Ferromagnetism in the Kondo-lattice model

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We propose a modified Rudermann, Kittel, Kasuya, Yosida (RKKY) technique to evaluate the magnetic properties of the ferromagnetic Kondo-lattice model. Together with a previously developed self-energy approach to the conduction-electron part of the model, we obtain a closed system of equations which can be solved self-consistently. The results allow us to study the conditions for ferromagnetism with respect to the band occupation n, the interband exchange coupling J, and the temperature T. Ferromagnetism appears for relatively low electron (hole) densities, while it is excluded around half-filling ($n=1$). For small J the conventional RKKY theory ($\sim J^2$) is reproduced, but with strong deviations for very moderate exchange couplings. For not-too-small n a critical J_c is needed to produce ferromagnetism with a finite Curie temperature T_C, which increases with J, then running into a kind of saturation, in order to fall off again and disappear above an upper critical exchange J. Spin waves show a uniform softening with rising temperature, and a nonuniform behavior as functions of n and J. The disappearance of ferromagnetism when varying T, J, and n is uniquely connected to the stiffness constant D becoming negative.

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I. INTRODUCTION

The Kondo-lattice model (KLM)[1–8] describes the interplay of itinerant electrons in a partially filled conduction band with quantum-mechanical spins (magnetic moments) localized at certain lattice sites. Typical model properties result from an interband exchange between the two subsystems. They are provided by a model Hamiltonian, which consists of two parts:

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

H_s describes uncorrelated electrons in a nondegenerate energy band (''s electrons''),

$$H_s=\sum_{ij\sigma} T_{ij}c_{i\sigma}^{\dagger}c_{j\sigma}, \tag{2}$$

where $c_{i\sigma}^{\dagger}(c_{i\sigma})$ creates (annihilates) an electron specified by the lower indices. The index i refers to a lattice site $\mathbf{R}_i$, and $\sigma=\uparrow,\downarrow$ is the spin projection. T_{ij} are the hopping integrals. The second term in Eq. (1) represents an interband exchange as an intra-atomic interaction between the itinerant electron spin $\boldsymbol{\sigma}_i$ and the localized spin $\mathbf{S}_i$ (''f electrons''):

$$H_{sf}=-J\sum_i \boldsymbol{\sigma}_i\cdot\mathbf{S}_i. \tag{3}$$

According to the sign of the exchange coupling J, a parallel ($J>0$) or antiparallel ($J<0$) alignment of itinerant and localized spins is favored with remarkable differences in the physical properties. We restrict our considerations in the following to the $J>0$ case, sometimes referred to as the ''ferromagnetic Kondo-lattice model,'' and also known as the ''$s-f$ ($s-d$) model.'' The applications of the KLM are manifold. In the 1970s it was extensively used to describe the electronic and magneto-optic properties of magnetic semiconductors such as the ferromagnetic EuO and EuS.[6,7] Most studies focused on the spectacular temperature dependence of the unoccupied band states. The well-known ''red shift'' of the optical absorption edge upon cooling from $T=T_C$ to $T=0$ K (Ref. 9) can be understood as a corresponding temperature shift of the lower $5d$ conduction-band edge. Recent quasiparticle band-structure calculations for bulk EuO (Ref. 10) confirmed the observed striking temperature dependence as a consequence of a ferromagnetic exchange J of the order of some tenth of an eV. The interest in EuO, in particular in thin films, was strongly revived by the observation of a temperature driven metal-insulator transition below T_C with a large drop of the resistivity by as much as eight orders of magnitude.[11] This gives rise to a colossal magnetoresistance (CMR) much stronger than that of manganites like $La_{1-x}Sr_xMnO_3$. Recently, a surface half-metal-insulator transition was predicted[12] for a Eu(100) film. The reason for this is an exchange-split surface state.

In ferromagnetic local moment metals like Gd a RKKY-type interaction is believed to cause the ferromagnetic order, so that in principle a self-consistent description of magnetic and electronic properties by the KLM is possible. The $T=0$ moment of Gd is found to be $7.63\mu_B$[13]. $7\mu_B$ stems from the exactly half-filled $4f$ shell. The excess moment of $0.63\mu_B$ is due to an induced spin polarization of the *a priori* nonmagnetic ($5d$,$6s$) conduction bands, indicating a weak or intermediate $J>0$.[14]

An interesting application of the KLM refers to semimagnetic semiconductors, where randomly distributed Mn^{2+} or Fe^{2+} ions provide localized magnetic moments which influence, via the exchange coupling J, the band states of systems like $Ga_{1-x}Mn_xAs$.[15] The Mn^{2+} ions provide both localized spins ($S=\frac{5}{2}$, concentration x) and free carriers (holes, concentration $x^{\star}<x$). The latter take care of an indirect coupling between the moments leading for small x even to a ferromagnetic order. The highest T_C so far reached is 110 K for $x=0.053$,[16] where $x^{\star}$ is found to be about 15% of the Mn concentration x. The origin of ferromagnetism in $Ga_{1-x}Mn_xAs$ is controversial. The authors of Ref. 16

claimed that the measured $T_C(x)$ is consistent with a conventional RKKY interaction between the Mn ions. However, they estimated the exchange coupling J to be of several eV, therewith comparable to the Fermi energy. This makes second-order perturbation theory with respect to J rather questionable. The RKKY interaction should be modified to account for higher-order spin polarization terms. In this paper we are going to present a respective proposal.

Another modern application of the $J>0$ KLM aims at manganese oxides with perovskite structures $T_{1-x}D_x\mathrm{MnO_3}$ (T=La, Pr, Nd; D=Sr,Ca, Ba, Pb) which have attracted great scientific interest because of their CMR.[17,18] Parent compounds like the protagonist $\mathrm{La^{3+}Mn^{3+}O_3}$ are antiferromagnetic insulators with a localized Mn spin of $S=2$. Replacing a trivalent $\mathrm{La^{3+}}$ ion by a divalent earth-alkali ion ($\mathrm{Ca^{2+}}$) leads to a homogenuous valence mixture of the manganese ion ($\mathrm{Mn^{3+}_{1-x}}$,$\mathrm{Mn^{4+}_x}$). The three $3d-t_{2g}$ electrons of $\mathrm{Mn^{4+}}$ can be considered as almost localized, forming an $S=\frac{3}{2}$ spin. Furthermore, $(1-x)$ electron per Mn site are of $3d-e_g$ type, and itinerant. They are exchange coupled to the $S=\frac{3}{2}$ spins, realizing just the situation which is modeled by the KLM. The exchange coupling is ferromagnetic ($J>0$). Since manganites are bad electrical conductors, it can be assumed that the intraatomic exchange $J>0$ is much larger than the hopping-matrix element $|t|$. With estimated e_g bandwidths of 1–2 eV and lower limits for J of $\gtrsim 1$ eV,[19] the manganites surely belong to the group of strongly coupled materials. Many fascinating features of the CMR materials can be traced back to an intimate correlation between magnetic and electronic components. One of the not yet fully understood features concerns the spin dynamics in the ferromagnetic phase [$x\sim 0.3$ in $\mathrm{La_{1-x}(Ca,Pb)_xMnO_3}$]. It was reported[20] that the spin-wave dispersion (SWD) of materials with high T_C (e.g., $\mathrm{La_{0.7}Pb_{0.3}MnO_3}$; with $T_C=355$ K) can be reasonably well reproduced by a simple Heisenberg model with nearest-neighbor exchange only. Furukawa[21] gave qualitative arguments that this can be understood for strongly exchange coupled materials. However, strong deviations from the typical Heisenberg behavior of the SWD have been found for manganites with lower T_C's.[22–24] It is a challenging question whether or not the softening of the SWD at the zone boundary can be explained by a better treatment of the KLM, i.e., by an appropriate consideration of the influence of the conduction-electron self-energy on the effective coupling between the localized magnetic moments.[25,26] Solovyev and Terakura[27] argued in such a way claiming that the SWD softening has a purely magnetic background. By contrast, Mancini *et al.*[28] believe that the KLM [Eq. (1)] has to be extended by a direct (superexchange) interaction between the Mn moments to account for antiferromagnetic tendencies, which dominate the magnetic behavior of the parent material $\mathrm{LaMnO_3}$.

The above-presented, of course incomplete, list documents the rich variety of, at least, qualitative applications for the KLM. However, the model Hamiltonian [Eq. (1)] provokes such a complicated many-body problem that approximations have to be tolerated. Most of the recent theoretical work on the KLM, aiming at the CMR-materials, assumed classical spins,[29–31] mainly in order to apply ''dynamical mean-field theory'' (DMFT). It is surely not unfair to consider this assumption as rather problematic.

It is the intention of the present paper to use a special RKKY technique for a detailed investigation of the magnetic properties of the KLM with ferromagnetic interband exchange. We exploit the method of Ref. 32 to map the interaction operator H_{sf} [Eq. (3)] to the Heisenberg model Hamiltonian. This is done by averaging out the conduction electron degrees of freedom by properly defined ''restricted'' Green functions. The resulting effective Heisenberg exchange integrals are functionals of the itinerant-electron self-energy, and therewith strongly temperature and band occupation dependent. These dependencies manifest themselves in fundamental magnetic properties such as phase diagrams, Curie temperature, spin-wave dispersions, and so on.

The electronic self-energy is taken from a ''moment conserving decoupling approach'' introduced in Ref. 32 and successfully applied to several topics in previous papers.[10,12,14,33] The method carefully interpolates between nontrivial limiting cases.

The final goal of our effort is to obtain quantitative information about magnetism and the temperature-dependent electronic structure of real local-moment systems. For this purpose it is intended in a future work to combine our model study with a ''first principles'' band-structure calculation. First attempts in this direction were made done for EuO (Ref. 10) and Gd (Ref. 14); however, the magnetic ordering could not be derived self-consistently. The paper is organized as follows. In Sec. II we briefly formulate the many-body problem of the KLM. Section III reproduces the most important aspects of the ''modified RKKY interaction'' (M-RKKY interaction), the understanding of which is vital for a correct interpretation of the results, which are discussed in Sec. IV.

II. KONDO-LATTICE MODEL

The model Hamiltonian [Eq. (1)] provokes a nontrivial many-body problem, which can be rigorously solved only for a small number of limiting cases. For practical reasons, it sometimes appears convenient to use the second quantized form of the exchange interaction [Eq. (3)],

$$H_{sf}=-\frac{1}{2}J\sum_{i\sigma}\left(z_\sigma S_i^z n_{i\sigma}+S_i^\sigma c_{i-\sigma}^\dagger c_{i\sigma}\right), \tag{4}$$

with the abbreviations

$$n_{i\sigma}=c_{i\sigma}^\dagger c_{i\sigma}, \quad z_\sigma=\delta_{\sigma\uparrow}-\delta_{\sigma\downarrow}, \quad S_i^\sigma=S_i^x+iz_\sigma S_i^y. \tag{5}$$

The first term in Eq. (4) describes an Ising-like interaction between the z components of the localized and itinerant spins. The second term comprises spin-exchange processes between the two subsystems.

To study the conduction-electron properties, we use the single-electron Green function

$$G_{ij\sigma}(E)=\langle\langle c_{i\sigma};c_{j\sigma}^\dagger\rangle\rangle_E. \tag{6}$$

Its equation of motion reads

$$\sum_m (E\delta_{im} - T_{im}) G_{mj\sigma}(E)$$

$$= \hbar\delta_{ij} - \frac{1}{2} J\{z_\sigma I_{ii,j\sigma}(E) + F_{ii,j\sigma}(E)\}, \tag{7}$$

where the two types of interaction terms in Eq. (4) are causing the ''spin-flip function''

$$F_{im,j\sigma}(E) = \langle\langle S_i^{-\sigma} c_{m-\sigma}; c_{j\sigma}^\dagger\rangle\rangle_E \tag{8}$$

and the ''Ising function''

$$I_{im,j\sigma}(E) = \langle\langle S_i^z c_{m\sigma}; c_{j\sigma}^\dagger\rangle\rangle_E . \tag{9}$$

These two ''higher'' Green functions prevent a direct solution of the equation of motion (7). A formal solution of the Fourier-transformed single-electron Green function,

$$G_{\mathbf{k}\sigma}(E) = \frac{1}{N}\sum_{ij} G_{ij\sigma}(E) e^{i\mathbf{k}\cdot(\mathbf{R}_i - \mathbf{R}_j)}, \tag{10}$$

defines the complex electron self-energy $\Sigma_{\mathbf{k}\sigma}(E)$:

$$G_{\mathbf{k}\sigma}(E) = \frac{\hbar}{E + \mu + i0^+ - \varepsilon(\mathbf{k}) - \Sigma_{\mathbf{k}\sigma}(E)}, \tag{11}$$

$$\langle\langle [c_{\mathbf{k}\sigma}, H_{sf}]_-; c_{\mathbf{k}\sigma}\rangle\rangle_E = \Sigma_{\mathbf{k}\sigma}(E) G_{\mathbf{k}\sigma}(E). \tag{12}$$

$\varepsilon(\mathbf{k})$ are the Bloch energies:

$$\varepsilon(\mathbf{k}) = \frac{1}{N}\sum_{ij} T_{ij} e^{i\mathbf{k}\cdot(\mathbf{R}_i - \mathbf{R}_j)}. \tag{13}$$

By use of the quasiparticle density of states

$$\rho_\sigma(E) = -\frac{1}{\hbar\pi N}\sum_{\mathbf{k}} \mathrm{Im} G_{\mathbf{k}\sigma}(E-\mu), \tag{14}$$

we can calculate the spin-dependent occupation numbers

$$\langle n_\sigma\rangle = \int_{-\infty}^{+\infty} dE f_-(E)\rho_\sigma(E), \tag{15}$$

important to fix the band polarization $m = \langle n_\uparrow\rangle - \langle n_\downarrow\rangle$. $f_-(E) = [\exp(\beta(E-\mu)+1]^{-1}$ is the Fermi function.

The central quantity is the self-energy, the determination of which solves the problem. However, for finite temperatures and arbitrary band occupations an exact expression is not avaliable. In Ref. 32 a ''moment conserving decoupling approach'' (MCDA) predicted the following structure of the self-energy:

$$\Sigma_{\mathbf{k}\sigma}(E) = -\frac{1}{2} J z_\sigma \langle S^z\rangle + \frac{1}{4} J^2 D_{\mathbf{k}\sigma}(E). \tag{16}$$

The first term is linear in the coupling J, and proportional to the the local-moment magnetization $\langle S^z\rangle$. It represents the result of a mean-field approach which is correct in the weak-coupling limit. The second term is predominantly determined by spin-exchange processes between band electrons and localized moments. It is a complicated functional of the self-energy itself. Thus Eq. (16) is not an analytical solution at all, but an implicit equation for $\Sigma_{\mathbf{k}\sigma}(E)$. The MCDA, the details of which are presented in Ref. 32, is a nonperturbational decoupling approach to a set of properly defined Green functions that correctly reproduces the non-trivial exact limiting cases of the KLM. A weighty example is the special case of a ''ferromagnetically saturated semiconductor.''[8,34,35,36,37] The various ''higher'' Green functions have been approximated by linear combination of ''lower'' functions, where the rigorous spectral representations of these ''higher'' Green functions and their exact special cases ($S = 1/2$, ferromagnetic saturation, ...) justify the respective ansatz. Free parameters are finally fitted to exactly known spectral moments. In a certain sense, the method can be considered to be an interpolation scheme between important limiting cases. It was successfully used in the past for several applications.[10,12,14,33,36] We refer the reader for technical details to one of these papers.

The term $D_{\mathbf{k}\sigma}(E)$ in Eq. (16) contains certain expectations values such as

(a) $\langle n_{i\sigma}\rangle$, ...,
(b) $\langle S_i^z\rangle, \langle S_i^{\pm} S_i^{\mp}\rangle, \langle (S_i^z)^2\rangle$, ...,
(c) $\Delta_\sigma = \langle S_i^z n_{i\sigma}\rangle, \gamma_\sigma = \langle S_i^{-\sigma} c_{i\sigma}^\dagger c_{i-\sigma}\rangle$, ...

By use of the spectral theorem, terms (a) and (c) can exactly be expressed by the Green functions (6), (8) and (9). Besides Eq. (15) it holds that

$$\gamma_\sigma = -\frac{1}{\hbar\pi}\int_{-\infty}^{+\infty} dE\, f_-(E)\mathrm{Im} F_{ii,i\sigma}(E-\mu), \tag{17}$$

$$\Delta_\sigma = -\frac{1}{\hbar\pi}\int_{-\infty}^{+\infty} dE\, f_-(E)\mathrm{Im}\Gamma_{ii,i\sigma}(E-\mu). \tag{18}$$

It remains to express the pure ''local-moment'' correlations (b) in terms of the electronic self-energy $\Sigma_{\mathbf{k}\sigma}(E)$ to obtain a closed system of equations that can be solved self-consistently. A finite coupling between the spin operators must be of indirect nature since the model Hamiltonian [Eq. (1)] does not contain any direct exchange. To obtain the pure spin correlations (b), we map exchange interaction (4) on an effective Heisenberg model with effective exchange integrals $\hat{J}_{ij}$ being functionals of the electronic self-energy. The result is a ''modified'' RKKY interaction (M-RKKY interaction) that explicitly takes into account the exchange-induced spin polarization of the band electrons. In lowest order it reproduces the conventional RKKY method.

III. EFFECTIVE EXCHANGE OPERATOR

The starting idea is to map the interband ($s-f$) exchange on an effective spin Hamiltonian of the Heisenberg type:

$$H_f = -\sum_{ij} \hat{J}_{ij}\, \mathbf{S}_i \cdot \mathbf{S}_j . \tag{19}$$

For this purpose we use the $s-f$ interaction in the following form:

$$H_{sf} = -J\frac{\hbar}{N}\sum_{i\sigma\sigma'}\sum_{\mathbf{kq}} e^{-i\mathbf{q}\cdot\mathbf{R}_i}(\mathbf{S}_i\cdot\hat{\boldsymbol{\sigma}})_{\sigma\sigma'}c^{\dagger}_{\mathbf{k}+\mathbf{q}\sigma}c_{\mathbf{k}\sigma'}. \quad (20)$$

$\hat{\boldsymbol{\sigma}}$ is the band electron spin operator, the components of which are Pauli spin matrices. The above-mentioned mapping occurs by averaging out the band electron degrees of freedom:

$$H_{sf}\rightarrow\langle H_{sf}\rangle^{(c)}\equiv H_f. \quad (21)$$

Averaging only in the band electron subspace means that $\langle H_{sf}\rangle^{(c)}$ retains an operator character in the *f*-spin subspace. According to Eq. (20) we have to calculate

$$\langle c^{\dagger}_{\mathbf{k}+\mathbf{q}\sigma}c_{\mathbf{k}\sigma'}\rangle^{(c)} = \frac{1}{\Xi'}\mathrm{Tr}(e^{-\beta H'}c^{\dagger}_{\mathbf{k}+\mathbf{q}\sigma}c_{\mathbf{k}\sigma'}). \quad (22)$$

H' has exactly the same structure as the KLM Hamiltonian H [Eq. (1)], except for the fact that for the averaging procedure the *f*-spin operators are to be considered as *c* numbers, therefore not affecting the trace. Ξ' is the corresponding grand partition function. The expectation value [Eq. (22)] does not necessarily vanish for $\mathbf{q}\neq 0$ and $\sigma\neq\sigma'$, as it would when averaging in the full Hilbert space of the KLM. To calculate Eq. (22) we introduce a proper ''restricted'' Green function.

$$\hat{G}^{\sigma'\sigma}_{\mathbf{k},\mathbf{k}+\mathbf{q}}(E) = \langle\langle c_{\mathbf{k}\sigma'};c^{\dagger}_{\mathbf{k}+\mathbf{q}\sigma}\rangle\rangle^{(c)}_{E}, \quad (23)$$

which has the ''normal'' definition of a retarded Green function, only the averages have to be done in the Hilbert space of H'. The equation of motion is readily derived for both cases, namely, in the case that $c_{\mathbf{k}\sigma'}$ is the active operator or $c^{\dagger}_{\mathbf{k}+\mathbf{q}\sigma}$. By use of the ''free'' Green function,

$$G^{(0)}_{\mathbf{k}}(E) = \frac{\hbar}{E+\mu-\varepsilon(\mathbf{k})}, \quad (24)$$

we can combine both equations:

$$\begin{aligned}\hat{G}^{\sigma'\sigma}_{\mathbf{k},\mathbf{k}+\mathbf{q}}(E) &= \delta_{\sigma\sigma'}\delta_{\mathbf{q},0}G^{(0)}_{\mathbf{k}}(E)\\ &\quad -\frac{J}{2N}\sum_{i\mathbf{k}'\sigma''}\{e^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}_i}G^{(0)}_{\mathbf{k}}(E)\\ &\quad\times(\mathbf{S}_i\cdot\hat{\boldsymbol{\sigma}})_{\sigma'\sigma''}\hat{G}^{\sigma''\sigma}_{\mathbf{k}',\mathbf{k}+\mathbf{q}}(E)\\ &\quad + e^{-i[\mathbf{k}'-(\mathbf{k}+\mathbf{q})]\cdot\mathbf{R}_i}G^{(0)}_{\mathbf{k}+\mathbf{q}}(\mathbf{S}_i\cdot\hat{\boldsymbol{\sigma}})_{\sigma''\sigma}\hat{G}^{\sigma'\sigma''}_{\mathbf{k},\mathbf{k}'}(E)\}. \end{aligned} \quad (25)$$

This equation is still exact and can be iterated up to any desired accuracy.

In first order, where the restricted Green function on the right-hand side of Eq. (25) is replaced by the ''free'' Green function [Eq. (24)], one obtains

$$\begin{aligned}[\hat{G}^{\sigma'\sigma}_{\mathbf{k},\mathbf{k}+\mathbf{q}}(E)]^{(1)} &= \delta_{\sigma\sigma'}\delta_{\mathbf{q},\mathbf{0}} - \frac{J}{N}\sum_i e^{i\mathbf{q}\cdot\mathbf{R}_i}G^{(0)}_{\mathbf{k}}(E)\\ &\quad\times(\mathbf{S}_i\cdot\hat{\boldsymbol{\sigma}})_{\sigma'\sigma}G^{(0)}_{\mathbf{k}+\mathbf{q}}. \end{aligned} \quad (26)$$

Exploiting the spectral theorem in the restricted Hilbert space, for the expectation value [Eq. (22)] one finds

$$\begin{aligned}&\left(\frac{1}{N}\sum_{\mathbf{k}}\langle c^{\dagger}_{\mathbf{k}+\mathbf{q}\sigma}c_{\mathbf{k}\sigma'}\rangle^{(c)}\right)^{(1)}\\ &\quad = \delta_{\sigma\sigma'}\delta_{\mathbf{q},0}\frac{1}{N}\sum_{\mathbf{k}} f_{-}[\varepsilon(\mathbf{k})]\\ &\quad\quad -\frac{J}{\hbar}\frac{1}{N}\sum_i e^{i\mathbf{q}\cdot\mathbf{R}_i}D^{(1)}_{\mathbf{q}}\cdot(\mathbf{S}_i\cdot\hat{\boldsymbol{\sigma}})_{\sigma'\sigma}. \end{aligned} \quad (27)$$

Here we have defined

$$\begin{aligned}D^{(1)}_{\mathbf{q}} &= -\frac{1}{\pi}\int_{-\infty}^{+\infty}dE\, f_{-}(E)\,\mathrm{Im}\frac{1}{N}\\ &\quad\times\sum_{\mathbf{k}} G^{(0)}_{\mathbf{k}}(E-\mu)G^{(0)}_{\mathbf{k}+\mathbf{q}}(E-\mu). \end{aligned} \quad (28)$$

Because of

$$\mathbf{S}_i\cdot\hat{\boldsymbol{\sigma}} = \frac{1}{2}\begin{pmatrix} S^z_i & S^-_i\\ S^+_i & -S^z_i\end{pmatrix}, \quad (29)$$

one realizes that the first term in Eq. (27) does not contribute to Eq. (21), while the second term gives rise to an effective Hamiltonian like Eq. (19), with the following effective exchange integrals:

$$\hat{J}^{(1)}_{ij} = \frac{1}{N}\sum_{\mathbf{q}}\hat{J}^{(1)}(\mathbf{q})e^{-i\mathbf{q}\cdot(\mathbf{R}_i-\mathbf{R}_j)}, \quad (30)$$

$$\begin{aligned}\hat{J}^{(1)}(\mathbf{q}) &= -\frac{1}{2}J^2D^{(1)}_{\mathbf{q}}\\ &= -\frac{1}{2}J^2\hbar^2\sum_{\mathbf{k}}\frac{f_{-}[\varepsilon(\mathbf{k}+\mathbf{q})]-f_{-}[\varepsilon(\mathbf{k})]}{\varepsilon(\mathbf{k}+\mathbf{q})-\varepsilon(\mathbf{k})}. \end{aligned} \quad (31)$$

This is the classical RKKY exchange, which comes out in first order of our Green-function procedure.

It is well known that Eq. (31) can be equivalently derived by conventional second-order perturbation theory starting from an unpolarized conduction electron gas. In order to incorporate the exchange-induced spin polarization of the conduction electrons to higher order, it almost suggests itself to modify approximation (26) by replacing in the exact expression [Eq. (25)] the restricted Green function by the full, all-polarization processes containing the single-electron Green function [Eq. (6)]:

$$\hat{G}^{\sigma''\sigma}_{\mathbf{k}',\mathbf{k}+\mathbf{q}}(E)\rightarrow\delta_{\sigma''\sigma}\delta_{\mathbf{k}'\mathbf{k}+\mathbf{q}}G_{\mathbf{k}+\mathbf{q}\sigma}(E), \quad (32)$$

$$\hat{G}^{\sigma'\sigma''}_{\mathbf{k},\mathbf{k}'}(E)\rightarrow\delta_{\sigma'\sigma''}\delta_{\mathbf{k}\mathbf{k}'}G_{\mathbf{k}\sigma'}(E). \quad (33)$$

For the required expectation value [Eq. (22)], we obtain an expression similar to Eq. (27), only $D^{(1)}_{\mathbf{q}}$ has to be replaced by

$$D_{\mathbf{q}}^{\sigma\sigma'} = -\frac{1}{\pi}\int_{-\infty}^{+\infty} dE\, f_-(E)\,\mathrm{Im}\frac{1}{N}\sum_{\mathbf{k}}\,[G_{\mathbf{k}}^{(0)}(E-\mu) \times G_{\mathbf{k}+\mathbf{q}\sigma}(E-\mu) + G_{\mathbf{k}+\mathbf{q}}^{(0)}(E-\mu)G_{\mathbf{k}\sigma'}(E-\mu)]. \tag{34}$$

Exploiting the inversion symmetry, one proves

$$D_{\mathbf{q}}^{\uparrow\downarrow} = D_{\mathbf{q}}^{\downarrow\uparrow} = \frac{1}{2}\sum_{\sigma} D_{\mathbf{q}}^{\sigma\sigma}. \tag{35}$$

This guarantees an isotropic effective exchange operator [Eq. (19)] with the exchange integral

$$\hat{J}(\mathbf{q}) = -\frac{J^2}{4}\int_{-\infty}^{+\infty} dE\, f_-(E)\frac{1}{N}\sum_{\mathbf{k}\sigma}\left(-\frac{1}{\pi}\mathrm{Im}[\,G_{\mathbf{k}}^{(0)}(E-\mu) \times G_{\mathbf{k}+\mathbf{q}\sigma}(E-\mu)]\right). \tag{36}$$

Via $G_{\mathbf{k}+\mathbf{q}\sigma}(E-\mu)$ the effective exchange is a functional of the electronic self-energy [Eq. (16)], and therefore receives a distinct temperature and electron density dependence.

To obtain the magnetic properties of the local-moment system from the effective operator [Eq. (19)], we use the spin Green function

$$P_{ij}^{(a)}(E) = \langle\langle S_i^+ ; e^{aS_j^z} S_j^- \rangle\rangle_E \tag{37}$$

first proposed by Callen.[38] a is a real number, which eventually helps to derive several spin-correlation functions by applying the spectral theorem to $P_{ij}^{(a)}(E)$ and a proper differentiation with respect to a. The equation of motion of $P_{ij}^{(a)}(E)$ reads

$$EP_{ij}^{(a)}(E) = 2\hbar\,\delta_{ij}\langle A_a\rangle - 2\hbar\sum_m \hat{J}_{im} \times \langle\langle (S_m^+ S_i^z - S_i^+ S_m^z); e^{aS_j^z} S_j^- \rangle\rangle_E. \tag{38}$$

Here we have abbreviated

$$\langle A_a\rangle = \langle [S_i^+ , e^{aS_i^z} S_i^-]_- \rangle. \tag{39}$$

It is well known that a simple random-phase-approximation decoupling of the higher Green function on the right-hand side of Eq. (38) yields surprisingly convincing results in the low- and high-temperature regions (''Tyablikov-approximation''). Doing so, after a Fourier transformation we obtain

$$P_{\mathbf{q}}^{(a)}(E) = \frac{2\hbar^2\langle A_a\rangle}{E - E(\mathbf{q}) + i0^+}. \tag{40}$$

In this approximation the magnon energies $E(\mathbf{q})$ are real:

$$E(\mathbf{q}) = 2\hbar\langle S^z\rangle[\hat{J}_0 - \hat{J}(\mathbf{q})], \tag{41}$$

$$\hat{J}_0 = \hat{J}(\mathbf{q}=0) = \sum_i \hat{J}_{ij}. \tag{42}$$

Following the Callen method[38] for the local-moment magnetization one obtains

$$\langle S^z\rangle = \hbar\,\frac{(1+S+\varphi)\varphi^{2S+1} + (S+\varphi)(1+\varphi)^{2S+1}}{(1+\varphi)^{2S+1} - \varphi^{2S+1}}. \tag{43}$$

φ is the average magnon number,

$$\varphi(S) = \frac{1}{N}\sum_{\mathbf{q}}\frac{1}{e^{\beta E(\mathbf{q})} - 1}, \tag{44}$$

which determines $\langle S^z\rangle$ and many other spin correlations. Some typical examples are

$$\langle S^- S^+\rangle = 2\hbar\langle S^z\rangle\varphi(S), \tag{45}$$

$$\langle (S^z)^3\rangle = \hbar^3 S(S+1)\varphi(S) + \hbar^2\langle S^z\rangle[S(S+1) + \varphi(S)] - \hbar\langle (S^z)^2\rangle[1 + 3\varphi(S)]. \tag{46}$$

Via Eq. (36) all spin correlations [Eqs. (43)–(46)] are fixed by the electron self-energy [Eq. (16)].

The key quantity of ferromagnetism is the Curie temperature T_C. Performing the limiting processes

$$T \to T_C^{(-)}, \langle S^z\rangle \to 0^+ \tag{47}$$

one finds, from Eq. (43),

$$k_B T_C = \frac{2}{3}\hbar^2 S(S+1)\left[\frac{1}{N}\sum_{\mathbf{q}}\,[\hat{J}_0 - \hat{J}(\mathbf{q})]_{T_C}^{-1}\right]^{-1}. \tag{48}$$

The effective exchange integrals are temperature-dependent and have to be used here for $T \to T_C$.

For the results presented in Sec. IV, we used the self-energy approach of Ref. 32. The same approach was applied in Refs. 10,12,14,33, and 37, so that here we can refrain from presenting details of the method. As explained after Eq. (16) the self-energy of Ref. 32 depends on three types of expectation values: purely electronic terms, which are accessible with the single-particle Green function [Eq. (6)]; pure local-spin correlations such as Eqs. (43), (45), and (46); and mixed itinerant-electron–local–moment correlations, which are expressible by the ''higher'' Green functions [Eqs. (8) and (9)]. Eventually, we have a closed system of equations that can be solved self-consistently for the desired electronic and magnetic properties of the KLM. Some typical results are discussed in Sec. IV.

IV. MAGNETIC PROPERTIES

We have evaluated our theory for a s.c. lattice (bandwidth $W=1$ eV) to find out the magnetic properties of the ferromagnetic KLM. The latter are, however, strongly correlated with the electronic properties, so that they cannot be understood without referring to the itinerant-electron subsystem.

Figure 1 shows the quasiparticle density of states (QDOS) $\rho_\sigma(E)$ for a band occupation $n=0.2$ and a moderate exchange coupling $J=0.2$ eV. The self-consistently determined Curie temperature turns out to be $T_C=232$ K. In the low-temperature, ferromagnetic phase the QDOS exhibits a

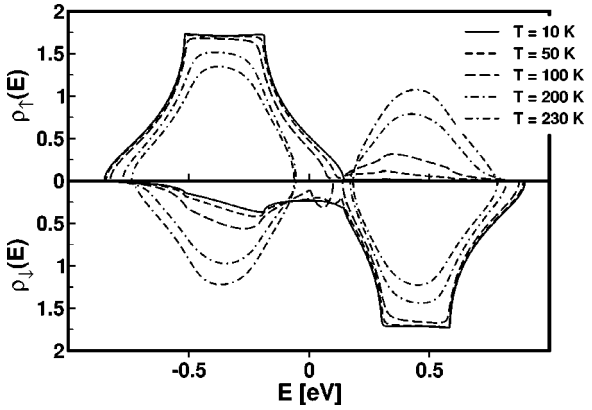

FIG. 1. Temperature-dependent quasiparticle density of states $\rho_\sigma(E)$ as a function of energy (the Kondo-lattice model). Parameters: $W=1$ eV, $S=7/2$, $J=0.2$ eV, and $n=0.2$. For these parameters the theory yields: $T_C=232$ K. The upper half for $\sigma=\uparrow$, and the lower half for $\sigma=\downarrow$.

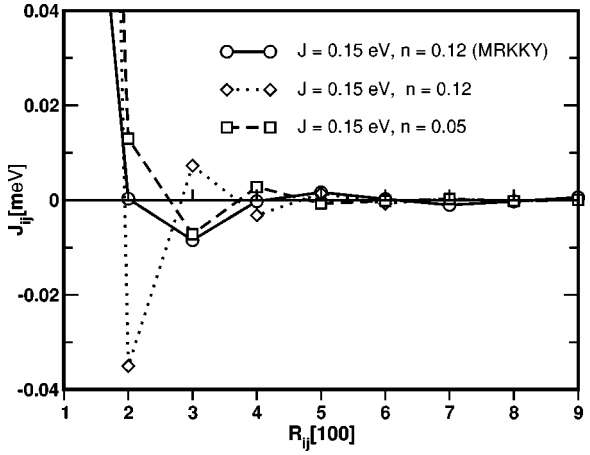

FIG. 2. Conventional RKKY exchange integrals $\hat{J}_{ij}$ as a function of the distance R_{ij} between lattice sites in the (100) direction for various band occupations n and different interband exchange couplings J. Parameters: $T=0$ K, $W=1$ eV, and the s.c. lattice. For comparison a respective example of the modified RKKY model is included.

remarkable temperature-dependence. For $T\rightarrow 0$ the $\uparrow$ spectrum becomes rather simple (the solid line in the upper half of Fig. 1) since the local-moment system is ferromagnetically saturated. A $\uparrow$ electron therefore has no chance to exchange its spin with the localized spins. That means that only the Ising-like interaction term in Eq. (4) really works, leading to a rigid shift of about $-\frac{1}{2}JS$ of $\rho_\uparrow(E)$ with respect to the ''free'' Bloch DOS $\rho_0(E)$. The down-spin QDOS, however, is much more complicated, because a $\downarrow$ electron can exchange its spin with the saturated localized spins. That can, e.g., be done by emitting a magnon $\hbar\omega(\mathbf{q})$, where the electron reverses its spin from $\downarrow$ to $\uparrow$. Consequently, this part of the spectrum (''scattering states'') exactly coincides with $\rho_\uparrow(E)$. However, the $\downarrow$ electron has another possibility to exchange its spin with the perfectly aligned localized spins. It can polarize its local spin surrounding by repeated magnon emission and reabsorption. This can even lead to a bound state, which we call the ''magnetic polaron.'' For the special case of a single $\downarrow$ electron in an otherwise empty conduction band coupled to a ferromagnetically saturated spin system the formation of the magnetic polaron can rigorously be shown[34,35] (see Fig. 1 in Ref. 37). Formation of a polaron costs more energy than magnon emission. Polaron states therefore build the upper part of the $\downarrow$ spectrum.

Magnon emission by the $\downarrow$ electron is equivalent to magnon absorption of a $\uparrow$ electron, with one exception: magnon absorption can occur only if there are mangnons in the system. This is not the case at $T=0$ in the ferromagnetic saturation. At finite temperature, however, there are magnons to be absorbed by $\uparrow$ electrons. Consequently, scattering states appear in the $\uparrow$ spectrum too. Because of the possible spin exchange both spin spectra occupy exactly the same energy region at all $T>0$. It is interesting to observe that even for such a moderate coupling $J=0.2$ eV ($W=1$ eV) there opens a gap between lower and upper quasiparticle subbands. In the lower subband the electron hops mainly over lattice sites, where it orients its spin parallel to the local moment, either without or with a preceding spin-flip by magnon emission (absorption). The upper subband consists of polaron states with finite lifetimes. Note that such elementary processes are not recognizable when for mathematical simplicity classical spins ($S\rightarrow\infty$) are assumed.[29–31] The QDOS in Fig. 1 clearly demonstrates that the conduction electrons are not at all fully spin polarized as is often used in DMFT treatments of the KLM.[21,31] This is true only in the unphysical limit $S\rightarrow\infty$.

Let us now concentrate the rest of the discussion on the magnetic properties of the KLM. They are determined by the respective behavior of the effective exchange integrals $\hat{J}_{ij}$.

Figure 2 shows the distance dependence of $\hat{J}_{ij}$ as it follows from the conventional RKKY exchange [Eq. (30)]. We recognize the well-known oscillating and long-range behavior. According to Eq. (31) two parameters influence the oscillation: the interband exchange J and the band occupation n. The ''amplitude'' of the oscillation is proportional to J^2, while the ''period'' is fixed via the chemical potential μ by the conduction electron density n. A remarkable temperature dependence is not observed.

The oscillations of the effective exchange integrals $\hat{J}_{ij}$ are less regular when higher-order polarization effects in the conduction band are taken into account [Eq. (36)]. Figures 3 and 4 show two examples for two different J's and several band occupations n.

The nonregular J dependence results from the fact, that J appears in Eq. (36) not only as a prefactor J^2 but also in a complicated manner via the Green function $G_{\mathbf{k}+\mathbf{q}\sigma}(E-\mu)$ of the polarized itinerant electron system. Of similar complexity is the n-dependence. In the conventional RKKY exchange [Eq. (31)] it comes into play only through the chemical potential in the Fermi function, while in the full expression [Eq. (36)], several correlation functions such as $\langle n_\sigma\rangle$, $\langle S^z n_\sigma\rangle$, $\langle S^{-\sigma}c^\dagger_\sigma c_{-\sigma}\rangle$, ... [Eqs. (15), (17), and (18)] contribute to the n dependence.

To demonstrate the influence of the interband exchange coupling J on the effective Heisenberg exchange integrals $\hat{J}_{ij}$

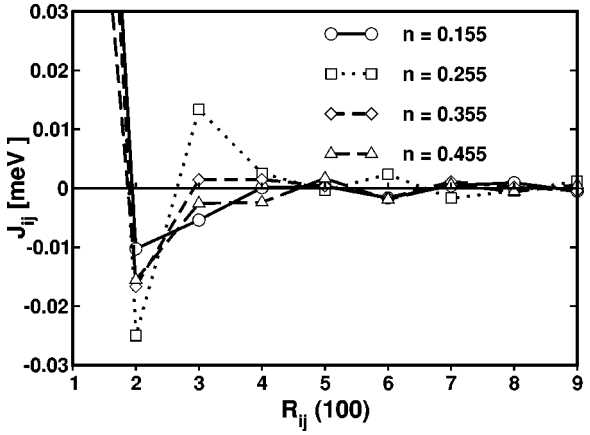

FIG. 3. Effective exchange integrals $\hat{J}_{ij}$ of the "modified" RKKY model as a function of the distance R_{ij} in the (100) direction for various band occupations. Parameters: $T=0$ K, $J=0.1$ eV, $W=1$ eV, s.c. lattice, and $S=7/2$.

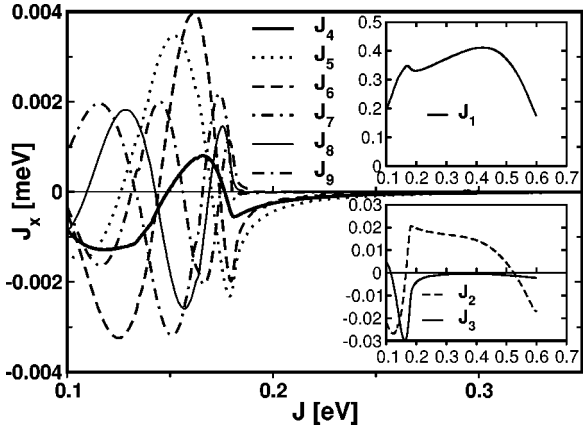

FIG. 5. Effective exchange integrals J_x between xth nearest neighbors ($x=1,2,\ldots,9$) in the (100) direction (s. c. lattice) as a function of the interband exchange coupling J. Parameters: $T=0$ K, $n=0.2$, $W=1$ eV, and $S=7/2$. Note the different energy scales.

in more detail, in Fig. 5 we plot the J dependence of J_x, $x=1,\ldots,9$. J_x is the effective exchange integral between sites separated along the (100) direction by x lattice constants.[39] $J_4,\ldots,J_9$ show an oscillatory structure in the weak-coupling region, being, one order of magnitude smaller than $J_{2,3}$ and even two orders of magnitude smaller than J_1. For $J>0.2$ eV $J_3,\ldots,J_9$ are so strongly damped that the magnetism of the KLM can be described by a Heisenberg model with next- and next-nearest-neighbor exchanges only. In the weak-coupling regime, however, the higher terms $J_3,\ldots,J_9$ cannot be neglected. Similar statements can be found in Ref. 21. Maybe this is an explanation of why CMR materials with high T_C can be described reasonably well[20] by a short-range Heisenberg model, while other materials with lower T_C exhibit strong deviations.[22–24] In any case the main contribution to the exchange coupling stems from the next-nearest-neighbors. J_1 determines the stable magnetic configuration. The negative slope of J_1 for $J>0.5$ eV already indicates a breakdown of the ferromagnetic order, which is more carefully inspected below.

The effective exchange integrals J_x exhibit an interesting electron-density dependence (Fig. 6). For low densities up to $n=0.3$, only J_1 is of importance. A Heisenberg model with nearest-neighbor exchange will appropriately describe the magnetic properties of the KLM.

For larger n, however, the exchange interaction becomes very long ranged; even J_9 is not negligible. From Figs. 5 and 6 we learn that the interplay between the local interband exchange J and the band occupation n does lead to a rather complicated behavior of the effective RKKY exchange integrals.

The effective exchange integrals determine the magnetic properties of the KLM. On the other hand, they depend on the electron self-energy which is strongly influenced by magnetic correlation functions. Typical examples of the latter are plotted in Fig. 7, calculated for $n=0.2$ and $J=0.2$ eV.

The electronic part of the self-consistent solution, represented by the temperature dependent QDOS, is shown in Fig. 1. The Curie temperature is found as to be $T_C=232$ K. The local-moment magnetization $\langle S^z\rangle$ is of Brillouin function type, and the qualitative shapes of the other spin correlations are also familiar from the pure Heisenberg model.

The key-quantity of ferromagnetism is the Curie tempera-

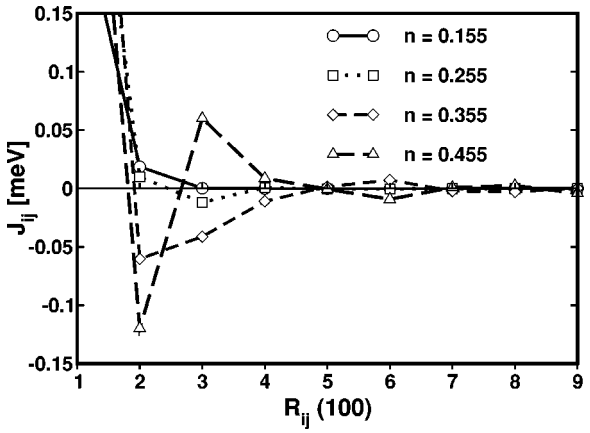

FIG. 4. The same as in Fig. 3, but for $J=0.2$ eV.

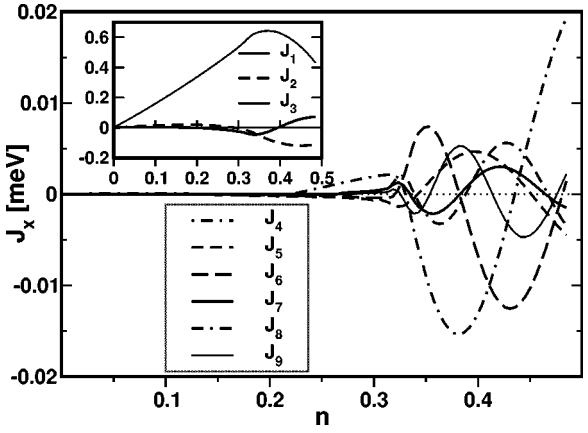

FIG. 6. The same effective exchange integrals as in Fig. 5, but now as a function of the band occupation n for $J=0.2$ eV.

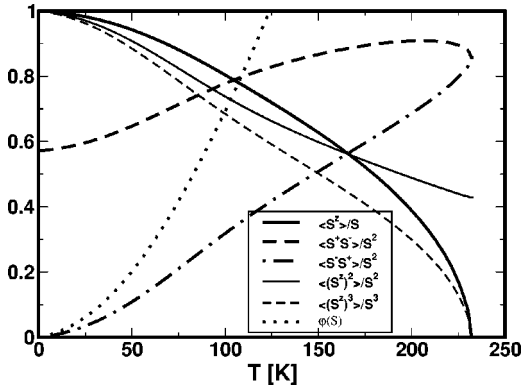

FIG. 7. Several spin correlation functions of the local-moment system in dependence on the temperature T. Parameters: $J=0.2$ eV, $n=0.2$, $S=7/2$, s. c. lattice, and $W=1$ eV. The theory, presented in the text, self-consistently yields $T_C=232$ K.

ture T_C. $T_C>0$ comes out as a consequence of an indirect coupling between the local moments, mediated by the spin polarization of itinerant electrons. Therefore, a strong particle density dependence of T_C has to be expected. Figure 8 shows $T_C(n)$ for various J's from the weak- to moderate-coupling regime, calculated according to Eq. (48).

Ferromagnetism appears for low particle (hole) densities, where the ferromagnetic region increases with increasing J. The T_C values are of realistic order. A similar n dependence of T_C was found in Ref. 40 within an extended multiband KLM. No ferromagnetism appears around half-filling ($n=1$). It can be speculated that antiferromagnetism, which is not considered within our approach, becomes stable near $n=1$. It is interesting to compare the results of Fig. 8 with their conterparts from conventional RKKY results (Fig. 9), using the same model parameters and the same T_C formula [Eq. (48)]. The T_C values are higher, but the region of n where ferromagnetism appears is distinctly narrower. The critical n, where T_C vanishes again, is independent of J, in contrast to the full theory in Fig. 8.

The Curie temperature shows a remarkable J dependence, which is plotted in Fig. 10 for various band occupations.

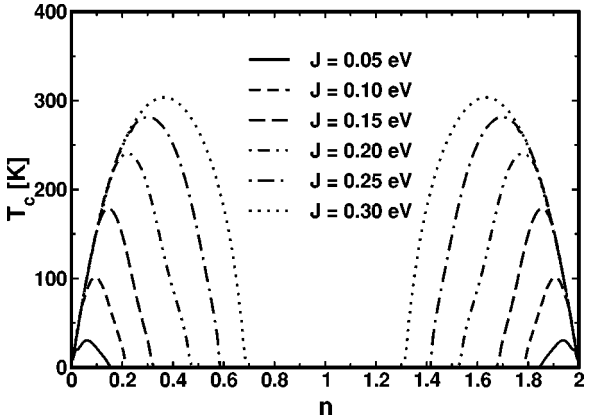

FIG. 8. Curie temperature as a function of the band occupation n for various interband exchanges couplings ($S=7/2$, s. c. lattice, and $W=1$ eV), calculated by use of the "modified" RKKY model.

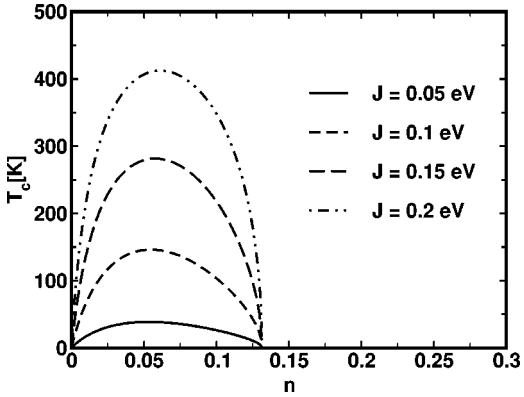

FIG. 9. The same as in Fig. 8, but calculated by use of the "conventional" RKKY model.

According to conventional RKKY theory one would expect a monotonic increase of J with increasing T_C. A mean-field evaluation of the effective Heisenberg model would lead to $T_C\sim J^2$.

Our theory becomes identical to conventional RKKY theory for small J. However, because of the random-phase approximation (RPA) treatment of the Heisenberg operator [Eq. (19)], there are deviations of T_C from J^2 behavior even for small J. A more important and very characteristic feature of our modified RKKY approach is the appearance of a critical coupling J_c for band occupations $n\geqslant 0.13$, below which no ferromagnetism occurs. This occurs for band fillings for which conventional RKKY theory fails to yield a ferromagnetic RPA solution (Fig. 9). Therefore the existence of J_c does not disprove the agreement of conventional and modified RKKY theory for small J. After a steep increase as function of J, T_C runs into a kind of saturation, where the plateau becomes smaller the larger the bandfilling n. With a further increase of J, the Curie temperature again decreases and disappears at an upper critical J value. The upper critical value of J coincides with the point where the stiffness constant D of the spin-wave dispersion [Eq. (41)] [$E(\mathbf{q})\approx D\mathbf{q}^2$ near the Γ point] becomes negative due to a respective behavior of the dominating effective exchange integrals J_1, J_2, and J_3

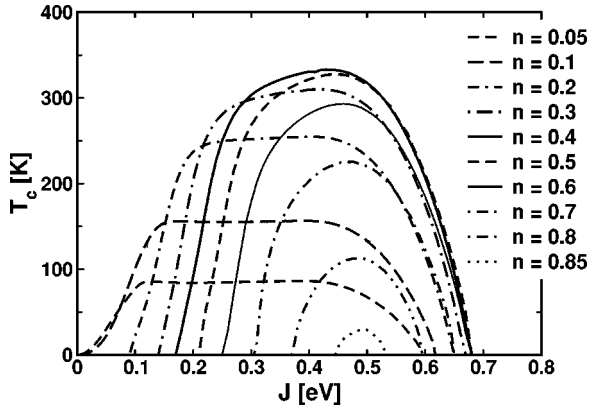

FIG. 10. Curie temperature as a function of the interband exchange coupling J for various band occupations n ($S=7/2$, s. c. lattice, and $W=1$ eV).

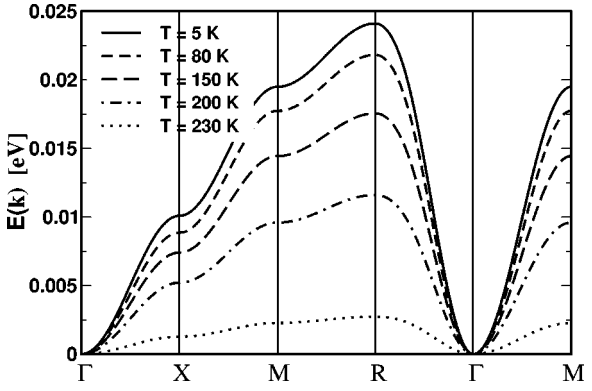

FIG. 11. Spin-wave dispersion of the ferromagnetic Kondo-lattice model as a function of the wave vector for different temperatures T. Parameters: $J=0.2$ eV, $n=0.2$, $S=7/2$, s. c. lattice, and $W=1$ eV. The self-consistently calculated Curie temperature is $T_C=232$ K.

[see the inset in Fig. 5 and Eq. (48)]. It can be suspected that for larger J the local-moment system becomes antiferromagnetic. Furthermore, it cannot be excluded that for still larger J's the system returns to ferromagnetism. These features are far beyond conventional RKKY theory, and due to higher-order polarization terms which come into play via the full Green function $G_{\mathbf{k}+\mathbf{q}\sigma}(E-\mu)$ in expression (36) for the effective exchange integral $\hat{J}(\mathbf{q})$. The manganites are often treated as ferromagnets with $J\rightarrow\infty$ in the KLM.[21] According to Fig. 10 this must be reexamined. For $J\approx W$ ferromagnetism is unlikely in the KLM.

The spin-wave dispersion, plotted in Fig. 11 for the main symmetry direction, obtains, via the effective exchange integrals, a distinct temperature dependence. Upon heating the dispersion relation uniformly softens, disappearing above T_C. This agrees qualitatively with a neutron-scattering study of the spin dynamics in the manganite $Pr_{0.63}Sr_{0.37}MnO_3$.[22] However, we do not share the view of the authors of Ref. 22, i.e., that the unexcepted results rule out a simple Heisenberg Hamiltonian. In our opinion the Heisenberg model works as long as the exchange integrals are renormalized in a proper way by the conduction-electron self-energy.

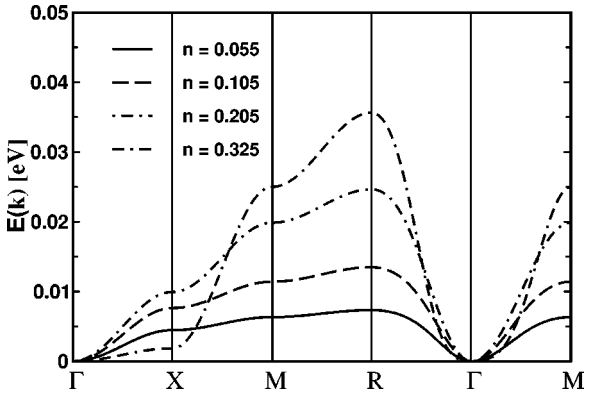

FIG. 12. The same as in Fig. 11, but for different band occupations at $T=0$ K.

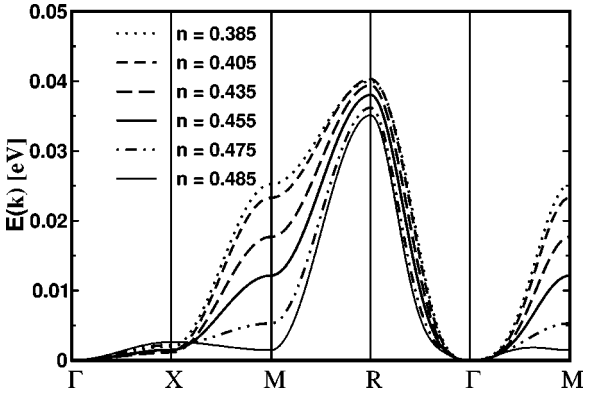

FIG. 13. The same as in Fig. 12, but for higher band occupations.

Changing the doping in III-V-based diluted magnetic semiconductors such as $Ga_{1-x}Mn_xAs$, or in manganites like $La_{1-x}Ca_xMnO_3$, does alter the band occupation and therewith the physics of these materials. It is therefore worthwhile to inspect the n dependence of the spin-wave dispersion. This is done in Fig. 12 for low band occupations and in Fig. 13 for higher band occupations for a model system with $J=0.2$ eV. For low densities (Fig. 12) we observe a strengthening of the magnon dispersion with increasing band filling, while the opposite is true for stronger fillings (Fig. 13). This is in accordance with the T_C behavior of Fig. 8. The band occupation $n=0.485$ is very close to the upper critical point of the $J=0.2$ eV curve in Fig. 8. The corresponding spin-wave dispersion in Fig. 13 shows a strong softening. A slightly higher n leads to parts of the Brillouin zone with negative magnon energies, i.e., the ferromagnetic ground state becomes unstable. The stiffness D, defined by $E(\mathbf{q})\approx D\mathbf{q}^2$ for $\mathbf{q}\rightarrow 0$, becomes negative. The points where $D=D(n,J,T=0)=0^+$ exactly coincide in our theory with those for which $T_C=T_C(n,J)\rightarrow 0$, directly derived from the local-moment magnetization $\langle S^z\rangle$. Qualitatively the same n dependence of the spin-wave dispersion was reported in Ref. 41.

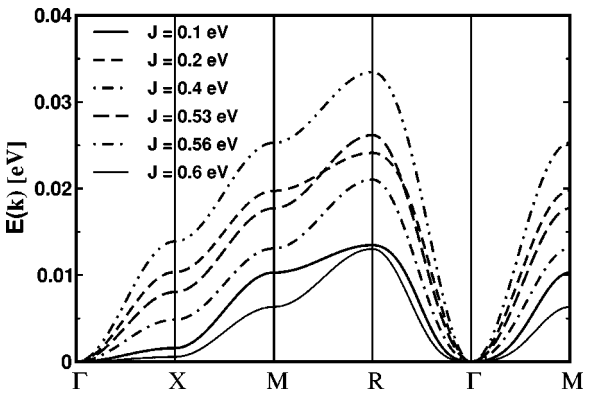

FIG. 14. The same as in Fig. 11, but for different interband exchange couplings J at $T=0$ K.

An example of the J dependence of the spin wave dispersion is presented in Fig. 14 ($n=0.2$, $T=0$ K, $S=\frac{7}{2}$). Again we observe a strengthening of the dispersion with J when T_C increases simultaneously and a softening, when T_C decreases (see Fig. 10).

V. SUMMARY

We have evaluated an approximate but self-consistent theory for a local-moment ferromagnet described within the framework of the ferromagnetic Kondo-lattice model. Candidates for this model are magnetic semiconductors (EuO, EuS), diluted magnetic semiconductors ($Ga_{1-x}Mn_xAs$), magnetic metals (Gd, Dy, Tb) and CMR materials ($La_{1-x}Ca_xMnO_3$). We have used a previously developed Green-function technique[32] for a detailed investigation of the magnetic properties of the exchange-coupled local moment-itinerant electron system. An extended RKKY mechanism has been worked out to derive magnetic phase diagrams as well as spin-wave dispersions. The latter show strong dependencies on the temperature (uniform softening for $T\rightarrow T_C$), on the band occupation n, and on the exchange coupling J. These dependencies are due to the respective behaviors of the effective exchange integrals between the localized moments. The effective exchange integrals are found by mapping the interband exchange ($s-f$) coupling, characteristic of the KLM, to an effective Heisenberg model. They turn out to be functions of the electronic self-energy, being therewith strongly temperature and electron density dependent. For small J our theory reproduces the results of the well-known conventional RKKY treatment, which represents second-order perturbation theory to the KLM. Already for very moderate J, however, substantial deviations appear, when higher-order terms of the induced conduction electron spin polarization bring their influence to bear.

For the future the presented approach has to be extended to antiferromagnetic moment configurations in order to complete the phase diagram. Furthermore, Coulomb correlations have to be introduced for the conduction-electron subsystem. Their neglect in the original KLM seems to be only poorly justified.

It is further intended to apply the modified RKKY procedure to the antiferromagnetic KLM ($J<0$) in order to investigate the interplay between ''Kondo screening'' and the RKKY procedure. This, however, requires a serious check whether or not the MCDA in its present form[32] is able to account for the subtle low-temperature (Kondo) physics.

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