Ferromagnetism and temperature-dependent electronic structure of hcp gadolinium

C. Santos and W. Nolting*
Lehrstuhl Festkörpertheorie, Institut für Physik, Humboldt-Universität zu Berlin, Newtonstraße 15, 12489 Berlin, Germany

V. Eyert
Institut für Physik, Universität Augsburg, Universitätsstraße 1, 86135 Augsburg, Germany
(Received 19 January 2004; published 16 June 2004)

We use a combination of a many-body model analysis with an "*ab initio*" band-structure calculation to derive the temperature-dependent electronic quasiparticle structure of the rare-earth metal gadolinium. As a local-moment system Gd is properly represented by the ferromagnetic (multiband) Kondo-lattice model [*s*-f(d-f) model]. The single-particle part of the model Hamiltonian is taken from an augmented spherical wave band calculation. The proposed method avoids the double counting of relevant interactions by exploiting an exact limiting case of the model and takes into account the correct symmetry of atomic orbitals. The weakly correlated 5d conduction bands become polarized via interband exchange coupling to the localized 4f levels with a distinct temperature dependence. This results in a Rudermann-Kittel-Kasuya-Yosida-type mechanism of coupling leading to the ferromagnetism of Gd. We get a self-consistently derived Curie temperature of 294.1 K and a $T=0$ moment of $7.71\mu_B$, surprisingly close to the experimental values. The striking temperature dependence of the 5d conduction bands provides insight into the origin of the temperature dependence of the photoemission data. The only parameter of the theory (interband exchange coupling J) is uniquely fixed by the band calculation.

DOI: 10.1103/PhysRevB.69.214412 PACS number(s): 71.10.−w, 75.30.Et, 71.20.Eh

I. INTRODUCTION

The rare-earth metal Gd is one of the four room-temperature elemental ferromagnetic metals; the others are Fe, Co, Ni. It crystallizes in the hcp structure with a lattice constant $a=3.629$ Å, $c/a=1.597$.[1] Magnetic properties result from the half-filled 4f shell ($L=0$, $J=S=\frac{7}{2}$) which gives rise to strictly localized magnetic moments. Conduction is dominated by the partially filled 5d/6s conduction bands. Gd is considered an almost ideal Heisenberg ferromagnet with a Curie temperature of $T_C=293.2$ K and a zero-temperature moment of $\mu(T=0)=7.63\mu_B$.[2] The moment indicates an induced polarization of the conduction bands of at least $0.63\mu_B$ due to an interband exchange coupling between itinerant band electrons and localized 4f electrons. The rather strict localization of the 4f wave function[3] prevents sufficient overlap for a direct exchange interaction between the 4f moments. The coupling between the moments is therefore mediated by polarized 5d/6s conduction electrons [Rudermann-Kittel-Kasuya-Yohida (RKKY)], i.e., strongly influenced by the electronic structure.

Although the ground-state of Gd is ferromagnetic, it is still a matter of debate how to get this fact by an "*ab initio*" band-structure calculation. Numerous investigations of the electronic ground-state properties of Gd have been performed in the recent past,[4–12] all in the framework of density functional theory (DFT). They provide a convincing description of ground-state properties such as the lattice constant, the hcp-crystal stability, the c/a ratio, the magnetic moment,[4,7] the bulk modulus, and the Fermi-surface paramters.[7,9] On the other hand, a standard local-density approach (LDA) to DFT predicts an antiferromagnetic ground state if the 4f electrons are considered as valence electrons. In a detailed analysis Kurz *et al.*[11] have demonstrated that the reason for the incorrect prediction of antiferromagnetism is the well known difficulty of LDA to correctly describe strongly localized electrons. The LDA calculation of Ref. 11 poses the nearly dispersionsless majority 4f bands some 4.5 eV below the Fermi energy while the minority 4f bands are directly above the Fermi energy leading to a certain itinerancy of the 4f electrons. These findings are at variance with the results of combined direct x-ray photoemission spectroscopy and inverse bremsstrahlung isochromat spectroscopy photoemission experiments[13] which observe occupied $4f\uparrow$ states at the binding energy (-7.44 ± 0.1) eV and unoccupied $4f\downarrow$ states at $(+4.04\pm0.2)$ eV, i.e., distinctly away from the Fermi edge. A special consequence of the incorrect binding energy of the down-spin 4f states is an extremely high density of states close to the Fermi edge and concomitant unrealistically large γ value of the electronic heat capacity.[4] The most important consequence of the incorrect $4f\downarrow$ binding energy, however, is the prediction of antiferromagnetism. All calculations, which treat the 4f electrons as valence electrons, irrespective of whether the LDA or the generalized gradient approximation (GGA) is applied, end up with an antiferromagnetic Gd ground state.[11,14] Today it is clear how to remove this inadequacy of LDA (GGA) "by hand."[11] One has to remove the $4f\downarrow$ states from the Fermi energy. This can be done simply by considering the 4f states as "core states," so that they are not allowed to hybridize with any other states on neighboring atoms.[11,14] Another way is to apply the so-called LDA+U method,[15] which introduces strong intra-atomic interactions of the localized states in a Hartree-Fock-like manner. The main effect is a splitting apart from the occupied and unoccupied 4f states,[10,16,11] i.e., in particular a removal of the minority 4f states from the Fermi edge.

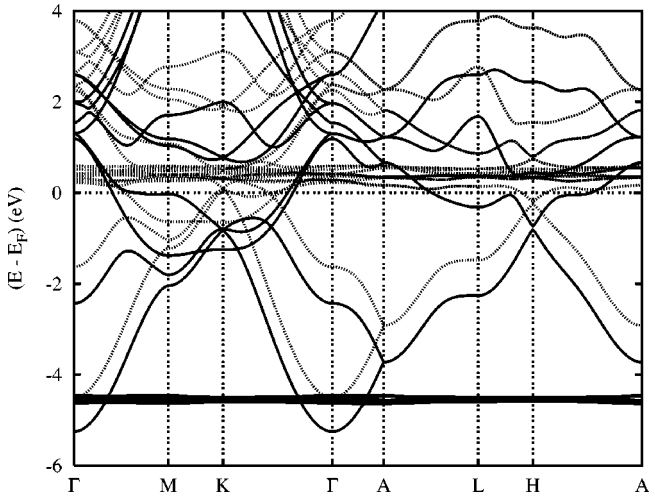

FIG. 1. Spin-resolved $T=0$ band structure of ferromagnetic Gd as a function of the wave vector, obtained by a scalar-relativistic ASW calculation. Solid lines for ↑ states, dotted lines for ↓ states. The energy zero is defined by the Fermi edge. The flat dispersions are the $4f$ levels.

Needless to say that in a real *ab initio* DFT procedure all electron states, including the $4f$ states, should be treated as valence states. To declare the $4f$ states as core states or to introduce at a convenient place the "Hubbard-U" surely corrupts the "first principles" character of the band calculation. The only motivation is to compensate the LDA artifact which prevents the correct ferromagnetic Gd ground state. We have recalculated the Gd band structure using a new implementation of the augmented spherical wave (ASW) method.[17,18] The $4f$ electrons have been treated as valence electrons and the scalar-relativistic approximation of the Dirac equation has been used. Although the antiferromagnetic configuration turns out to minimize the total energy, the ferromagnetic order has been assumed, first because it is closer to reality; second because we need these data for our further procedure. The results for some high-symmetry directions are plotted in Fig. 1. We recognize that the rather flat $4f$ dispersions are at incorrect binding energies. The fairly broad $5d/6s$ dispersions exhibit an exchange splitting with a weak **k** dependence. The result is in good agreement with other first-principles calculations that treat the $4f$ states as valence states.[6,8,11] The occupied and almost dispersionless $4f\uparrow$ bands provide the major part of the magnetic moment ($\approx 7\mu_B$) while the $5d$ splitting at the Fermi edge accounts for the experimentally observed excess moment of $\approx 0.63\mu_B$.

The mentioned $5d/6s$ exchange splitting must be induced since the $5d/6s$ electrons can be considered as only weakly correlated and *a priori* nonmagnetic. The splitting is obviously due to a strong interband exchange interaction between the flat $4f$ states and the extended $5d/6s$ conduction states. Precondition for that is a ferromagnetic order of the localized magnetic moments built by the half-filled $4f$ shells. The next neighbor distance is too large for a direct exchange interaction so the moment coupling is of indirect nature mediated by spin-polarized conduction electrons.

The induced $5d/6s$ exchange splitting is still a matter of controversial debate, in particular what concerns its temperature dependence.[12] Is it collapsing or noncollapsing for $T \rightarrow T_C$? Photoemission data appear to be not unique. Some experiments point to a collapsing ("Stoner-like") behavior,[19] others exhibit a splitting that does not shift very much with temperature ("spin-mixing") persisting in the paramagnetic phase.[20,21] In the latter case the demagnetization for $T \rightarrow T_C$ is reached by a redistribution of spectral weight rather than by a gradually increasing overlap of respective spin peaks. To find out what is really going on one needs a theory for the full temperature dependence of the electronic structure. Pure *ab initio* band calculations are restricted to $T=0$ being therefore insufficient for this purpose. Sandratskii and Kübler[22] have proposed a DFT-based theory where finite-temperature effects are simulated to a certain degree by a respective directional disorder of the spatially localized $4f$ moments. Even though this is an interesting ansatz, it certainly cannot replace the full statistical mechanics of the local-moment ferromagnet.

A key quantity of ferromagnetism is the Curie temperature T_C. It is the aim of each theory for a ferromagnetic material to approach T_C as quantitatively as possible. On the other hand, it is a very sensitive term to get. Several attempts have been started to estimate T_C from total-energy calculations by use of the LDA-DFT scheme.[11,23,24] For this purpose the energy data are inserted into simple mean-field formula for the magnetic transition temperature, very often arriving at astonishingly accurate T_C values. However, it is surely not unfair to state that such estimates cannot replace a full theory of the Gd ferromagnetism. The latter requires access to an electronic structure calculation which fully accounts for the temperature and correlations effects. Such a complete theory does not yet exist for the prototypical local-moment ferromagnet Gd. It is the aim of this paper to present a methodical approach to the temperature-dependent electronic structure of ferromagnetic local-moment metals with a direct application to Gd.

We present a theory of the electronic quasiparticle structure of the ferromagnetic $4f$ metal Gd that yields in a self-consistent manner the electronic as well as the magnetic properties. The approach shall work for arbitrary temperatures regarding in particular electron correlations effects. To get in this sense a realistic picture of Gd we combine a first principles band structure calculation with a many-body evaluation of a properly chosen theoretical model similar to previous work on the band ferromagnets Fe, Co, and Ni.[25] We consider the underlying proposal a continuation and extension of a previously published paper[26] that already dealt with the electronic quasiparticle structure of Gd. However, in the previous case we did not succeed in getting the ferromagnetism self-consistently, i.e., simply via the special electronic structure. There appeared a serious ambiguity on how to handle the d-band degeneracy, i.e., how to perform the necessary decomposition of $5d$ band in nondegenerate subbands. The decomposition in Ref. 26 did not emphasize the correct symmetry of atomic orbitals. Separate model calculations[27,28] revealed that the then-used d-band decomposition is inconvenient for a ferromagnetic order of the local $4f$ moments. We propose in this paper a new ansatz by which one gets correctly the electronic structure as well as the magnetic order of Gd.

The general procedure is briefly described in the following section. Central part of the procedure is a many-body evaluation of a properly chosen theoretical model. In Sec. III we introduce and justify the (multiband) Kondo-lattice model (KLM) as a good starting point for an at least qualitative understanding of local-moment ferromagnets such as Gd. We explain how to combine KLM with an LDA-DFT band calculation to arrive at quantitative statements. The KLM introduces a nontrivial many-body problem which for the general case cannot be treated rigorously. In Sec. IV our theoretical approach is represented. In the last step (Sec. V) we combine the model analysis with a band-structure calculation to get the electronic quasiparticle spectrum of Gd and its temperature dependence that, on the other hand, fixes the magnetic properties of the rare-earth metal as, e.g., the Curie temperature and the magnetic moment.

II. GENERAL PROCEDURE

Our study aims at a quantitative determination of the temperature-dependent electronic structure of the $4f$ ferromagnet Gd. The general concept is rather straightforward and consists of three steps. The important first step is the choice of a suitable theoretical model. The main physics is due to the existence and the mutual influence of two well defined subsystems, quasi-free electrons in rather broad conduction bands $(5d/6s)$ and localized electrons with extremely flat dispersions $(4f)$. The theoretical model is defined by its Hamiltonian

$$H = H_0 + H_1, \tag{1}$$

more strictly, by its interaction part H_1. This particular operator shall incorporate all those interactions which are responsible for the characteristic phenomena under study. In the present case H_1 should cover the decisive electron correlations which determine the magnetic properties and the characteristic temperature dependence of the electron quasiparticle spectrum. Our proposal for H_1 is discussed in the following section.

While there is no contribution of the f electrons to the kinetic energy, the part of the band electrons reads

$$\begin{aligned} H_0 &= \sum_{ij\sigma mm'} (T_{ij}^{mm'} - \mu\delta_{ij}\delta_{mm'}) c_{im\sigma}^{+} c_{jm'\sigma} \\ &= \sum_{\mathbf{k}\sigma mm'} (T_{\mathbf{k}}^{mm'} - \mu\delta_{mm'}) c_{\mathbf{k}m\sigma}^{+} c_{\mathbf{k}m'\sigma}. \end{aligned} \tag{2}$$

$c_{jm\sigma}^{+}(c_{jm\sigma})$ is the creation (annihilation) operator for a Wannier electron at site $\mathbf{R}_j$ in the orbital m with spin $\sigma(\sigma = \uparrow, \downarrow)$. $c_{\mathbf{k}m\sigma}^{+}(c_{\mathbf{k}m\sigma})$ is the respective Fourier transform

$$c_{\mathbf{k}m\sigma} = \frac{1}{\sqrt{N}} \sum_j c_{jm\sigma} e^{i\mathbf{k}\cdot\mathbf{R}_j}, \tag{3}$$

$$c_{jm\sigma} = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} c_{\mathbf{k}m\sigma} e^{-i\mathbf{k}\cdot\mathbf{R}_j}. \tag{4}$$

The hopping process from site $\mathbf{R}_j$ to site $\mathbf{R}_i$ may be accompanied by an orbital change $(m' \rightarrow m)$. $T_{ij}^{mm'}$ are the respective hopping integrals:

$$T_{ij}^{mm'} = \frac{1}{N} \sum_{\mathbf{k}} T_{\mathbf{k}}^{mm'} e^{i\mathbf{k}\cdot(\mathbf{R}_i - \mathbf{R}_j)}. \tag{5}$$

μ is the chemical potential. The single-particle part H_0 of the model Hamiltonian H stands, as usual, for the kinetic energy of the itinerant charge carriers and for their interaction with the lattice potential. However, it shall furthermore account for all those interactions which are not directly covered by H_1. By construction these interactions are not important for the magnetic properties and the temperature dependence of the electronic spectrum of the underlying ferromagnetic material (Gd). Nevertheless they may influence the rough structure of the spectrum so that they cannot be neglected if our study really aims at a quantitative description of Gd. For this reason we perform in the second step of our procedure a full self-consistent band-structure calculation within the LDA-DFT scheme in order to replace the single-particle energies $T_{\mathbf{k}}^{mm'}$ in H_0 by the effective energies of the LDA:

$$T_{\mathbf{k}}^{mm'} \rightarrow T_{\mathbf{k}}^{mm'}(\mathrm{LDA}). \tag{6}$$

Since the "other" interactions are, by construction of the model, not responsible for the temperature effects, we can incorporate them for any temperature, e.g., for $T=0$ where LDA works. It is therefore guaranteed that all the other interactions are well accounted by the single-particle energy renormalization (6). However, the problem of a double counting of just the relevant interactions, namely once explicitly in H_1 and then once more implicitly in H_0, must carefully be avoided. How we circumvent this problem in the case of the local-moment ferromagnet Gd is explained at a later stage.

In the third step we apply a many-body formalism in order to investigate how the effective single-particle energies change under the influence of the "relevant" interaction H_1 into temperature-dependent, carrier concentration-dependent (n), and possibly spin-dependent quasiparticle energies:

$$H_1 : T_{\mathbf{k}}^{mm'}(\mathrm{LDA}) \rightarrow E_{m\sigma}(\mathbf{k}, T, n). \tag{7}$$

The quasiparticle energies are derived as the poles of the single-electron Green-function matrix:

$$\hat{G}_{\mathbf{k}\sigma}(E) = \hbar[(E+\mu)\hat{\mathrm{I}} - \hat{T}_{\mathbf{k}} - \hat{M}_{\mathbf{k}\sigma}(E)]^{-1}. \tag{8}$$

$\hat{M}_{\mathbf{k}\sigma}(E)$ is the self-energy matrix, the determination of which solves the problem. $\hat{T}_{\mathbf{k}}$ is the hopping matrix. The elements of the Green-function matrix are the retarded single-electron Green functions:

$$\langle\langle c_{\mathbf{k}m\sigma}; c^{+}_{\mathbf{k}m'\sigma}\rangle\rangle_E = -i\int_0^{+\infty} dt e^{i/\hbar Et}\langle[c_{\mathbf{k}m\sigma}(t), c^{+}_{\mathbf{k}m'\sigma}(0)]_+\rangle. \tag{9}$$

$[\ldots,\ldots]_{+(-)}$ means the anticommutator (commutator) and $\langle\ldots\rangle$ is the thermodynamic average. The elements of the self-energy matrix formally solve the Green-function equation of motion:

$$\langle\langle[c_{\mathbf{k}m\sigma}, H_1]_-; c^{+}_{\mathbf{k}m'\sigma}\rangle\rangle_E \equiv \sum_{m''} M^{mm''}_{\mathbf{k}\sigma}(E)\langle\langle c_{\mathbf{k}m''\sigma}; c^{+}_{\mathbf{k}m'\sigma}\rangle\rangle_E. \tag{10}$$

We will discuss our results in terms of spectral densities and quasiparticle densities of states (Q-DOS), because both have a direct relationship to experiment. Except for respective transition matrix elements the spectral density expresses the bare line shape of an angle- and spin-resolved (direct or inverse) photoemission spectrum:

$$S_{\mathbf{k}m\sigma}(E-\mu) = -\frac{1}{\pi}\mathrm{Im}G^{mm}_{\mathbf{k}\sigma}(E-\mu) = -\frac{1}{\pi}\mathrm{Im}\langle\langle c_{\mathbf{k}m\sigma}; c^{+}_{\mathbf{k}m\sigma}\rangle\rangle_{E-\mu}. \tag{11}$$

An additional $\mathbf{k}$ summation yields the quasiparticle density of states (angle-averaged photoemission spectrum)

$$\rho_{m\sigma}(E) = \frac{1}{N\hbar}\sum_{\mathbf{k}} S_{\mathbf{k}m\sigma}(E-\mu) \tag{12}$$

that in general will be temperature dependent, carrier-concentration dependent, lattice structure dependent, and in particular for ferromagnetic systems, explicitly spin dependent.

III. THEORETICAL MODEL

A. Model Hamiltonian

We still have to fix the interaction part H_1 of the model Hamiltonian (1) for the correlated system of localized ($4f$) and delocalized ($5d$) electrons. We presume from the very beginning an on-site Coulomb interaction between electrons of different subbands:

$$H_1 = \frac{1}{2}\sum_{L_1,\ldots,L_4}\sum_{\sigma\sigma'} U_{L_1,\cdots,L_4} c^{+}_{L_1\sigma} c^{+}_{L_2\sigma'} c_{L_3\sigma'} c_{L_4\sigma}. \tag{13}$$

For simplicity we drop for the moment the site index i and the respective summation that will be reintroduced at the end of the following consideration. $L_1,\ldots,L_4$ denote the different bands, and $U_{L_1,\cdots,L_4}$ are the Coulomb matrix elements. Restricting the electron scattering processes caused by the Coulomb interaction to two involved subbands only, we get instead of Eq. (13)

$$H_1 = \frac{1}{2}\sum_{LL'}\sum_{\sigma\sigma'}\{U_{LL'} c^{+}_{L\sigma} c^{+}_{L'\sigma'} c_{L'\sigma'} c_{L\sigma} + J_{LL'} c^{+}_{L\sigma} c^{+}_{L'\sigma'} c_{L\sigma'} c_{L'\sigma} + J^{\star}_{LL'} c^{+}_{L\sigma} c^{+}_{L\sigma'} c_{L'\sigma'} c_{L'\sigma}\}. \tag{14}$$

In the case of Gd the band indices L and L' can be attributed either to a flat $4f$ band ($L\to f$) or to a broad ($5d/6s$) conduction band ($L\to m$). In an obvious manner we can then split the Coulomb interaction into three different parts,

$$H_1 = H_{dd} + H_{ff} + H_{df}, \tag{15}$$

depending on whether both interacting particles stem from a conduction band H_{dd}, or both from a flat band H_{ff}, or one from a flat band the other from a conduction band H_{df}. The first term, H_{dd}, refers to electron correlations in the broad conduction bands. We consider them not to be decisive for the characteristic Gd physics. According to our concept (Sec. II) H_{dd} does not enter explicitly our model being rather accounted for by the single-particle energy renormalization (6). H_{ff} is built by pure $4f$ correlations. The main influence of the $4f$ electrons on the Gd physics is due to the fact that they form permanent localized magnetic moments. So H_{ff} is unimportant as part of our model Hamiltonian and we are left with the interaction between localized and itinerant electrons:

$$H_{df} = \sum_{mf\sigma\sigma'}\Big\{U_{mf} c^{+}_{m\sigma} c^{+}_{f\sigma'} c_{f\sigma'} c_{m\sigma} + J_{mf} c^{+}_{m\sigma} c^{+}_{f\sigma'} c_{m\sigma'} c_{f\sigma} + \tfrac{1}{2} J^{\star}_{mf} c^{+}_{m\sigma} c^{+}_{m\sigma'} c_{f\sigma'} c_{f\sigma} + \tfrac{1}{2} J^{\star}_{fm} c^{+}_{f\sigma} c^{+}_{f\sigma'} c_{m\sigma'} c_{m\sigma}\Big\}. \tag{16}$$

The last two terms do not contribute since the Gd^{3+} $4f$ shell has its maximum spin $S=7/2$. All the seven $4f$ electrons have to occupy different subbands and none of the seven subbands will be doubly occupied. By use of the electron-spin operator,

$$\sigma^{+} = \hbar c^{+}_{\uparrow} c_{\downarrow}; \quad \sigma^{-} = \hbar c^{+}_{\downarrow} c_{\uparrow}; \quad \sigma^{z} = \frac{\hbar}{2}(n_{\uparrow} - n_{\downarrow}), \tag{17}$$

($n_\sigma = c^{+}_{\sigma} c_{\sigma}$) we get H_{df} in the following compact form

$$H_{df} = -\frac{2}{\hbar^2}\sum_{mf} J_{mf}\boldsymbol{\sigma}_m\cdot\boldsymbol{\sigma}_f + \sum_{mf}\left(U_{mf} - \frac{1}{2}J_{mf}\right) n_m n_f \tag{18}$$

with $n_{m(f)} = n_{m(f)\uparrow} + n_{m(f)\downarrow}$. For all processes of interest the number of f electrons per site is fixed, n_f is therefore only a c number. The last term in Eq. (18) does not really provide an fd interaction. It leads only to a rigid shift of the atomic levels being therefore fully accounted for by the renormalization (6) of the single-particle part of the Hamiltonian. By defining the spin operator $\mathbf{S}$ of the local f moment

$$\mathbf{S} = \sum_{f}\boldsymbol{\sigma}_f, \tag{19}$$

and by assuming that the interband exchange J_{mf} is independent of the special index-pair m,f

$$J_{mf} \equiv \tfrac{1}{2}J, \tag{20}$$

the interaction term reads after the reintroduction of the lattice site dependence

$$H_{df} = -\frac{J}{\hbar^2}\sum_{im}\boldsymbol{\sigma}_{im}\cdot\mathbf{S}_i = -\frac{J}{2\hbar}\sum_{im\sigma}\{z_\sigma S^z_i n_{im\sigma} + S^{\sigma}_i c^{+}_{im-\sigma} c_{im\sigma}\}. \tag{21}$$

Here we have used the abbreviations

$$S_j^\sigma = S_j^x + iz_\sigma S_j^y; \quad z_\sigma = \delta_{\sigma\uparrow} - \delta_{\sigma\downarrow}. \tag{22}$$

The single-band version (nondegenerate *s* band) of Eq. (21) is well known as the interaction part of the so-called (KLM),[29] in the older literature more appropriately denoted as *s-f* or *s-d* model.[12,30,31] In the multiband case we have in H_{df} simply an additional summation over the orbital index *m*. The first term of Eq. (21) describes an Ising-like interaction of the two spin operators, while the other provides spin-exchange processes between localized moment and itinerant electron. Spin exchange may happen by three different elementary processes: Magnon emission by an itinerant ↓-electron, magnon absorption by a ↑-electron, and also formation of a quasiparticle, which is called "magnetic polaron." The latter can be understood as a propagating electron "dressed" by a virtual cloud of repeatedly emitted and reabsorbed magnons corresponding to a polarization of the immediate localized spin neighborhood.

Our model Hamiltonian, built up by the partial operators (2) and (21),

$$H = H_0 + H_{df}, \tag{23}$$

can be considered as "multiband Kondo-lattice model." While in the interaction part H_{df} the multiband aspect appears only as an additional summation, the subbands are intercorrelated via the single-particle term H_0.

An important model parameter is of course the effective coupling constant JS/W where *W* is the width of the "free" Bloch-band and *S* the local spin value. It turns out that in particular the sign of *J* is decisive. Other model parameters are the lattice structure and the band occupation

$$n = \sum_{m\sigma} \langle n_{m\sigma} \rangle. \tag{24}$$

In case of an *s* band *n* is a number in between 0 and 2.

B. Exact limiting case

The many-body problem provoked by the model Hamiltonian (23) is rather sophisticated, up to now not exactly solvable for the general case. Fortunately, however, there exists a nontrivial, very illustrative limiting case which is rigorously tractable, nevertheless exhibiting all the above mentioned elementary excitations processes.[32–34] It refers to a single electron in an otherwise empty conduction band being coupled to a ferromagnetically saturated moment system. Such a situation is met, e.g., for the ferromagnetic semiconductor EuO at $T=0$. Because of the empty band and the totally aligned spin system the hierarchy of equation of motions of the single-electron Green function (8) and (9) decouples exactly. One can exploit exact relationships of the following kind:

$$\langle \ldots c_{im\sigma} \rangle = \langle c_{im\sigma}^+ \ldots \rangle = 0; \quad \langle \ldots S_i^+ \rangle = \langle S_i^- \ldots \rangle = 0,$$

$$\langle \ldots S_i^z \rangle = \langle S_i^z \ldots \rangle = \hbar S \langle \ldots \rangle. \tag{25}$$

A troublesome but straightforward calculation then arrives at the following result for the self-energy matrix (8)

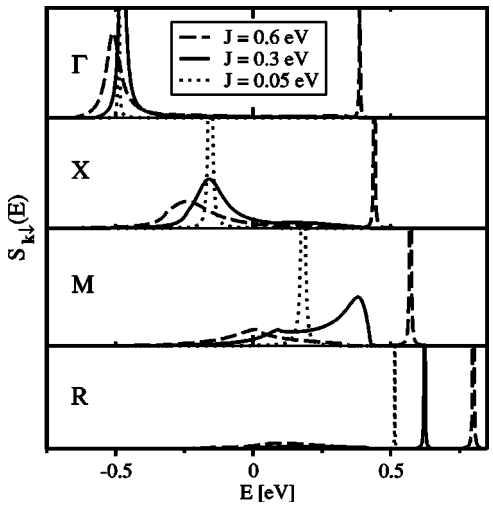

FIG. 2. Exact ↓ spectral density of the single-band Kondo-lattice model at $T=0$ as function of the energy for several symmetry points in the first Brillouin zone and for different exchange couplings *J*. Parameters: $S=1/2$, $W=1$ eV, $n=0$, *sc* lattice.

$$\hat{M}_{\mathbf{k}\sigma}(E) = -\frac{1}{2} z_\sigma JS\hat{\mathbf{I}} + (1 - z_\sigma) \frac{\frac{1}{4} J^2 S \frac{1}{\hbar} \hat{G}_0\left(E + \frac{1}{2}JS\right)}{\hat{\mathbf{I}} - \frac{1}{2} J \frac{1}{\hbar} \hat{G}_0\left(E + \frac{1}{2}JS\right)}, \tag{26}$$

$$\frac{1}{\hbar}\hat{G}_0(E) = \frac{1}{N}\sum_{\mathbf{k}} [(E+\mu)\hat{\mathbf{I}} - \hat{T}_{\mathbf{k}}]^{-1}. \tag{27}$$

The ↑ spectrum is especially simple because the ↑ electron cannot exchange its spin with the parallely aligned local spin system. Only the Ising-type interaction in Eq. (21) takes care for a rigid shift of the self-energy by $-\frac{1}{2}JS$. The spectral densities (11) are δ functions representing quasiparticles with infinite lifetimes. Real correlation effects appear, however, in the ↓ spectrum. The essentials can be seen already for a nondegenerate *s* band. Figure 2 shows the energy dependence of the ↓ spectral density $S_{\mathbf{k}\downarrow}(E)$ for some symmetry points. Furthermore, we have chosen a *sc* lattice, $S=1/2$ and $W=1$ eV.

For weak coupling (e.g., $J=0.05$ eV) the spectral density consists of a single pronounced peak. The finite width points to a finite quasiparticle lifetime due to some spin-flip processes, but the sharpness of the peaks indicates a long living quasiparticle. This changes drastically even for rather moderate effective exchange couplings JS/W. One observes in certain parts of the Brillouin zone, for strongly coupled systems even in the whole Brillouin zone, that the spectral density splits into two parts. The sharp high-energy peak belongs to the formation of the magnetic polaron while the broad low-energy part consists of scattering states due to magnon emission by the ↓ electron. As long as the polaron peak is above the scattering spectrum the quasiparticle has even an infinite lifetime. The scattering spectrum is in general rather broad because the emitted magnon can carry away any wave

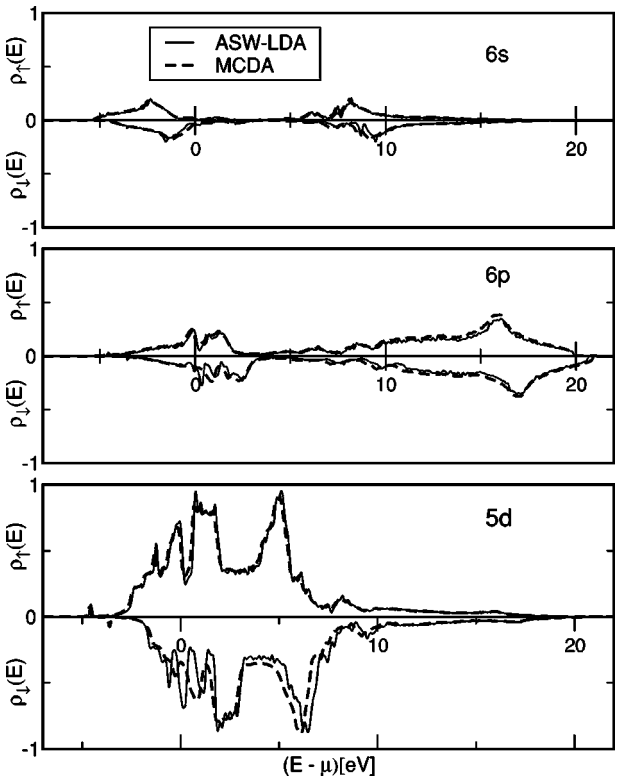

FIG. 3. Spin resolved densities of states of the 6*S*, 6*P*, and 5*d* bands of Gd as functions of the energy at $T=0$. Full lines for the ASW-LDA calculation, broken lines for our theory. $\uparrow(\downarrow)$ spectra in upper (lower) halves of the figures.

vector from the first Brillouin zone. Because of the concomitant spin-flip magnon emission can happen only if there are $\uparrow$ states within reach. Therefore, the scattering part extends just over that energy region where $\rho_{\uparrow}(E) \neq 0$. Sometimes, as e.g., for $J=0.6$ eV at the Γ point (Fig. 2), the scattering part is surprisingly bunched together to a prominent peak, therefore certainly visible in a respective (inverse) photoemission experiment. Note that the results in Fig. 2 are exact and free of any uncontrollable approximation. They exhibit typical correlation effects which are by no means reproducible by a single-electron theory.

Very important for the following procedure is the simple $\uparrow$ result. It tells us that at ($T=0$, $n=0$) the df exchange interaction takes care only for a rigid shift of the total energy spectrum without any deformation, which is therefore identical to the free Bloch spectrum. Furthermore for this special case a mean-field approximation turns out to be exact. Though not exactly provable, many reliable approaches[27,32,35] show that this holds, at least to a good approximation, for finite band occupation, too. This will be demonstrated in Fig. 3 for the actual case of Gd.

C. Band-structure calculations

As described in Sec. II the hopping integrals $T_{ij}^{mm'}$ in the single-particle Hamiltonian H_0 (2) have to absorb the influences of all those interactions which are not directly covered by our model Hamiltonian (23). For this purpose we have performed a spin polarized scalar-relativistic ASW band calculation for ferromagnetic hcp Gd. The result is plotted in Fig. 1. The $4f$ electrons are considered as valence electrons. In Sec. I we already commented on the wrong position of the $4f$ levels. However, we need as input for the determination of the quasiparticle spectrum only the $\uparrow$ part of the band-structure because of the following reason. The main problem, when using LDA-DFT results as renormalized single-particle input, is to avoid a double counting of the key interband exchange coupling, namely, explicit inclusion in the interaction part H_1 of the model Hamiltonian and then inclusion once more implicitly by the effective single-particle input. The exact limiting case of the last section gives the hint on how to circumvent this double-counting problem. For an empty band ($n=0$) and ferromagnetic saturation the $\uparrow$ spectrum is only rigidly shifted by a constant energy amount $\left(-\frac{1}{2}JS\right)$ compared to the free Bloch spectrum. As mentioned, model approaches convincingly demonstrate that this remains true for finite band occupation (less than half-filled bands) too, at least to a very good approximation. For less than half-filled bands we therefore can identify the ($T=0$, $\sigma=\uparrow$)-LDA results (without the $4f$ part) with the free Bloch energies. By this procedure we do not explicitly switch off the df interband exchange but rather exploit the fact that for the mentioned special case the interband exchange leads only to a trivial rigid shift. The incorrect binding energy of the LDA-$4f\downarrow$ states is not of overwhelming concern because we need only the $\uparrow$ spectrum. On the other hand, we can be sure that all other interactions, e.g., the Coulomb interaction of the $5d$ electrons, are excellently accounted for by the LDA. We note in passing, that for more than half-filled bands we would have to take the ($T=0$, $\sigma=\downarrow$)-spectrum (particle-hole symmetry).

After defining the single-particle input there remains only one parameter, namely the exchange coupling J. It is not considered as a free parameter, but taken from the band-structure calculation. It is commonly accepted that an LDA treatment of ferromagnetism is quite compatible with a mean-field ansatz,[17,36] so that the exchange splitting in Fig. 1 should amount to $\Delta=JS$ (see following section). We realize, however, that the assumption of a rigid splitting is too simple. A slight energy and wave-vector dependence of the exchange splitting is found by LDA, too. We have therefore averaged the $T=0$ splitting over N_p prominent features in the Q-DOS of Gd arriving at

$$J=\frac{1}{N_p S}\sum_p \Delta_p = 0.3 \text{ eV}. \tag{28}$$

This is of the same order of magnitude as found for Eu chalcogenides.[37,38] There are then no other parameters in our theory.

The validity of the above assumptions will be demonstrated later with Fig. 3.

IV. MANY-BODY EVALUATION

The multiband-KLM (23) does not allow a rigorous solution except for some special cases. Approximations are un-

avoidable. There are two partial problems to be solved, one concerning the ferromagnetism mainly provoked by the localized $4f$ moments, the other dealing with the temperature reaction of the conduction-band states due to the magnetic state of the moment system. In addition, both parts have to be combined in a self-consistent manner.

For the electronic part we have developed in the past several approaches,[27,35,39] which all lead, at least qualitatively, to the same result. The "interpolating self-energy approach"[35,39] is in particular trustworthy for almost empty or almost full bands. For intermediate fillings as in the case of Gd the "moment conserving decoupling approach" (MCDA) (Ref. 27) seems to be more recommendable. So we use it here. Since this approach has been exhibited in detail in Ref. 27 we can restrict ourselves in the following to the central parts which are vital for the understanding of the underlying procedure. To make the representation as clear as possible we develop the method in the following section for the special case of a nondegenerate band. The orbital index is then superfluous. The generalization for the degenerate case is straightforward. The investigation of the magnetic part follows in the Sec. IV B.

A. Conduction electron self-energy

According to Eq. (8) the solution of the problem needs the knowledge of the self-energy $M_{\mathbf{k}\sigma}(E)$. The above-mentioned MCDA is a nonperturbational Green-function theory. It starts from the equation of motion of the site-dependent single-electron Green function (8):

$$\sum_m [(E+\mu)\delta_{im} - T_{im}]G_{mj\sigma}(E) = \hbar\,\delta_{ij} - \frac{1}{2}J[z_\sigma \Gamma_{ii,j\sigma}(E) + F_{ii,j\sigma}(E)]. \tag{29}$$

Our approximation attacks the equations of motion of the "Ising function"

$$\Gamma_{im,j\sigma}(E) = \langle\langle S_i^z c_{m\sigma}; c_{j\sigma}^+\rangle\rangle_E, \tag{30}$$

and the "spin-flip function":

$$F_{im,j\sigma}(E) = \langle\langle S_i^{-\sigma} c_{m-\sigma}; c_{j\sigma}^+\rangle\rangle_E. \tag{31}$$

These equations of motion contain still higher Green functions which are decoupled to get a closed system of equations. Let us exemplify the procedure by a "higher" Green function of the type $\langle\langle A_i[c_{l\sigma}, H_{df}]_-; c_{j\sigma}^+\rangle\rangle_E$, where A_i is any combination of local moment and band operators. The off-diagonal terms $i \neq m$ are approached by use of the self-energy elements $M_{lr\sigma}$ ("self-energy trick"), in a certain sense as a generalization of the exact Eq. (10):

$$\langle\langle A_i[c_{l\sigma}, H_{df}]_-; c_{j\sigma}^+\rangle\rangle_E \Rightarrow \sum_r M_{lr\sigma}(E)\langle\langle A_i c_{r\sigma}; c_{j\sigma}^+\rangle\rangle_E. \tag{32}$$

The right-hand side is a linear combination of "lower" Green functions with the self-energy elements as self-consistently to be determined coefficients. To account for the strong local correlations the diagonal terms $i=l$ are handled with special care:

$$\langle\langle A_i[c_{i\sigma}, H_{df}]_-; c_{j\sigma}^+\rangle\rangle_E = \alpha_\sigma G_{ij\sigma}(E) + \beta_\sigma \Gamma_{ii,j\sigma}(E) + \gamma_\sigma F_{ii,j\sigma}(E). \tag{33}$$

Such an ansatz is constructed in such a way that all known exact limiting cases (atomic limit, ferromagnetic saturation, local spin $S=1/2$, $n=0$, $n=2,\ldots$) are exactly fulfilled. The at first unknown coefficients α_σ, β_σ, γ_σ are eventually found by equating exact high-energy expansion (spectral moments) of the self-energy. As the other above-mentioned methods[35,39] the MCDA arrives at the following structure of the self-energy:

$$M_{\mathbf{k}\sigma}(E) = -\tfrac{1}{2}Jz_\sigma\langle S^z\rangle + J^2 D_{\mathbf{k}}(E;J). \tag{34}$$

Restriction to the first term, only, is just the mean-field approach to the KLM, which is correct for sufficiently weak couplings J, being mainly due to the Ising-part in Eq. (21). Without the second part it would give rise to a spin-polarized splitting of the conduction band. The term $D_{\mathbf{k}\sigma}(E;J)$ is more complicated being predominantly determined by spin exchange processes due to the spin-flip term in the Hamiltonian (21). It is a complicated functional of the self-energy itself, and that for both spin directions, i.e., Eq. (34) is an implicit equation for $M_{\mathbf{k}\sigma}(E)$ and not at all an analytic solution. $D_{\mathbf{k}\sigma}(E;J)$ depends, furthermore, on mixed spin correlations such as $\langle S_i^z n_{i\sigma}\rangle$, $\langle S_i^+ c_{i\downarrow}^+ c_{i\uparrow}\rangle,\ldots$, built up by combinations of localized-spin and itinerant-electron operators. Fortunately, all these mixed correlations can rigorously be expressed via the spectral theorem by any of the Green functions involved in the hierarchy of the MCDA. However, there are also pure local-moment correlation functions of the form $\langle S_i^z\rangle$, $\langle S_i^+ S_i^-\rangle$, $\langle (S_i^z)^2\rangle,\ldots$ which also have to be expressed by the electronic self-energy $M_{\mathbf{k}\sigma}(E)$.

B. Modified RKKY interaction

To get such expectation values of local-spin combinations we map the interband exchange operator (21) on an effective Heisenberg Hamiltonian:[27,28]

$$H_{df} = -\frac{J}{\hbar^2}\sum_{im} \boldsymbol{\sigma}_{im}\cdot\mathbf{S}_i \Rightarrow -\sum_{ij} J_{ij}^{\mathrm{eff}}\mathbf{S}_i\cdot\mathbf{S}_j. \tag{35}$$

We use here again the full multiband version. The mapping is done by averaging out the electronic degrees of freedom $\boldsymbol{\sigma}_{im} \rightarrow \langle\boldsymbol{\sigma}_{im}\rangle^{(c)}$. That means, in the last analysis, to determine the expectation value $\langle c_{\mathbf{k}+\mathbf{q}m\sigma}^+ c_{\mathbf{k}m\sigma'}\rangle^{(c)}$. The averaging $\langle\cdots\rangle^{(c)}$ has to be done in the conduction electron subspace where the local spins $\mathbf{S}_i$ can be treated as classical variables:

$$\langle c_{\mathbf{k}+\mathbf{q}m\sigma}^+ c_{\mathbf{k}m\sigma'}\rangle^{(c)} = \frac{1}{\Xi'}\mathrm{Tr}(e^{-\beta H'} c_{\mathbf{k}+\mathbf{q}m\sigma}^+ c_{\mathbf{k}m\sigma'}). \tag{36}$$

H' is formally the same as in Eq. (23), except for the fact that for the averaging process the f spin operators are to be considered as c numbers, therefore not affecting the trace. Ξ' is the corresponding grand partition function. We use the spectral theorem for the "restricted" Green function,

$$G^{mm'}_{\mathbf{k}\sigma',\mathbf{k}+\mathbf{q}\sigma}(E) = \langle\langle c_{\mathbf{k}m\sigma'}; c^{+}_{\mathbf{k}+\mathbf{q}m'\sigma}\rangle\rangle_E, \tag{37}$$

to fix the expectation value (36). Equation (37) stands for the usual definition (9) of a retarded Green function, only the averages have to be done in the Hilbert space of H'. The equation of motion of $\hat{G}$ reads (in matrix representation with respect to the orbital indices m, m')

$$\begin{aligned}\hat{G}_{\mathbf{k}\sigma',\mathbf{k}+\mathbf{q}\sigma}(E) = {} & \delta_{\mathbf{q},\mathbf{0}}\delta_{\sigma\sigma'}\hat{G}^{(0)}_{\mathbf{k}}(E) \\ & - \frac{J}{2N}\sum_{i\sigma''\mathbf{k}'}\left[e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}_i}(\mathbf{S}_i\cdot\sigma)_{\sigma'\sigma''}\cdot\hat{G}^{(0)}_{\mathbf{k}}(E)\right. \\ & \times\hat{G}_{\mathbf{k}'\sigma',\mathbf{k}+\mathbf{q}\sigma}(E) + e^{i[\mathbf{k}'-(\mathbf{k}+\mathbf{q})]\cdot\mathbf{R}_i}(\mathbf{S}_i\cdot\sigma)_{\sigma''\sigma} \\ & \left.\times\hat{G}_{\mathbf{k}\sigma',\mathbf{k}'\sigma''}(E)\hat{G}^{(0)}_{\mathbf{k}+\mathbf{q}}(E)\right].\end{aligned} \tag{38}$$

This equation is exact and can be iterated up to any desired accuracy. $\hat{G}^{(0)}_{\mathbf{k}}(E)$ is the Green-function matrix of the free electron system:

$$\hat{G}^{(0)}_{\mathbf{k}}(E) = \hbar[(E+\mu)\hat{\mathbf{I}} - \hat{T}_{\mathbf{k}}]^{-1}. \tag{39}$$

If we stop the iteration in Eq. (38) after the first nontrivial step, i.e., replacing $\hat{G}$ in the right-hand side by the free Green-function matrix, then we arrive at the well-known RKKY result,[28] which can be equivalently derived by use of conventional second-order perturbation theory with respect to J starting from the unpolarized conduction electron gas. To incorporate the exchange-induced conduction electron-spin polarization to a higher degree we replace the restricted Green function in the right-hand side of Eq. (38) not by the free but by the full single-electron Green-function matrix $\hat{G}_{\mathbf{k}\sigma}(E)$ defined in Eq. (8):

$$\hat{G}_{\mathbf{k}'\sigma'',\mathbf{k}+\mathbf{q}\sigma}(E) \rightarrow \delta_{\mathbf{k}',\mathbf{k}+\mathbf{q}}\delta_{\sigma''\sigma}\hat{G}_{\mathbf{k}+\mathbf{q}\sigma}(E), \tag{40}$$

$$\hat{G}_{\mathbf{k}\sigma',\mathbf{k}'\sigma''}(E) \rightarrow \delta_{\mathbf{k},\mathbf{k}'}\delta_{\sigma'\sigma''}\hat{G}_{\mathbf{k}\sigma'}(E). \tag{41}$$

After some manipulations that replacement leads to the following effective exchange integrals

$$\begin{aligned}J^{\mathrm{eff}}_{ij} = {} & \frac{J^2}{8N\pi}\sum_{\mathbf{k}\mathbf{q}m\sigma} e^{-i\mathbf{q}\cdot(\mathbf{R}_i-\mathbf{R}_j)}\int_{-\infty}^{+\infty} dE f_{-}(E) \\ & \times\mathrm{Im}[(\hat{G}_{\mathbf{k}\sigma}(E-\mu)\hat{G}^{(0)}_{\mathbf{k}+\mathbf{q}}(E-\mu))^{mm} \\ & + (\hat{G}^{(0)}_{\mathbf{k}}(E-\mu)\hat{G}_{\mathbf{k}+\mathbf{q}\sigma}(E-\mu))^{mm}].\end{aligned} \tag{42}$$

These effective exchange integrals are functionals of the electronic self-energy $\hat{M}_{\mathbf{k}\sigma}(E)$ getting therewith a distinct temperature—and carrier concentration dependence. Neglecting $\hat{M}_{\mathbf{k}\sigma}(E)$, i.e., replacing in Eq. (42) the full by the free Green function, leads to the multiband version of the conventional RKKY-exchange integrals.[40,41] Via $\hat{M}_{\mathbf{k}\sigma}(E)$ higher-order terms of the conduction electron spin polarization enter the "modified" RKKY (42) which is therefore not restricted to weak couplings only.

To get from the effective Heisenberg Hamiltonian (35) the magnetic properties of the multiband KLM we apply the standard Tyablikow approximation[42] which is known to yield convincing results in the low as well as high-temperature region. All the above mentioned local-moment correlations are then expressed by the electronic self-energy. We therefore end up with a closed system of equations that can be solved self-consistently for all quantities of interest. For a detailed discussion of the so-found properties of the single-band KLM the reader is referred to our previous publications.[27,28,43] We use the theory in the following section to find the electronic and the magnetic properties of the ferromagnetic $4f$ metal Gadolinium.

V. MAGNETIC AND ELECTRONIC PROPERTIES OF GADOLINIUM

Figure 3 shows the partial ($5d$, $6s$, $6p$) quasiparticle densities of states at $T=0$, as they are found by our method and compared to the pure ASW-LDA. The $\uparrow$ spin parts are almost identical for both methods. That confirms our procedure, explained in Sec. III C, for the combination of the many-body model evaluation and the first principles band-structure calculation. Obviously a double counting of any decisive interaction has almost perfectly been avoided. The still observable very small deviations might be due to the finite band occupation. The statement that the up-spin spectrum at $T=0$ is only rigidly shifted, Eq. (26), compared to the free spectrum can be proved, strictly speaking, only for empty bands. As mentioned, a lot of reliable approaches[27,28,39] support the assumption that this is true, at least to a very good approximation, for finite carrier densities, too. However, slight deviations may appear. Furthermore, the band- and wave-vector-independence of the exchange coupling J (28) is surely an oversimplification and may also contribute to the deviations in the $\uparrow$ spectrum. Nevertheless, the near perfect agreement between LDA and model results demonstrate that there are hardly any exchange-caused correlation effects in the $\uparrow$ spectrum of the local-moment ferromagnet at $T=0$ (ferromagnetic saturation).

The $\downarrow$ part of the $T=0$ spectrum, however, exhibits already strong correlation effects due to the exchange coupling of the band states to the $4f$ moment system, predominantly in the $5d$ subband. They follow from magnon emission processes of the down-spin electrons and to a lesser extent from the formation of magnetic polarons.

Integration up to the Fermi edge yields the $T=0$ contribution of the conduction electrons to the magnetic moment. We find

$$\Delta\mu = 0.71\mu_{\mathrm{B}}. \tag{43}$$

Since in our model the $4f$ moments have a fixed value of $7\mu_{\mathrm{B}}$ the total moment amounts to $7.71\mu_{\mathrm{B}}$ very close to the experimental value of $7.63\mu_{\mathrm{B}}$.[2] Our value is a bit smaller than that from the LDA+U calculation in Ref. 11.

The procedure explained in the preceding sections allows for a determination of the full temperature dependence of the energy spectrum and the magnetic properties of Gd. The self-consistent evaluation yields a ferromagnetic low-temperature

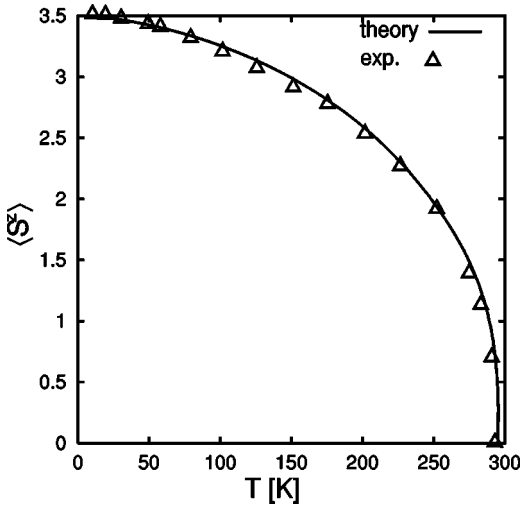

FIG. 4. Magnetization curve for ferromagnetic Gadolinium as function of the temperature. The triangles represent experimental data taken from Ref. 2.

phase with astonishing precise key data. Figure 4 exhibits the magnetization curve in comparison to the experimental data of Ref. 2. There is practically an exact agreement. In particular the calculated Curie temperature,

$$T_C = 294.1 \text{ K}, \tag{44}$$

being known as a very sensitive entity of magnetism, hardly deviates from the experimental value of 293.2 K. Note that there is in principle no fitting parameter in our theory; even the exchange constant J (28) is taken from the LDA input. We therefore have to conclude that the modified RKKY theory (Sec. IV B), with the effective exchange integrals being functionals of the conduction electron self-energy, describes the ferromagnetism of Gd in an absolutely convincing manner.

Since we did not consider a direct exchange interaction between the localized $4f$ moments the induced spin polarization of the conduction electrons mediates the indirect coupling. The *a priori* only slightly correlated $5d/6s/6p$ band states therefore exhibit a distinct temperature dependence as

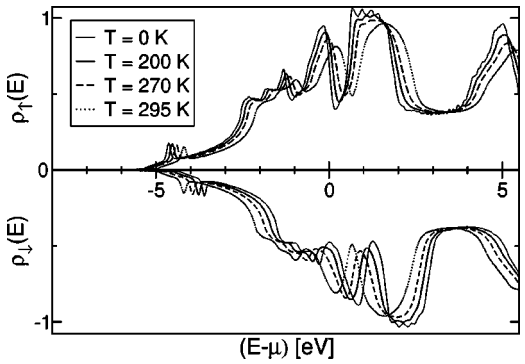

FIG. 5. Quasiparticle density of states of the valence and conduction bands of Gd as function of the energy $(E-\mu)$ (μ: chemical potential) for four different temperatures. The total densities of states consist of $5d$, $6s$, and $6p$ contributions.

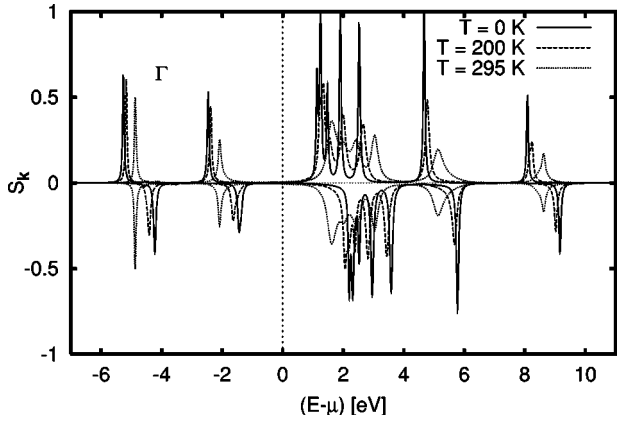

FIG. 6. Spin resolved single-electron spectral density of Gd at the Γ point as function of the energy $(E-\mu)$ for three different temperatures. Upper half: ↑ spectrum, lower half: ↓ spectrum.

can be seen for the total quasiparticle density of states in Fig. 5. The $T=0$ splitting is responsible for the band contribution (43) to the total magnetic moment. With increasing temperature the induced splitting reduces steadily collapsing at T_C.

The shift is not at all rigid ("Stoner-like"), but with clear deformations. The latter point to a substantial influence of nonlinear effects such as magnon emission and absorption and magnetic polaron formation, in particular what concerns the d states. The lower edge of the ↑ spectrum, predominantly built up by $6s$ states (Fig. 3), shows a red shift upon cooling below T_C as it is typical for local-moment systems, first observed for insulators and semiconductors such as EuO and EuS.[44] The temperature behavior at the chemical potential is not so clear.

The single-electron spectral density (11) represents the bare line shape of an angle- and spin-resolved photoemission experiment. Pronounced peaks in the spectral density define the quasiparticle band structure. For four high-symmetry points (Γ, A, H, M) we have calculated the energy dependence of the spectral density in the valence and conduction-band region. The results for three different temperatures ($T=0$, 200, 295 K) are represented in Figs. 6–9. The $T=0$-↑ spectra always consist of relatively sharp peaks pointing at quasiparticles with long, sometimes even infinite, lifetimes. In case of infinite lifetime (real self-energy) the spectral den-

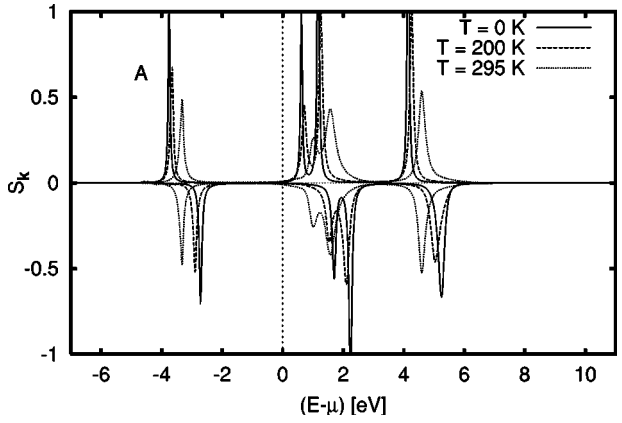

FIG. 7. The same as in Fig. 6 but for the A point.

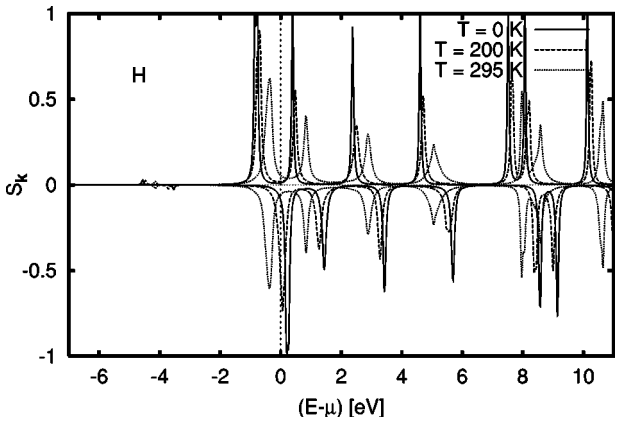

FIG. 8. The same as in Fig. 6 but for the H point.

sity is a δ function. For plotting reasons we have then added a small imaginary part ($i\Delta;\Delta=0.01$) to the electronic self-energy. For empty energy bands the $\uparrow$ spectrum would consist at $T=0$ exclusively of δ peaks. This is just the exact limiting case discussed in Sec. III B. It means nothing else than that a $\uparrow$ electron cannot undergo any scattering process if the localized $4f$ moments are ferromagnetically saturated. However, for finite and not fully spin-polarized band occupations some spin-exchange processes may happen giving rise to slight quasiparticle dampings. Figure 6 shows the energy dependence of the spectral density at the Γ point for three different temperatures. $T=0$ means ferromagnetic $4f$ saturation (Fig. 4) while $T=295$ K is slightly above the calculated Curie temperature (44). At $T=200$ K the moment system is partially ordered. The low energy peaks belong to $6s$ states (Figs. 1 and 3). They are spin split in the ferromagnetic phase, where the induced exchange splitting diminishes continuously with increasing temperature, collapsing at $T=T_C$ (Stoner-like behavior). This agrees with the photoemission data of Kim *et al.*[19] Similar temperature behavior is found for the other quasiparticle peaks too, and also for the other symmetry points A, H, and M (Figs. 7–9). These theoretical results partly contradict our previous investigation[26] according to which in some cases a persisting splitting in the paramagnetic phase should be possible. Experimental evidence for this behavior has been reported by Maiti *et al.*[21]

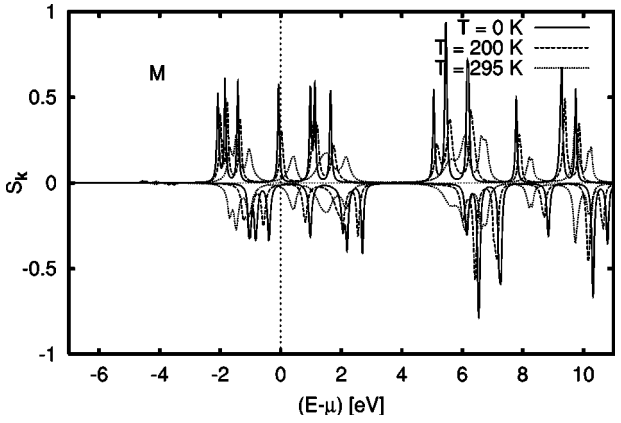

FIG. 9. The same as in Fig. 6 but for the *M* point.

The ambiguity comes along with the necessary decomposition of the total spectrum into nondegenerate subbands. That can be done, in principle, in different ways, and, at least in our opinion, it is not *a priori* clear which is the correct procedure. In this work we have used a method that retains the full atomic-orbital symmetry. The resulting rather broad subbands (Fig. 3) cause correspondingly small effective exchange couplings J/W. The self-energy $M_{\mathbf{k}\sigma}(E)$ (34) is then dominated by the first term and therewith relatively close to the mean-field solution of the sf (Kondo-lattice) model. The "Stoner collapsing" is a typical feature of the weak-coupling (mean-field) region. The band decomposition used in Ref. 26 leads to substantially smaller subband widths and therewith to stronger effective exchange couplings.

A general observation is that with increasing temperature the excitation peaks are getting broader, i.e., quasiparticle lifetimes decrease. Raising temperature means enhancing magnon densities and therewith a higher probability for electron-magnon spin-flip scattering. The *d* like states near and around the chemical potential μ exhibit stronger correlation effects than the low-energy $6s$ states or high-energy $6p$ states, again due to the larger effective coupling constant J/W. Contrary to the *H* and *M* points (Figs. 8 and 9) there is no spectral weight at the chemical potential for the Γ and *A* points (Figs. 6 and 7).

VI. SUMMARY

In this paper we have used a combination of a many-body approach to the Kondo-lattice (s-f) model with an LDA-DFT band structure calculation to get in a realistic and self-consistent manner the electronic and magnetic properties of the rare-earth metal gadolinium. The many-body approach has previously been developed and tested in several model studies. It consists of a moment-conserving decoupling approach for the single-electron Green function, which fulfills a maximum number of exact limiting cases, and a modified RKKY theory for the localized moment system. The effective exchange integrals between the localized spins turn out to be functionals of the electronic self-energy. In the weak coupling limit the approach agrees with the conventional RKKY theory.

As single-electron (Bloch) energies we have used the results of an ASW band-structure calculation therewith guaranteeing that all those interactions which are not explicitly covered by the Kondo-lattice model are taken into account in a rather realistic manner. An exact limiting case of the model could be exploited to avoid the well-known double counting problem. In a strict sense the method does not contain any really free parameter. The $4f$-$5d$ exchange coupling constant J, which enters the theory via the Kondo-lattice model, is fitted by the LDA input.

The results of our theoretical investigation agree astonishingly well with the experimental data of Gd. The self-consistent approach predicts correctly a ferromagnetic low-temperature phase. The magnetic $T=0$ moment is with $7.71\mu_B$ very close to the experimental value of $7.63\mu_B$. Even the extremely sensitive Curie temperature hardly deviates from the real Gd value (theory: 294.1 K, experiment:

293.2 K). The valence and conduction bands exhibit a remarkably induced temperature dependence. The $T=0$ exchange splitting explains the excess moment of $0.63\mu_B$ (or $0.71\mu_B$) that cannot be ascribed to the seven $4f$ electrons. The temperature dependence of the exchange splitting roughly scales with the macroscopic magnetization collapsing at T_C ("Stoner behavior") as it has been observed in photoemission experiments. Correlation effects lead to a distinct temperature dependence of the quasiparticle damping.

We believe that the proposed combination of a careful many-body treatment of a proper theoretical model with an *ab initio* band-structure calculation yields a rather realistic description of the ferromagnetic $4f$ metal Gadolinium.

ACKNOWLEDGMENTS

Financial support by the "Deutsche Forschungsgemeinschaft" within the SFB 290 ("Metallische dünne Filme: Struktur, Magnetismus und elektronischen Eigenschaften") and the SFB 484 ("Kooperative Phänomene im Festkörper: Metall-Isolator-Übergänge und Ordnung mikroskopischer Freiheitsgrade") are gratefully acknowledged.

*Electronic address: Wolfgang.Nolting@physik.hu-berlin.de

[1] S. Legvold, *Ferromagnetic Materials* (North Holland, Amsterdam, 1980), Vol. 1, Chap. 3.
[2] H. E. Nigh, S. Legvold, and F. H. Spedding, Phys. Rev. **132**, 1092 (1963).
[3] B. N. Harmon and A. J. Freeman, Phys. Rev. B **10**, 1979 (1974).
[4] J. Sticht and J. Kübler, Solid State Commun. **53**, 529 (1985).
[5] M. Richter and H. Eschrig, Solid State Commun. **72**, 263 (1989).
[6] B. C. H. Krutzen and F. Springelkamp, J. Phys.: Condens. Matter **1**, 8369 (1989).
[7] W. M. Temmermann and P. A. Sterne, J. Phys.: Condens. Matter **2**, 5529 (1990).
[8] D. J. Singh, Phys. Rev. B **44**, 7451 (1991).
[9] R. Ahuja, S. Anbluck, and B. Johansson, Phys. Rev. B **50**, 5147 (1994).
[10] R. F. Sabiryanov and S. S. Jaswal, Phys. Rev. B **55**, 4117 (1997).
[11] P. Kurz, G. Bihlmayer, and S. Blügel, J. Phys.: Condens. Matter **14**, 6353 (2002).
[12] *Magnetism and Electronic Correlations in Local-Moment Systems; Rare-Earth Elements and Compounds*, edited by M. Donath, P. A. Dowben, and W. Nolting (World Scientific, Singapore, 1998).
[13] J. K. Lang, Y. Baer, and P. A. Cox, J. Phys. F: Met. Phys. **11**, 121 (1981).
[14] O. Eriksson, R. Ahuja, A. Ormeci, J. Trygg, O. Hjortstam, P. Söderlind, B. Johansson, and J. M. Wills, Phys. Rev. B **52**, 4420 (1995).
[15] V. I. Anisimov, F. Aryasetiawan, and A. I. Lichtenstein, J. Phys.: Condens. Matter **9**, 767 (1996).
[16] A. B. Shick, A. I. Liechtenstein, and W. E. Pickett, Phys. Rev. B **60**, 10 763 (1999).
[17] A. R. Williams, J. Kübler, and C. D. Gelatt, Phys. Rev. B **19**, 6094 (1979).
[18] V. Eyert, Int. J. Quantum Chem. **77**, 1007 (2000).
[19] B. Kim, A. B. Andres, J. L. Erskine, K. J. Kim, and B. N. Harmon, Phys. Rev. Lett. **68**, 1931 (1992).
[20] D. Li, J. Zhang, P. A. Dowben, and M. Onellion, Phys. Rev. B **45**, 7272 (1992).
[21] K. Maiti, M. C. Malagoli, A. Dallmeyer, and C. Carbone, Phys. Rev. Lett. **88**, 167205 (2002).
[22] L. M. Sandratskii and J. Kübler, Europhys. Lett. **23**, 661 (1993).
[23] L. M. Sandratskii and P. Bruno, Phys. Rev. B **66**, 134435 (2002).
[24] I. Turek, J. Kudrnovsky, G. Bihlmayer, and S. Blügel, J. Phys.: Condens. Matter **15**, 2771 (2003).
[25] W. Nolting, W. Borgiel, V. Dose, and T. Fauster, Phys. Rev. B **40**, 5015 (1989).
[26] S. Rex, V. Eyert, and W. Nolting, J. Magn. Magn. Mater. **192**, 529 (1999).
[27] W. Nolting, S. Rex, and S. M. Jaya, J. Phys.: Condens. Matter **9**, 1301 (1997).
[28] C. Santos and W. Nolting, Phys. Rev. B **65**, 144419 (2002).
[29] S. Doniach, Physica B & C **B91**, 231 (1977).
[30] W. Nolting, Phys. Status Solidi B **96**, 11 (1979).
[31] S. G. Ovchinnikov, *Phase Transitions* (Gordon Breach, Bergs, UK, 1991), Vol. 36.
[32] D. Meyer, C. Santos, and W. Nolting, J. Phys.: Condens. Matter **13**, 2531 (2001).
[33] B. S. Shastry and D. C. Mattis, Phys. Rev. B **24**, 5340 (1981).
[34] S. R. Allan and D. M. Edwards, J. Phys. C **15**, 2151 (1982).
[35] W. Nolting, G. G. Reddy, A. Ramakanth, and D. Meyer, Phys. Rev. B **64**, 155109 (2001).
[36] J. F. Janak and A. R. Williams, Phys. Rev. B **14**, 4199 (1976).
[37] R. Schiller and W. Nolting, Solid State Commun. **110**, 121 (1999).
[38] W. Müller and W. Nolting, Phys. Rev. B **66**, 085205 (2002).
[39] W. Nolting, G. G. Reddy, A. Ramakanth, D. Meyer, and J. Kienert, Phys. Rev. B **67**, 024426 (2003).
[40] T. Kasuya, Prog. Theor. Phys. **16**, 45 (1956).
[41] K. Yosida, Phys. Rev. **106**, 893 (1957).
[42] N. N. Bogliubov and S. Tyablikow, Dokl. Akad. Nauk SSSR **126**, 53 (1959) [Sov. Phys. Dokl. **4**, 589 (1959)].
[43] W. Nolting, W. Müller, and C. Santos, J. Phys. A **36**, 9275 (2003).
[44] B. Batlogg, A. S. E. Kaldis, and P. Wachter, Phys. Rev. B **12**, 3940 (1975).