{\mathrm{pm}}(E)\Sigma_{\mathrm{pm}}^2(E)}{(1+R_{\mathrm{pm}}(E)\Sigma_{\mathrm{pm}}(E))^2}+x\frac{1-D_{\mathrm{pm}}(E)(M_{\mathrm{pm}}(E)-\Sigma_{\mathrm{pm}}(E))^2}{(1-R_{\mathrm{pm}}(E)(M_{\mathrm{pm}}(E)-\Sigma_{\mathrm{pm}}(E)))^2}, \tag{A8}$$

$$A_x(E)=\frac{1-x}{(1+R_{\mathrm{pm}}(E)\Sigma_{\mathrm{pm}}(E))^2}+\frac{x}{(1-R_{\mathrm{pm}}(E)(M_{\mathrm{pm}}(E)-\Sigma_{\mathrm{pm}}(E)))^2}, \tag{A9}$$

$$B_x(E)=\frac{x}{(1-R_{\mathrm{pm}}(E)(M_{\mathrm{pm}}(E)-\Sigma_{\mathrm{pm}}(E)))^2}. \tag{A10}$$

Obviously, for the concentrated case, where $x=1$ and $M_{\mathrm{pm}}(E)=\Sigma_{\mathrm{pm}}(E)$, the algebraic equations have a much simpler structure.

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