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Magnetism and electronic structure of a local moment ferromagnet

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Abstract. We propose a self-consistent approximate solution of the s–f model for describing the exchange coupling of a local moment system with a partially filled energy band. Induced electronic correlations account for the characteristic quasiparticle band effects which become manifest via striking temperature dependencies, band deformations and splittings. For weak s–f exchange interactions a ‘Stoner-like’ spin splitting of the conduction band proportional to the f magnetization occurs. As soon as the coupling exceeds a critical value an additional spin splitting of the quasiparticle dispersion sets in, which is due to different elementary excitations. One of these appears as a repeated emission and reabsorption of a magnon by the conduction electron, resulting in an effective electron–magnon attraction. This gives rise to a polaron-like quasiparticle (a ‘magnetic polaron’). Other elementary processes are connected to magnon emission or absorption by the conduction electron (‘scattering states’). The polarization of the conduction band due to the s–f exchange interaction J feeds back to the localized spin system leading to an indirect coupling between the spins. For weak s–f coupling the RKKY mechanism dominates ($T_c \propto J^2$), but with remarkable deviations for intermediate and strong couplings. The Curie temperature saturates with increasing J , where the saturation value is strongly dependent on the band occupation n . The oscillating behaviour of the effective exchange integral connecting the localized spins restricts ferromagnetism to special regions for n . The magnetization curve, the spin polarization of the itinerant electrons, and f–f as well as s–f spin correlation functions are worked out for a simple cubic lattice and discussed in terms of the band occupation n and the s–f exchange coupling J .

1. Introduction

Ferromagnetism (antiferromagnetism) is bound by two basic preconditions. The system has to be a solid, and this solid must contain permanent magnetic moments. Dividing the solids roughly into insulators and metals, and the moments into itinerant and localized ones, one gets four possible combinations, which provide a fairly reasonable classification of magnetism. The magnetic insulators with localized magnetic moments are excellently described by the Heisenberg model [1], as far as the purely magnetic properties are concerned. The prototype is the ferromagnetic 4f insulator EuO [2–6]. As insulators with itinerant moments one might consider the so-called ‘Mott–Hubbard insulators’ such as antiferromagnetic NiO [7], the permanent moments of which stem from a 1–3 eV broad, fully occupied quasiparticle subband [8]. If the electrical conductivity and the spontaneous magnetization of a magnetic metal are caused by the same group of electrons, then we speak of a band magnet. The permanent moments are then necessarily itinerant (Fe, Co, Ni). Band magnetism is far from being fully understood [9]. Limited insight can be derived

from ‘local spin-density approximations’ [10–12], local band theories [13] and multiband Hubbard models [14].

Magnetic metals, for which electrical conduction and magnetization are caused by different groups of electrons, are called ‘local moment magnets’. The prototype is the ferromagnetic rare-earth metal Gd [15] that takes its magnetism from a strongly localized half-filled 4f shell, while itinerant electrons in rather broad 6s, 5d bands determine the conductivity. Many striking properties of these materials are due to exchange correlations between the two well-defined electronic subsystems. As far as such ‘cross-effects’ are concerned the so-called s–f (or s–d) model [3] has turned out to be a reasonable theoretical framework. It traces back these ‘cross-effects’ to an intra-atomic exchange interaction between the conduction electron spin and the localized moment (the f spin). The mutual influence of the two electronic subsystems leads, on the one hand, to a remarkable spin, temperature and carrier concentration dependence of the quasiparticle band-structure of the itinerant electrons, and, on the other hand, to an indirect coupling of the localized f spins. This indirect coupling, called RKKY interaction [16–18], is mediated by an exchange-induced spin polarization of the ‘*a priori*’ non-magnetic conduction band. It is the main goal of our study to present a complete theory of the s–f model, which inspects in a self-consistent manner the mutual influence of the local moment magnetism and the quasiparticle nature of the conduction band. In this paper we restrict considerations exclusively to ferromagnetic systems. Antiferromagnetism will be discussed in a forthcoming paper.

The final goal of our investigation is that of establishing the magnetic and electronic properties of real substances such as the ferromagnetic 4f metals Gd, Tb, and Dy. It is a fact that the appearance of ferromagnetism is basically caused by strong electron correlations. A full understanding therefore requires a thorough investigation of the electronic structure of the underlying material. In previous papers we have done such a study for the classical band ferromagnets Fe [19], Co [20], and Ni [14]. The method is based on a combination of a many-body evaluation of an appropriate theoretical model with a single-electron band-structure calculation on the basis of density functional theory. It can be demonstrated how magnetic and electronic properties are intercorrelated. Rather realistic values for the Curie temperatures were derived. A similar procedure has been applied to Gd [15], where, however, the magnetic properties cannot yet be determined self-consistently. The magnetization curve of the localized spin system has been described by an $S = 7/2$ Brillouin function and only the exchange-induced reaction of the conduction band on the assumed magnetic state of the spin system has been inspected. It could be shown that the s–f exchange leads to a remarkable non-uniform magnetic behaviour of the energy spectrum at different positions in the Brillouin zone, and in particular to different temperature dependencies. Special splittings of the single-electron spectral density, caused by the s(d)–f exchange interaction, lead one to expect complicated photoemission spectra. However, nothing could be said about the origin of the Gd ferromagnetism. We present in this paper a many-body evaluation of the s–f model that allows a self-consistent inspection of the interplay between magnetism and electronic structure. Effective exchange integrals for the spin–spin interaction are expressed in terms of the electronic self-energy which in turn depends decisively on correlation functions of the spin system. This gives rise to a sensitive influence of the conduction electron density on the Curie temperature and on other magnetic properties. Exchange-induced spin polarizations of the band electrons feed back to the localized spins accounting for an indirect coupling between them. In lowest order the well-known RKKY result is reproduced. Because of the pronounced many-particle aspects covered by the ‘s–f problem’ it is necessary to start with an exhaustive model study. Only then will we be able to interpret unambiguously the results for real substances such as Gd. Thus we present in

this paper a detailed inspection of the s–f model, which for the band part means an extension and improvement of our earlier theory [15], while the f-moment part is now self-consistently included.

The paper is organized as follows. In section 2 the model Hamiltonian is introduced and the corresponding many-body problem is formulated. The s–f model is thought appropriate for an exchange-coupled system of itinerant conduction electrons and localized magnetic moments. For this reason intercorrelated electronic and magnonic self-energies mould the physical properties. In section 3 we first develop an approximate solution for the electronic self-energy part, and that in terms of certain f spin correlation functions. The approach treats the site-dependent single-electron Green function and applies to all known exactly solvable limiting cases. The subsequent treatment of the magnon part consists of two steps. First we transform the s–f interaction operator into an ‘effective’ Heisenberg exchange operator. The ‘effective’ exchange integrals turn out to be functionals of the electronic self-energy part (section 4). Magnon energies and spin correlation functions are then derived from a properly defined f spin Green function. Eventually a rather complicated, but closed system of equations is formed that can be solved self-consistently for all interesting physical properties. The results of the numerical evaluation are presented and discussed in section 5. Comprehensive conclusions and an outlook with respect to applications to real substances are given in section 6.

2. The model Hamiltonian and its many-body problem

The principal goal of our study is a qualitative understanding of temperature and correlation effects in the electronic structure of metallic ‘local moment’ ferromagnets such as the 4f elements Gd (the prototype!), Tb, and Dy. Since a direct interaction of the localized moments must be negligible in these materials, the collective magnetic order below a critical temperature is explicable only by an indirect interaction via the ‘*a priori*’ non-magnetic itinerant conduction electrons. This physical background is well accounted for by the s–f model [1, 3], the Hamiltonian of which

$$H = H_s + H_{sf} \quad (2.1)$$

describes an exchange-coupled system of itinerant conduction electrons and localized magnetic f moments. The conduction electrons are treated as s electrons without explicit Coulomb interaction:

$$H_s = \sum_{ij\sigma} T_{ij} c_{i\sigma}^+ c_{j\sigma} = \sum_{k\sigma} \varepsilon(\mathbf{k}) c_{k\sigma}^+ c_{k\sigma}. \quad (2.2)$$

The contributions of the Coulomb interaction between s electrons are considered to be not decisive for the phenomena that we are interested in. They might be incorporated by a proper renormalization of the Bloch energies $\varepsilon(\mathbf{k})$, e.g. by using the results of a corresponding LDA calculation [15]. This will indeed be done in forthcoming papers which will extend our present model study to real substances like Gd.

The hopping integrals T_{ij} and Bloch energies $\varepsilon(\mathbf{k})$ are connected by Fourier transformation:

$$T_{ij} = \frac{1}{N} \sum_{\mathbf{k}} \varepsilon(\mathbf{k}) e^{i\mathbf{k} \cdot (\mathbf{R}_i - \mathbf{R}_j)}. \quad (2.3)$$

c^+ (c) represents the creation (annihilation) operator of an electron specified by the lower indices: i, j stand for the lattices sites $\mathbf{R}_i, \mathbf{R}_j$. $\sigma = \uparrow, \downarrow$ is the electron spin projection, and \mathbf{k} is a wave vector from the first Brillouin zone.

Each lattice site \mathbf{R}_i possesses a localized magnetic f moment represented by a spin operator \mathbf{S}_i . As already mentioned, a direct interaction is not considered but a coupling of the spin system to the band electrons by an intra-atomic s-f exchange is considered:

$$H_{sf} = -J \sum_i \boldsymbol{\sigma}_i \cdot \mathbf{S}_i \quad (2.4)$$

$\boldsymbol{\sigma}_i$ is the Pauli spin operator. For the following application the second-quantization representation appears to be convenient:

$$H_{sf} = -\frac{1}{2} J \sum_{i,\sigma} (z_\sigma S_i^z n_{i\sigma} + S_i^\sigma c_{i-\sigma}^+ c_{i\sigma}) \quad (2.5)$$

$$S_j^\sigma = S_j^x + i z_\sigma S_j^y \quad z_\uparrow = +1 \quad z_\downarrow = -1. \quad (2.6)$$

$n_{i\sigma} = c_{i\sigma}^+ c_{i\sigma}$ is the occupation number operator. With respect to applications to materials like Gd we shall assume a *ferromagnetic* exchange coupling, i.e. a positive s-f coupling constant J . In the case where $J < 0$ the model Hamiltonian (2.1) is that of the so-called ‘Kondo lattice’ [21, 22].

In spite of its simple structure the operator (2.1) creates a rather sophisticated many-body problem which cannot be solved exactly in the general case. There exist, however, some non-trivial, rigorously tractable limiting cases [23–25] which can be used as tests for the otherwise unavoidable approximations.

The goal is a self-consistent description of how the not directly interacting f spins may be forced into a collective magnetic order by a proper spin polarization in the conduction band mediated by H_{sf} , and how the electronic quasiparticle band-structure reacts to the magnetic state of the localized spin system. All of the information that we are interested in can be read off from the two retarded Green functions

$$G_{ij\sigma}(E) = \langle\langle c_{i\sigma}; c_{j\sigma}^+ \rangle\rangle_E = -i \int_0^\infty dt \exp\left(-\frac{i}{\hbar} Et\right) \langle\langle [c_{i\sigma}(t), c_{j\sigma}^+(0)]_+ \rangle\rangle \quad (2.7)$$

$$P_{ij}^{(a)}(E) = \langle\langle S_i^+; e^{aS_j^z} S_j^- \rangle\rangle_E = -i \int_0^\infty dt \exp\left(-\frac{i}{\hbar} Et\right) \langle\langle [S_i^+(t), (e^{aS_j^z} S_j^-)(0)]_- \rangle\rangle. \quad (2.8)$$

$\langle\langle \dots \rangle\rangle$ means thermodynamic averaging, while $[\dots]_+$ ($[\dots]_-$) denotes the anti-commutator (commutator). $G_{ij\sigma}(E)$ is known as the ‘single-electron Green function’, while we shall refer to $P_{ij}^{(a)}(E)$ as the ‘magnon Green function’. a is a real parameter chosen for convenience in order to derive from the magnon function all interesting spin correlation functions, and that for arbitrary (!) spin values S . This form of $P_{ij}^{(a)}(E)$ has been introduced by Callen [26] to investigate the Heisenberg model. Its usefulness will become evident in section 4. Besides the site-dependent functions $G_{ij\sigma}(E)$ and $P_{ij}^{(a)}(E)$, the wave-vector-dependent Fourier transforms are convenient (translational symmetry!), e.g.

$$G_{\mathbf{k}\sigma}(E) = \frac{1}{N} \sum_{i,j} G_{ij\sigma}(E) e^{i\mathbf{k}\cdot(\mathbf{R}_i - \mathbf{R}_j)} = \langle\langle c_{\mathbf{k}\sigma}; c_{\mathbf{k}\sigma}^+ \rangle\rangle_E. \quad (2.9)$$

$c_{\mathbf{k}\sigma}$ is the Fourier transform of $c_{i\sigma}$. The equation of motion of the single-electron Green function is formally solved by introducing the self-energy $M_{\mathbf{k}\sigma}(E)$:

$$\langle\langle [c_{\mathbf{k}\sigma}, H_{sf}]_-; c_{\mathbf{k}\sigma}^+ \rangle\rangle_E = M_{\mathbf{k}\sigma}(E) G_{\mathbf{k}\sigma}(E). \quad (2.10)$$

The self-energy gathers together all of the influences of the s-f exchange interaction on the electronic structure of the conduction band, being therefore a central quantity of our study:

$$G_{\mathbf{k}\sigma}(E) = \frac{\hbar}{E - \varepsilon(\mathbf{k}) - M_{\mathbf{k}\sigma}(E)}. \quad (2.11)$$

A function of exactly the same usefulness as the Green function $G_{k\sigma}(E)$ is the single-electron spectral density, which is, except for as regards the corresponding dipole transition matrix elements, directly related to angle- and spin-resolved (inverse) photoemission experiments:

$$S_{k\sigma}(E) = -\frac{1}{\pi} \text{Im} G_{k\sigma}(E) = -\frac{\hbar}{\pi} \frac{\text{Im} M_{k\sigma}(E)}{(E - \varepsilon(\mathbf{k}) - \text{Re} M_{k\sigma}(E))^2 + (\text{Im} M_{k\sigma}(E))^2}. \quad (2.12)$$

An additional wave-vector summation yields the quasiparticle density of states (QDOS) in terms of which we shall discuss a great part of our results:

$$\rho_\sigma(E) = \frac{1}{N\hbar} \sum_{\mathbf{k}} S_{k\sigma}(E). \quad (2.13)$$

By use of the so-called spectral theorem [27] the spectral density helps, e.g., to determine the (\mathbf{k}, σ) -dependent average occupation number:

$$\langle n_{k\sigma} \rangle = \langle c_{k\sigma}^+ c_{k\sigma} \rangle = \int_{-\infty}^{+\infty} dE f_-(E) S_{k\sigma}(E) \quad (2.14)$$

where $f_-(E)$ is the Fermi function. The local occupation number is related to the QDOS:

$$\langle n_{i\sigma} \rangle = \frac{1}{\hbar} \int_{-\infty}^{+\infty} dE f_-(E) S_{ii\sigma}(E) = \int_{-\infty}^{+\infty} dE f_-(E) \rho_\sigma(E). \quad (2.15)$$

In a translationally symmetric system this quantity will be site independent: $\langle n_{i\sigma} \rangle \equiv \langle n_\sigma \rangle$.

In the next section we derive an explicit expression for the electronic self-energy $M_{k\sigma}(E)$. The final result will contain a rather large number of equal-time correlation functions of purely s character such as (2.14) and (2.15), of purely f character like

$$\langle S_i^z \rangle \quad \langle (S_i^z)^2 \rangle \quad \langle (S_i^z)^3 \rangle \quad \langle S_i^\sigma S_i^{-\sigma} \rangle \quad (2.16)$$

and of ‘mixed’ s–f character:

$$\Delta_\sigma = \langle S_i^z n_{i\sigma} \rangle \quad \gamma_\sigma = \langle S_i^{-\sigma} c_{i\sigma}^+ c_{i-\sigma} \rangle \quad (2.17)$$

$$\mu_\sigma = \langle S_i^{-\sigma} S_i^\sigma n_{i\sigma} \rangle \quad \eta_\sigma = \langle S_i^{-\sigma} S_i^z c_{i\sigma}^+ c_{i-\sigma} \rangle \quad \vartheta_\sigma = \langle S_i^z n_{i\sigma} n_{i-\sigma} \rangle \equiv \vartheta. \quad (2.18)$$

All of these terms must be determined self-consistently within our (approximate) theory. We shall demonstrate in the next section how to express the ‘mixed’ correlations (2.17) and (2.18) in terms of the electronic self-energy. For the pure f spin correlations (2.16) we exploit the magnon Green function (2.8). The f spin operators commute with H_s , so $P_{ij}^{(a)}(E)$ is mainly determined by the s–f exchange. We propose in section 4 a transformation of H_{sf} into an indirect Heisenberg exchange operator. The corresponding exchange integrals \hat{J}_{ij} reduce in first order to the well-known RKKY result [16–18] being otherwise functions of the electronic self-energy $M_{k\sigma}(E)$. Thereby the polarization of the conduction electrons, provoked by the localized f spins, feeds back to the spin system. The function $P_{ij}^{(a)}(E)$ is calculated by use of the effective Heisenberg model Hamiltonian (4.14).

In the next section we propose a theory for the fundamental electronic self-energy $M_{k\sigma}(E)$.

3. The electronic self-energy

Our procedure for obtaining the conduction electron self-energy $M_{k\sigma}(E)$ starts from the equation of motion of the single-electron Green function (2.7):

$$\sum_m (E\delta_{im} - T_{im}) G_{mj\sigma}(E) = \hbar\delta_{ij} - \frac{1}{2} J(z_\sigma \Gamma_{ii,j\sigma}(E) + F_{ii,j\sigma}(E)) \quad (3.1)$$

that contains two ‘higher’ functions which we call the ‘spin-flip’ function:

$$F_{ip,j\sigma}(E) = \langle\langle S_i^{-\sigma} c_{p-\sigma}; c_{j\sigma}^+ \rangle\rangle_E \quad (3.2)$$

and the ‘Ising’ function, respectively:

$$\Gamma_{ip,j\sigma}(E) = \langle\langle S_i^z c_{p\sigma}; c_{j\sigma}^+ \rangle\rangle_E. \quad (3.3)$$

To achieve a solution for the single-electron Green function we construct in the next step the equations of motion of the ‘new’ functions (3.2) and (3.3). They are formally given by the following expressions:

$$\sum_r (E\delta_{pr} - T_{pr}) F_{ir,j\sigma}(E) = \langle\langle S_i^{-\sigma} [c_{p-\sigma}, H_{sf}]_-; c_{j\sigma}^+ \rangle\rangle_E + \langle\langle [S_i^{-\sigma}, H_{sf}]_- c_{p-\sigma}; c_{j\sigma}^+ \rangle\rangle_E \quad (3.4)$$

$$\begin{aligned} \sum_r (E\delta_{pr} - T_{pr}) \Gamma_{ir,j\sigma}(E) &= \hbar \langle S_i^z \rangle \delta_{pj} + \langle\langle S_i^z [c_{p\sigma}, H_{sf}]_-; c_{j\sigma}^+ \rangle\rangle_E \\ &+ \langle\langle [S_i^z, H_{sf}]_- c_{p\sigma}; c_{j\sigma}^+ \rangle\rangle_E. \end{aligned} \quad (3.5)$$

On the right-hand sides of these equations there appear further ‘higher’ Green functions, which prevent a direct solution of the problem. Our treatment of these functions consists of two steps. One concerns the case $i \neq p$, and the other the diagonal terms $i = p$. For $i \neq p$ we use a self-consistent ‘self-energy approach’ which leads to an effective decoupling of the hierarchy of the equations of motion. This approach turns out to be less convincing for the diagonal terms, $i = p$, therefore being replaced by a moment technique which takes the local correlations into account better. The latter appears to be correct for all known, rigorously calculable limiting cases. We first introduce the ‘self-energy approach’.

(a) $i \neq p$. The starting point is the definition equation of the electronic self-energy, the site-dependent analogue to (2.10):

$$\langle\langle [c_{p\sigma}, H_{sf}]_-; c_{j\sigma}^+ \rangle\rangle_E = \sum_r M_{pr\sigma}(E) \langle\langle c_{r\sigma}; c_{j\sigma}^+ \rangle\rangle_E. \quad (3.6)$$

This relation formally corresponds to the replacement

$$[c_{p\sigma}, H_{sf}]_- \Rightarrow \sum_r M_{pr\sigma}(E) c_{r\sigma} \quad (3.7)$$

within the brackets of the Green function. The spectral representations [27] of the two functions in equation (3.6) reveal that both are meromorphic functions, with exactly the same single-particle poles. They differ only in the spectral weights of these poles, where the equality of both sides in (3.6) is ensured by the self-energy components $M_{pr\sigma}(E)$. When we now inspect in the same sense the spectral decomposition of the following two functions:

$$\langle\langle S_i^z [c_{p\sigma}, H_{sf}]_-; c_{j\sigma}^+ \rangle\rangle_E \quad \langle\langle S_i^z c_{r\sigma}; c_{j\sigma}^+ \rangle\rangle_E$$

which are identical to those in equation (3.6) except for in that they contain the additional spin operator S_i^z , we come to the same conclusion—that they can differ only in their spectral weights, but must have exactly the same single-particle pole structure. By use of (3.7) and in analogy to (3.6) we therefore propose the following plausible *ansatz*:

$$\langle\langle S_i^z [c_{p\sigma}, H_{sf}]_-; c_{j\sigma}^+ \rangle\rangle_E \approx \sum_r M_{pr\sigma}(E) \langle\langle S_i^z c_{r\sigma}; c_{j\sigma}^+ \rangle\rangle_E \quad (i \neq p). \quad (3.8)$$

The same justification can be applied to the first term on the right-hand side of equation (3.4):

$$\langle\langle S_i^{-\sigma} [c_{p-\sigma}, H_{sf}]_-; c_{j\sigma}^+ \rangle\rangle_E \approx \sum_r M_{pr-\sigma}(E) \langle\langle S_i^{-\sigma} c_{r-\sigma}; c_{j\sigma}^+ \rangle\rangle_E \quad (i \neq p). \quad (3.9)$$

The two other ‘higher’ Green functions in the equations of motion (3.4) and (3.5) contain commutators of spin operators with the interaction part H_{sf} of the model Hamiltonian. In the next section H_{sf} is transformed, as mentioned above, into an effective exchange operator H_f for the localized spin system (4.14). If we already use at this stage the effective exchange operator, then we have now to apply, in strict analogy to (3.7),

$$[S_i^\sigma, H_f]_- = 2z_\sigma \sum_m \hat{J}_{im} (S_m^z S_i^\sigma - S_i^z S_m^\sigma) \quad (3.10)$$

$$[S_i^z, H_f]_- = \sum_m \hat{J}_{im} (S_i^+ S_m^- - S_i^- S_m^+). \quad (3.11)$$

The effective exchange integrals \hat{J}_{im} are explicitly derived in the next section (see equation (4.15)). For the final evaluation of the complete theory they have to be determined self-consistently because of being complicated functionals of the electronic self-energy. Performing the same approximations as in section 4 for the magnon system, we get for the spin-flip function in (3.4) by use of (3.11)

$$\langle\langle [S_i^{-\sigma}, H_{sf}]_- c_{p-\sigma}; c_{j\sigma}^+ \rangle\rangle_E \approx 2z_\sigma \langle S^z \rangle \sum_m \hat{J}_{im} (F_{mp,j\sigma}(E) - F_{ip,j\sigma}(E)) \quad (i \neq p) \quad (3.12)$$

while the corresponding Green function in the equation of motion (3.5) of the ‘Ising function’ $\Gamma_{ip,j\sigma}(E)$ is approximately zero:

$$\langle\langle [S_i^z, H_{sf}]_- c_{p\sigma}; c_{j\sigma}^+ \rangle\rangle \approx \sum_m \hat{J}_{im} (\langle S_i^+ S_m^- \rangle - \langle S_i^- S_m^+ \rangle) G_{pj\sigma}(E) = 0 \quad (i \neq p). \quad (3.13)$$

Using (3.8), (3.9), (3.12), and (3.13), the ‘higher’ Green functions in the equations of motion (3.4) and (3.5) are expressed for $i \neq p$ as linear combinations of the simpler functions (2.7), (3.2), and (3.3), where the coefficients are directly or indirectly (\hat{J}_{im}) determined by the electronic self-energy.

(b) $i = p$. For the diagonal terms on the right-hand side of (3.4) and (3.5), we cannot use the ‘self-energy approaches’ because of the strong intra-atomic correlations between the localized f spins and the itinerant conduction electron spin. We first inspect the ‘higher spin-flip functions’ of equation (3.4) starting with an explicit evaluation of the commutators:

$$\langle\langle [S_i^{-\sigma} c_{i-\sigma}, H_{sf}]_-; c_{j\sigma}^+ \rangle\rangle_E = \frac{1}{2} J (z_\sigma F_{ii,j\sigma}^{(1)}(E) - F_{ii,j\sigma}^{(2)}(E)) \quad (3.14)$$

$$\langle\langle [S_i^{-\sigma}, H_{sf}]_- c_{i-\sigma}; c_{j\sigma}^+ \rangle\rangle_E = \frac{1}{2} J (-F_{ii,j\sigma}^{(3)}(E) + 2z_\sigma F_{ii,j\sigma}^{(4)}(E)). \quad (3.15)$$

Here we have used the abbreviations

$$F_{ii,j\sigma}^{(1)}(E) = \langle\langle S_i^{-\sigma} S_i^z c_{i-\sigma}; c_{j\sigma}^+ \rangle\rangle_E \quad (3.16)$$

$$F_{ii,j\sigma}^{(2)}(E) = \langle\langle S_i^{-\sigma} S_i^\sigma c_{i\sigma}; c_{j\sigma}^+ \rangle\rangle_E \quad (3.17)$$

$$F_{ii,j\sigma}^{(3)}(E) = \langle\langle S_i^{-\sigma} n_{i\sigma} c_{i-\sigma}; c_{j\sigma}^+ \rangle\rangle_E \quad (3.18)$$

$$F_{ii,j\sigma}^{(4)}(E) = \langle\langle S_i^z c_{i-\sigma}^+ c_{i\sigma} c_{i-\sigma}; c_{j\sigma}^+ \rangle\rangle_E. \quad (3.19)$$

The ‘higher Ising functions’ on the right-hand side of (3.5) do not require the introduction of further Green functions since they can be exactly expressed in terms of the functions (2.7), (3.2), (3.3), (3.16), (3.17), and (3.18) already defined (see appendix A).

To get a closed system of equations we are eventually left with the determination of the functions $F_{ii,j\sigma}^{(n)}(E)$, $n = 1, \dots, 4$ which have some exactly calculable limiting cases. For

$S = 1/2$ and arbitrary temperatures, the functions $F_{ii,j\sigma}^{(1)}(E)$, $F_{ii,j\sigma}^{(2)}(E)$ obey the following exact relations:

$$F_{ii,j\sigma}^{(1)}(E) = \frac{1}{2} z_\sigma F_{ii,j\sigma}(E) \quad (3.20)$$

$$F_{ii,j\sigma}^{(2)}(E) = \frac{1}{2} G_{ij\sigma}(E) - z_\sigma \Gamma_{ii,j\sigma}(E). \quad (3.21)$$

The same two functions for arbitrary spin S , but for ferromagnetic saturation ($\langle S^z \rangle = S$), are given by

$$F_{ii,j\sigma}^{(1)}(E) \rightarrow \left(\left(S - \frac{1}{2} \right) + \frac{1}{2} z_\sigma \right) F_{ii,j\sigma}(E) \quad (3.22)$$

$$F_{ii,j\sigma}^{(2)}(E) \rightarrow S G_{ij\sigma}(E) - z_\sigma \Gamma_{ii,j\sigma}(E). \quad (3.23)$$

These exact limiting cases, (3.20) to (3.23), suggest the following structures for the general case:

$$F_{ii,j\sigma}^{(1)}(E) = \alpha_{1\sigma} G_{ij\sigma}(E) + \beta_{1\sigma} F_{ii,j\sigma}(E) \quad (3.24)$$

$$F_{ii,j\sigma}^{(2)}(E) = \alpha_{2\sigma} G_{ij\sigma}(E) + \beta_{2\sigma} \Gamma_{ii,j\sigma}(E). \quad (3.25)$$

The subsequent evaluation will yield $\alpha_{1\sigma} = 0$ in accordance with (3.20) and (3.22). Similar considerations for the exactly solvable limiting cases of empty ($n = 0$) and full ($n = 2$) conduction bands yield for the two other functions

$$F_{ii,j\sigma}^{(3)}(E) = \alpha_{3\sigma} G_{ij\sigma}(E) + \beta_{3\sigma} F_{ii,j\sigma}(E) \quad (3.26)$$

$$F_{ii,j\sigma}^{(4)}(E) = \alpha_{4\sigma} G_{ij\sigma}(E) + \beta_{4\sigma} \Gamma_{ii,j\sigma}(E). \quad (3.27)$$

For the complete solution we still need the coefficients $\alpha_{n\sigma}$ and $\beta_{n\sigma}$. The set of equations (3.24) to (3.27) contains seven different Green functions of the type $\langle\langle A; B \rangle\rangle_E$. For each of them we can calculate exactly the first two spectral moments according to the relation

$$M_{AB}^{(n)} \equiv \left\langle \left(i\hbar \frac{\partial}{\partial t} \right)^n [A(t), B(t')]_+ \right\rangle_{t=t'} \quad n = 0, 1, 2, \dots \quad (3.28)$$

Because of the equivalent equation

$$M_{AB}^{(n)} = -\frac{1}{\pi} \int_{-\infty}^{+\infty} dE E^n \text{Im} \langle\langle A; B \rangle\rangle_E \quad (3.29)$$

the moments can be used in the next step to fix the unknown coefficients $\alpha_{n\sigma}$, $\beta_{n\sigma}$ in the equations (3.24) to (3.27), and that in terms of certain expectation values. The spectral moments needed as well as the resulting coefficients are listed in appendix B. They are dominated by the expectation values of the f spin system listed in (2.16), and by the ‘mixed’ s–f correlation functions in (2.17) and (2.18). The latter appear essential for a precise interpretation of the rather sophisticated properties of the s–f exchange model. Fortunately, they can all be derived from the above-introduced Green functions.

By use of the spectral theorem for Green functions, the correlation functions in (2.17), Δ_σ and γ_σ , follow directly from the ‘Ising’ function $\Gamma_{ii,j\sigma}(E)$, and from the ‘spin-flip’ function $F_{ii,j\sigma}(E)$ respectively, while μ_σ , η_σ , and ϑ_σ , defined in (2.18), are predetermined by (3.25), (3.24), and (3.27):

$$\mu_\sigma = \alpha_{2\sigma} \langle n_\sigma \rangle + \beta_{2\sigma} \Delta_\sigma \quad (3.30)$$

$$\eta_\sigma = \alpha_{1\sigma} \langle n_\sigma \rangle + \beta_{1\sigma} \gamma_{-\sigma} \quad (3.31)$$

$$\vartheta_\sigma = -\alpha_{4\sigma} \langle n_\sigma \rangle - \beta_{4\sigma} \Delta_\sigma \equiv \vartheta. \quad (3.32)$$

We will demonstrate in section 4 how to get the pure f spin correlations (2.16). Since all expectation values in the coefficients $\alpha_{n\sigma}$, $\beta_{n\sigma}$ can be expressed in terms of one or more of the Green functions involved (see appendix B), we finally come to a closed system of equations which can be solved self-consistently for all quantities of interest. Some further details are given in appendix C.

The resulting implicit equation for determination of the electronic self-energy reads as follows:

$$M_{q\sigma}(E) = -\frac{1}{2}Jz_\sigma\langle S^z \rangle + \frac{1}{4}J^2\frac{D_{q\sigma}(E)}{N_{q\sigma}(E)}. \quad (3.33)$$

The first term represents the well-known mean-field contribution of the s-f exchange, which is linear in the coupling J , while the second term involves the higher-order consequences of the ‘Ising’ and the ‘spin-flip’ parts of the exchange interaction H_{sf} . The $D_{q\sigma}(E)$ as well as the $N_{q\sigma}(E)$ are complicated functionals of the self-energy for both spin directions:

$$\begin{aligned} D_{q\sigma}(E) &= (A_\sigma + B_\sigma\langle S^z \rangle) \left(1 - T_{0\sigma} + \frac{1}{2}Jz_\sigma c_\sigma \frac{1}{\hbar} G_{0\sigma}(E) \right) P_{q\sigma}(E) \\ &+ \left(\frac{1}{2}Ja_\sigma + z_\sigma\langle S^z \rangle \right) \left(\frac{1}{2}Jb_\sigma - M_{q\sigma}(E) \right) \frac{1}{\hbar} G_{0\sigma}(E) \\ &\times \left(1 - Q_{q\sigma}(E) + \frac{1}{2}Jz_\sigma B_\sigma P_{q\sigma}(E) \right) \end{aligned} \quad (3.34)$$

$$N_{q\sigma}(E) = c_\sigma B_\sigma \frac{1}{\hbar} G_{0\sigma}(E) P_{q\sigma}(E) - (1 - T_{0\sigma}(E))(1 - Q_{q\sigma}(E)). \quad (3.35)$$

Here we have used the following abbreviations for the propagators:

$$P_{q\sigma}(E) = \frac{1}{N} \sum_{\mathbf{p}} \frac{1}{E + z_\sigma \hat{E}(\mathbf{q} - \mathbf{p}) - \varepsilon(\mathbf{p}) - M_{\mathbf{p}-\sigma}(E)} \quad (3.36)$$

$$G_{0\sigma}(E) = \frac{1}{N} \sum_{\mathbf{p}} \frac{\hbar}{E - \varepsilon(\mathbf{p}) - M_{\mathbf{p}\sigma}(E)} = \frac{1}{N} \sum_{\mathbf{p}} G_{\mathbf{p}\sigma}(E) \quad (3.37)$$

$$Q_{q\sigma}(E) = \frac{1}{N} \sum_{\mathbf{p}} \frac{z_\sigma \hat{E}(\mathbf{q} - \mathbf{p}) - M_{\mathbf{p}-\sigma}(E) + (1/2)JC_\sigma}{E + z_\sigma \hat{E}(\mathbf{q} - \mathbf{p}) - \varepsilon(\mathbf{p}) - M_{\mathbf{p}-\sigma}(E)} \quad (3.38)$$

$$T_{0\sigma}(E) = \frac{1}{N} \sum_{\mathbf{p}} \frac{(1/2)Jb_\sigma - M_{\mathbf{p}\sigma}(E)}{E - \varepsilon(\mathbf{p}) - M_{\mathbf{p}\sigma}(E)}. \quad (3.39)$$

The $\hat{E}(\mathbf{p})$ are dressed magnon energies to be derived in equation (4.21). We recognize that (3.33) is not an analytic solution for $M_{q\sigma}(E)$ at all, but must be solved self-consistently. Via the coefficients a_σ , b_σ , c_σ and A_σ , B_σ , C_σ , which are explained in appendix C, a lot of expectation values are involved, e.g., the spin-dependent average occupation number $\langle n_\sigma \rangle$. According to (2.15) this quantity is directly related to the single-electron Green function. Of great importance are the ‘mixed’ correlation functions in equation (2.17) which can rigorously be expressed in terms of the electronic self-energy $M_{q\sigma}(E)$, simply by applying the spectral theorem to certain ‘higher’ Green functions involved in our theory. The result (3.33) for the electronic self-energy contains furthermore the pure f spin correlations, listed in (2.16). They cannot be expressed in terms of the Green functions used in this section. They have to be derived from the magnon Green function $P_{ij}^{(a)}(E)$, defined in (2.8). To evaluate this function we are going to transform in the next section the s-f interaction H_{sf} (2.4) into an effective Heisenberg exchange operator H_f .

4. The modified RKKY interaction

Our model does not consider a direct exchange interaction between the localized f spins. We discuss in this section the indirect f spin coupling mediated by the s–f exchange (2.4), which can be written in the following equivalent form:

$$H_{sf} = -J \frac{1}{N} \sum_{i,\sigma,\sigma'} \sum_{\mathbf{k},\mathbf{q}} e^{-i\mathbf{q}\cdot\mathbf{R}_i} (\mathbf{S}_i \cdot \hat{\boldsymbol{\sigma}})_{\sigma\sigma'} c_{\mathbf{k}+\mathbf{q}\sigma}^+ c_{\mathbf{k}\sigma'}. \quad (4.1)$$

The components of the band electron spin operator $\hat{\boldsymbol{\sigma}}$ are the Pauli spin matrices. We transform H_{sf} into an ‘effective’ spin Hamiltonian H_f by averaging H_{sf} in the subspace of the conduction electrons ($\langle \cdot \rangle^{(s)}$):

$$H_f = -J \frac{1}{N} \sum_{i,\sigma,\sigma'} \sum_{\mathbf{k},\mathbf{q}} e^{-i\mathbf{q}\cdot\mathbf{R}_i} (\mathbf{S}_i \cdot \hat{\boldsymbol{\sigma}})_{\sigma\sigma'} \langle c_{\mathbf{k}+\mathbf{q}\sigma}^+ c_{\mathbf{k}\sigma'} \rangle^{(s)}. \quad (4.2)$$

In the above-mentioned subspace the f spin operators do not act. On the other hand, the expectation value ($\langle \cdot \rangle^{(s)}$) may still have operator properties in the f spin subspace. For the same reason $\langle c_{\mathbf{k}+\mathbf{q}\sigma}^+ c_{\mathbf{k}\sigma'} \rangle^{(s)}$ does not necessarily vanish for $\mathbf{q} \neq \mathbf{0}$ and/or $\sigma \neq \sigma'$. We try to evaluate the expectation value on the right-hand side of (4.2) by solving the equation of motion of the Green function

$$\hat{G}_{\mathbf{k},\mathbf{k}+\mathbf{q}}^{\sigma\sigma'}(E) = \langle \langle c_{\mathbf{k}\sigma}; c_{\mathbf{k}+\mathbf{q}\sigma'}^+ \rangle \rangle_E^{(s)}. \quad (4.3)$$

This function is formally defined as in (2.7) and (2.9), only the averaging has to be done in the s subspace only. The equation of motion reads

$$(E - \varepsilon(\mathbf{k})) \hat{G}_{\mathbf{k},\mathbf{k}+\mathbf{q}}^{\sigma\sigma'} = \hbar \delta_{\mathbf{q},0} \delta_{\sigma\sigma'} - J \frac{1}{N} \sum_{i\mathbf{k}'\sigma''} e^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}_i} (\mathbf{S}_i \cdot \hat{\boldsymbol{\sigma}})_{\sigma'\sigma''} \hat{G}_{\mathbf{k}',\mathbf{k}+\mathbf{q}}^{\sigma''\sigma}(E). \quad (4.4)$$

This can be iterated up to any desired accuracy resulting in spin products of the type

$$(\mathbf{S}_i \cdot \hat{\boldsymbol{\sigma}})_{\sigma'\sigma''} (\mathbf{S}_j \cdot \hat{\boldsymbol{\sigma}})_{\sigma''\sigma'''} (\mathbf{S}_k \cdot \hat{\boldsymbol{\sigma}})_{\sigma'''\sigma''''} \cdots$$

For further unavoidable approximations it is recommended to write the equation of motion in an alternative, but equivalent way by considering the ‘second operator’ in (4.3), $c_{\mathbf{k}+\mathbf{q}\sigma'}^+$, as the ‘active’ operator:

$$(E - \varepsilon(\mathbf{k} + \mathbf{q})) \hat{G}_{\mathbf{k},\mathbf{k}+\mathbf{q}}^{\sigma\sigma'}(E) = \hbar \delta_{\mathbf{q},0} \delta_{\sigma\sigma'} - J \frac{1}{N} \sum_{i\mathbf{k}'\sigma''} e^{-i(\mathbf{k}'-(\mathbf{k}+\mathbf{q}))\cdot\mathbf{R}_i} (\mathbf{S}_i \cdot \hat{\boldsymbol{\sigma}})_{\sigma''\sigma} \hat{G}_{\mathbf{k},\mathbf{k}'}^{\sigma'\sigma''}(E). \quad (4.5)$$

Both equations (4.4) and (4.5) are still exact. Introducing the Green function of the ‘free’-electron system,

$$G_{\mathbf{k}}^{(0)}(E) = \frac{\hbar}{E - \varepsilon(\mathbf{k})} \quad (4.6)$$

we can combine the two equations of motion:

$$\begin{aligned} \hat{G}_{\mathbf{k},\mathbf{k}+\mathbf{q}}^{\sigma\sigma'}(E) &= \delta_{\sigma\sigma'} \delta_{\mathbf{q},0} G_{\mathbf{k}}^{(0)}(E) - J \frac{1}{2N\hbar} \sum_{i\mathbf{k}'\sigma''} \{ e^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}_i} G_{\mathbf{k}}^{(0)}(E) (\mathbf{S}_i \cdot \hat{\boldsymbol{\sigma}})_{\sigma'\sigma''} \hat{G}_{\mathbf{k}',\mathbf{k}+\mathbf{q}}^{\sigma''\sigma}(E) \\ &+ e^{-i(\mathbf{k}'-(\mathbf{k}+\mathbf{q}))\cdot\mathbf{R}_i} G_{\mathbf{k}+\mathbf{q}}^{(0)}(E) (\mathbf{S}_i \cdot \hat{\boldsymbol{\sigma}})_{\sigma''\sigma} \hat{G}_{\mathbf{k},\mathbf{k}'}^{\sigma'\sigma''}(E) \}. \end{aligned} \quad (4.7)$$

The first-order approximation on the r.h.s.

$$\begin{aligned} \hat{G}_{\mathbf{k}',\mathbf{k}+\mathbf{q}}^{\sigma''\sigma}(E) &\rightarrow \delta_{\sigma''\sigma} \delta_{\mathbf{k}',\mathbf{k}+\mathbf{q}} G_{\mathbf{k}+\mathbf{q}}^{(0)}(E) \\ \hat{G}_{\mathbf{k},\mathbf{k}'}^{\sigma'\sigma''}(E) &\rightarrow \delta_{\sigma'\sigma''} \delta_{\mathbf{k},\mathbf{k}'} G_{\mathbf{k}}^{(0)}(E) \end{aligned} \quad (4.8)$$

leads to the following compact expression:

$$[G_{\mathbf{k},\mathbf{k}+\mathbf{q}}^{\sigma'\sigma}(E)]^{(1)} = \delta_{\sigma\sigma'}\delta_{q0}G_{\mathbf{k}}^{(0)}(E) - J\frac{1}{\hbar N}\sum_i e^{i\mathbf{q}\cdot\mathbf{R}_i}G_{\mathbf{k}}^{(0)}(E)(\mathbf{S}_i\cdot\hat{\boldsymbol{\sigma}})_{\sigma'\sigma}G_{\mathbf{k}+\mathbf{q}}^{(0)}(E). \quad (4.9)$$

It will be shown that this approximate result reproduces the ‘normal’ RKKY interaction. In order to incorporate to higher order the exchange-induced polarization of the itinerant conduction electrons, we replace the Green functions on the r.h.s. of the exact equation (4.7) in accordance with (4.8) but using the full single-electron Green function (2.11), which we have evaluated in section 3. Defining

$$\hbar A_{\mathbf{k},\mathbf{k}+\mathbf{q}}^{\sigma'\sigma}(E) = G_{\mathbf{k}}^{(0)}(E)G_{\mathbf{k}+\mathbf{q}\sigma}(E) + G_{\mathbf{k}+\mathbf{q}}^{(0)}(E)G_{\mathbf{k}\sigma'}(E) \quad (4.10)$$

we get

$$\hat{G}_{\mathbf{k},\mathbf{k}+\mathbf{q}}^{\sigma'\sigma}(E) \approx \delta_{\sigma\sigma'}\delta_{q0}G_{\mathbf{k}}^{(0)}(E) - J\frac{1}{2N}\sum_i e^{i\mathbf{q}\cdot\mathbf{R}_i}(\mathbf{S}_i\cdot\hat{\boldsymbol{\sigma}})_{\sigma'\sigma}A_{\mathbf{k},\mathbf{k}+\mathbf{q}}^{\sigma'\sigma}(E). \quad (4.11)$$

In arriving at (4.11) we have neglected the non-diagonal terms ($\mathbf{q} \neq \mathbf{0}, \sigma' \neq \sigma''$) in the ‘restricted’ Green functions on the r.h.s. of (4.7). We believe that this is partly compensated by using in (4.10) the full electronic self-energy $M_{k\sigma}(E)$ (via $G_{k\sigma}(E)$) which in principle incorporates all scattering processes of the conduction electron.

For the effective spin Hamiltonian (4.2) we need the expectation value $\langle c_{\mathbf{k}+\mathbf{q}\sigma}^+ c_{\mathbf{k}\sigma'} \rangle^{(s)}$, which we express in terms of the imaginary part of the Green function (4.11) exploiting the spectral theorem [24]:

$$\frac{1}{N}\sum_{\mathbf{k}} \langle c_{\mathbf{k}+\mathbf{q}\sigma}^+ c_{\mathbf{k}\sigma'} \rangle^{(s)} \approx \frac{1}{2}\delta_{\sigma\sigma'}\delta_{q0}\langle n \rangle - J\frac{1}{2N}\sum_i e^{i\mathbf{q}\cdot\mathbf{R}_i} D_q^{\sigma\sigma'}(\mathbf{S}_i\cdot\hat{\boldsymbol{\sigma}})_{\sigma'\sigma}. \quad (4.12)$$

Here we have defined

$$D_q^{\sigma\sigma'} = -\frac{1}{\pi}\text{Im}\int_{-\infty}^{+\infty} dE f_-(E)\frac{1}{N\hbar}\sum_{\mathbf{k}} A_{\mathbf{k},\mathbf{k}+\mathbf{q}}^{\sigma'\sigma}(E). \quad (4.13)$$

$\langle n \rangle$ is the average number of electrons per site ($0 \leq \langle n \rangle \leq 2$). One recognizes that averaging in the s subspace does not remove operator properties for the f system. Equation (4.12) inserted into (4.2) transforms the f spin Hamiltonian H_f into an ‘effective’ Heisenberg Hamiltonian

$$H_f = -\sum_{i,j} \hat{J}_{ij} \mathbf{S}_i \cdot \mathbf{S}_j. \quad (4.14)$$

The influence of the conduction electrons on the localized spin system is thereby incorporated in an ‘effective’ spin–spin interaction, where the exchange integrals

$$\hat{J}_{ij} = \frac{1}{N}\sum_{\mathbf{q}} \hat{J}(\mathbf{q})e^{-i\mathbf{q}\cdot(\mathbf{R}_i-\mathbf{R}_j)} \quad (4.15)$$

are mainly determined by the electronic self-energy:

$$\hat{J}(\mathbf{q}) = -\frac{1}{8}J^2\sum_{\sigma} D_q^{\sigma\sigma}. \quad (4.16)$$

It is easy to demonstrate that the first-order approach (4.8) is equivalent to the ‘normal’ RKKY interaction. In this case (4.13) simplifies to

$$D_q^{\sigma\sigma} \Rightarrow D_q^{(1)} = -\frac{2}{\pi}\text{Im}\int_{-\infty}^{+\infty} dE f_-(E)\frac{1}{N\hbar^2}\sum_{\mathbf{k}} G_{\mathbf{k}}^{(0)}(E)G_{\mathbf{k}+\mathbf{q}}^{(0)}(E) \quad (4.17)$$

and the effective exchange integrals (4.16) become identical to the well-known RKKY expression

$$J^{(1)}(\mathbf{q}) = -\frac{1}{2N} J^2 \sum_{\mathbf{k}} \frac{f_{-}(\varepsilon(\mathbf{k} + \mathbf{q})) - f_{-}(\varepsilon(\mathbf{k}))}{\varepsilon(\mathbf{k} + \mathbf{q}) - \varepsilon(\mathbf{k})}. \quad (4.18)$$

We shall use the full result (4.16), by which the s-f exchange interaction H_{sf} has been transformed into an indirect coupling between the localized spins, to calculate the ‘pure’ spin correlations (2.16), in particular the f magnetization $\langle S_i^z \rangle$. The exchange integrals \hat{J}_{ij} are decisively influenced by the electronic self-energy, and have therefore to be fixed self-consistently.

In order to determine the f spin correlation functions (2.16) we evaluate the equation of motion of the magnon Green function $P_{ij}^{(a)}(E)$, defined in (2.8), by applying the f spin Hamiltonian (4.14). Within the so-called Tyablikov approximation [28] one finds straightforwardly for the wave-vector-dependent magnon Green function

$$P_q^{(a)}(E) = \frac{\langle A^{(a)} \rangle}{E - E(\mathbf{q}) + i0^+} \quad (4.19)$$

$$\langle A^{(a)} \rangle = \langle [S_i^+, e^{aS_i^z} S_i^-]_- \rangle. \quad (4.20)$$

The $E(\mathbf{q})$ are the ‘effective’ magnon energies

$$E(\mathbf{q}) = 2\langle S^z \rangle (\hat{J}_0 - \hat{J}(\mathbf{q})) \quad (4.21)$$

$$\hat{J}_0 = \sum_i \hat{J}_{im} = \sum_m \hat{J}_{im} = \hat{J}(\mathbf{q} = \mathbf{0}). \quad (4.22)$$

In determining $\langle S^z \rangle$ for an arbitrary spin value S we use the method first proposed by Callen [26]. Details are given in appendix D. We are listing here only the final results for the important f spin correlations.

The result for the f spin magnetization reads

$$\langle S^z \rangle = \hbar \frac{(1 + S + \varphi)\varphi^{2S+1} + (S - \varphi)(1 + \varphi)^{2S+1}}{(1 + \varphi)^{2S+1} - \varphi^{2S+1}}. \quad (4.23)$$

Here we have used the definition

$$\varphi(S) = \frac{1}{N} \sum_{\mathbf{q}} \frac{1}{e^{\beta E(\mathbf{q})} - 1}. \quad (4.24)$$

φ depends on S via the magnon energies $E(\mathbf{q})$. Since the conduction electron self-energy $M_{q\sigma}(E)$ appears in the ‘effective’ exchange integrals \hat{J}_{ij} , it decisively influences $\varphi(S)$ and thereby the f spin magnetization $\langle S^z \rangle$. Another quantity of interest is

$$\langle S^- S^+ \rangle = 2\hbar \langle S^z \rangle \varphi(S). \quad (4.25)$$

We combine (4.23) and (4.25) to get

$$\langle (S^z)^2 \rangle = \hbar^2 S(S + 1) - \hbar \langle S^z \rangle (1 + 2\varphi(S)). \quad (4.26)$$

Furthermore we need

$$\langle (S^z)^3 \rangle = \hbar^3 S(S + 1)\varphi(S) + \hbar^2 \langle S^z \rangle (S(S + 1) + \varphi) - \hbar \langle (S^z)^2 \rangle (1 + 3\varphi). \quad (4.27)$$

Using (4.23) to (4.27), all f spin correlations (2.16), which appear in the representation (3.33) of the electronic self-energy, are expressed in terms of $\varphi(S)$. Thereby we have found a closed system of equations that can be solved self-consistently for all quantities of interest, in particular those which inform us about the mutual influence of magnetic and electronic

properties of the exchange-coupled system of itinerant conduction electrons and localized f spins.

A central magnetic quantity is of course the Curie temperature T_c , which can be read off from (4.23), because

$$T \rightarrow T_c^{(-)} \Leftrightarrow \langle S^z \rangle \rightarrow 0^+. \quad (4.28)$$

One finds the rather simple formula:

$$k_B T_c = \frac{2}{3} S(S+1) \left[\frac{1}{N} \sum_q (\hat{J}_0 - \hat{J}(q))_{T_c}^{-1} \right]^{-1}. \quad (4.29)$$

The ‘effective’ exchange integrals are temperature dependent. They have to be used in (4.29) for $T \rightarrow T_c$.

5. Discussion of the results

We have evaluated our theory for a simple cubic tight-binding Bloch density of states as given by Jelitto [29]. Our considerations are restricted to paramagnetic and ferromagnetic phases only, so that we can exploit the translational symmetry of the full lattice. The main goal is to get a qualitatively correct picture of the mutual influence of localized magnetic moments and itinerant conduction electrons. The interest is especially focused on typical correlation effects and temperature dependencies in the energy spectra, which should be spectroscopically observable.

5.1. The electronic quasiparticle structure

A quantity which is more or less directly related to angle- and spin-resolved (inverse) photoemission experiments, except for as regards the dipole transition matrix element, is the wave-vector- and spin-dependent single-electron spectral density, defined in (2.12). The position of a prominent $S_{k\sigma}(E)$ -peak corresponds to a corresponding quasiparticle excitation. In spite of the fact that the conduction electrons are ‘*a priori*’ non-magnetic, drastic magnetic correlation effects appear as a consequence of the exchange coupling to the localized moment system. Figure 1 shows the energy dependence of the spectral density for wave vectors \mathbf{k} from the Λ direction, and that for different temperatures in between $T = 0$ K and $T = T_c$. The conduction band is for $n = 0.2$ less than half-filled. At $T = 0$ K the \uparrow spectral density consists of a very sharp, δ -function-like peak. The resulting quasiparticle dispersion $E_{\uparrow}(\mathbf{k})$ has a very simple shape, being almost identical to the ‘free’ dispersion $\varepsilon(\mathbf{k})$ except for a rigid shift of about $-(1/2)JS$. The reason for this is that for $T = 0$ K a spin exchange of the excited \uparrow electron with the parallel-aligned localized spin system is excluded. The ‘spin-flip’ terms in the s–f interaction (2.5) do not work; only the Ising part of H_{sf} accounts for the rigid shift. The \downarrow electron, on the other hand, can even at $T = 0$ K exchange its spin with the f system. This may happen according to two elementary processes. The first one is a repeated emission and reabsorption of a magnon by the itinerant electron resulting in an effective electron–magnon attraction. This gives rise to a polaron-like quasiparticle, which we call the ‘magnetic polaron’. The electron may be considered as moving through the lattice accompanied by a cloud of magnons. The other excitation is due to a direct magnon emission by the conduction electron without reabsorption. In that case the original \downarrow particle becomes a \uparrow particle. This process is of course possible only if there are \uparrow states within reach. Corresponding electron states therefore must coincide with the \uparrow spectrum. This can clearly be seen in figure 2 where we have plotted the quasiparticle density of states (2.13)

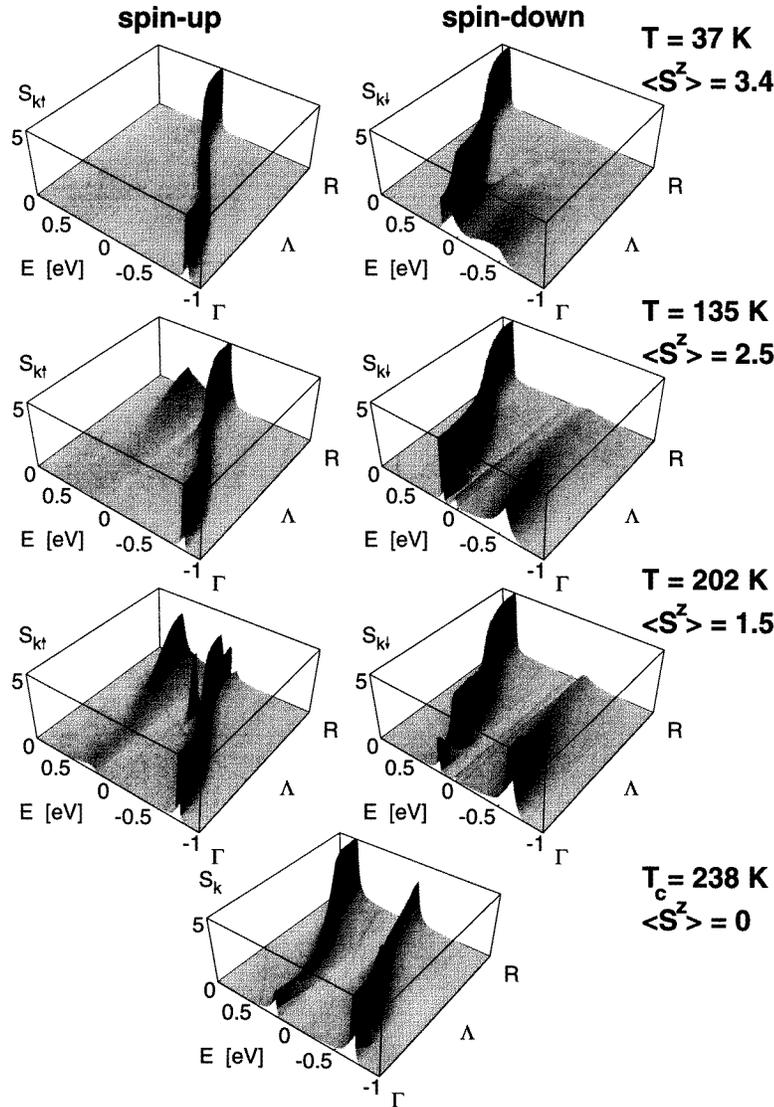


Figure 1. The spectral density as a function of the energy E for wave vectors from the Λ direction and for four different temperatures. $T_c = 238$ K is the self-consistently calculated Curie temperature. Parameters: $J = 0.2$ eV, $W = 1$ eV, $n = 0.2$, $S = 7/2$; sc lattice.

for the same parameter set and lattice type as for figure 1 for the spectral density. $\rho_{\downarrow}(E)$ has at $T = 0$ K a low-energy tail in the region of $\rho_{\uparrow}(E) \neq 0$ due to the above-mentioned magnon emission ('scattering states'). The high-energy part of $\rho_{\downarrow}(E)$ (for E -values with $\rho_{\uparrow}(E) = 0$) consists of quasiparticle excitations which correspond to the just-explained magnetic polaron states. In special situations [23, 25] (a single electron in an otherwise empty band) this may even lead to a bound state, i.e. to a quasiparticle with an infinite lifetime. The \downarrow spectral density near the R point (figure 1) consists at $T = 0$ K of a single, rather sharp peak that represents a well-defined—say, long-living—magnetic polaron. To

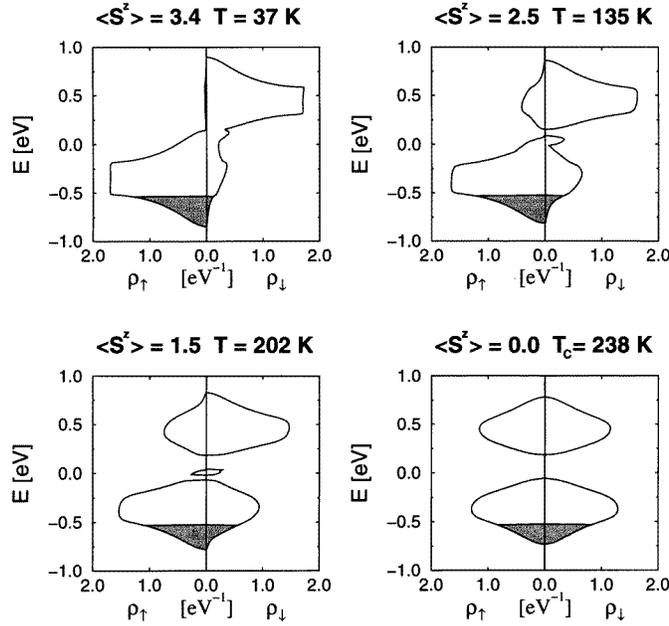


Figure 2. The spin-dependent quasiparticle density of states ρ_σ as a function of energy for four different temperatures. The parameters are as in figure 1; sc lattice. Left-hand part: $\sigma = \uparrow$; right-hand part: $\sigma = \downarrow$. The shaded region marks the occupied part of the energy band.

summarize, the QDOS $\rho_\downarrow(E)$ consists at $T = 0$ K (ferromagnetic saturation of the f spin system, $\langle S^z \rangle = S$) in the upper part of polaron states, and in the lower part of scattering states (figure 2). On the other hand $\rho_\uparrow(E)$ is fully built up of undamped quasi-free-electron states.

Magnon emission by a down-spin electron should be equivalent to magnon absorption by an up-spin electron. At finite temperatures the f magnetization $\langle S^z \rangle$ away from saturation produces a finite magnon density. The \uparrow electron has then indeed the chance to absorb a magnon, thereby reversing its own spin. One therefore recognizes at $T > 0$ scattering as well as polaron states in the \uparrow spectrum, too. The low-energy peak of the spectral density $S_{k\sigma}(E)$ (figure 1) corresponds, strictly speaking, to two elementary processes. The excited σ -electron retains its spin and enters the local frame of the localized spin parallel to the f spin. The other possibility is that the s electron first flips its spin via magnon emission/absorption and then enters as a $-\sigma$ -electron the local frame. At finite temperature both the σ -electron and the $-\sigma$ -electron can orientate the spin in the local frame with finite probability parallel to the f spin. In the second process a magnon is involved, in the first not. For $T \neq 0$ the low-energy peak of the spectral density is only partly built up of pure scattering states. The ferromagnetically saturated spin system represents a special case (a 'magnon vacuum') because then the low-energy part of the spectral density consists for $\sigma = \downarrow$ only of scattering states, and for $\sigma = \uparrow$ only of states without magnon contributions. Because of the normalization condition

$$\int_{-\infty}^{+\infty} dE S_{k\sigma}(E) = 1 \quad (5.1)$$

each peak in the spectral density is connected with a spectral weight which strongly changes

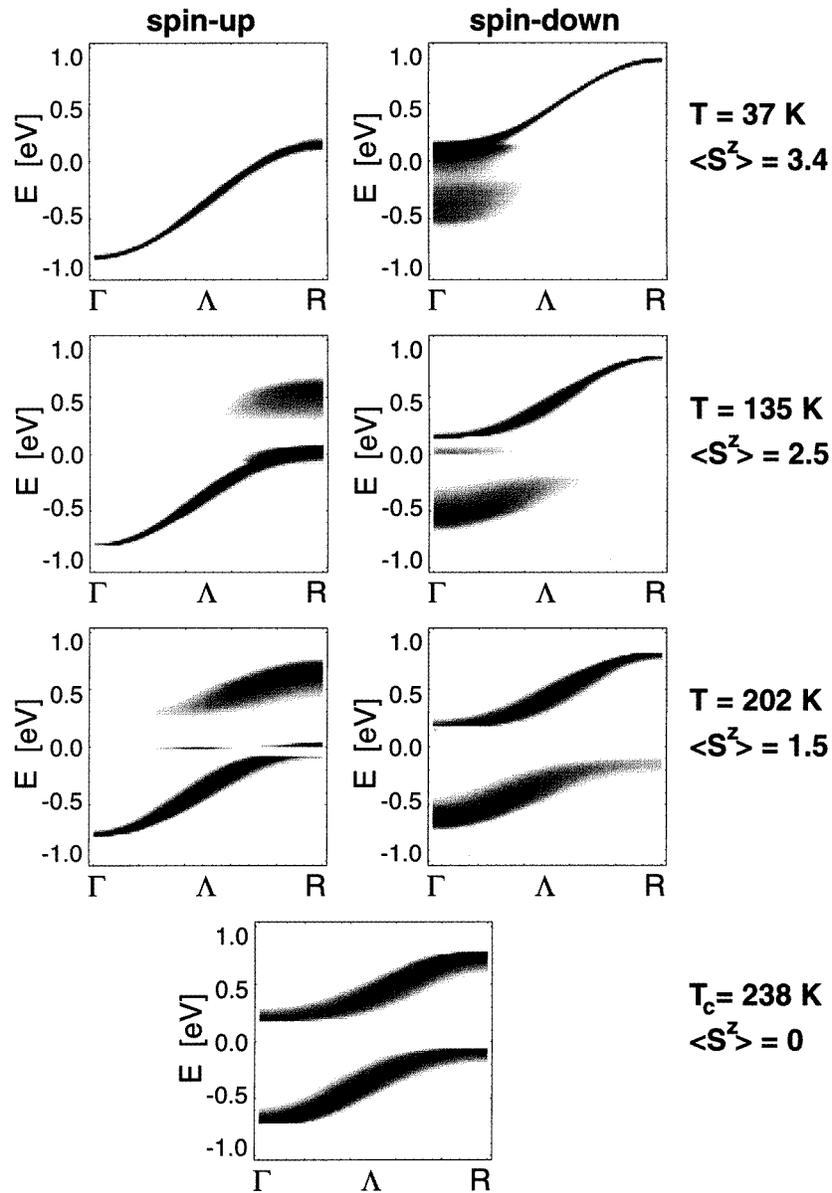


Figure 3. The quasiparticle dispersion, represented as a density plot of the spectral density, for wave vectors from the Λ direction and for four different temperatures. The parameters are as in figure 1; sc lattice. The degree of shading is a measure of the spectral weight of the quasiparticle excitation.

with temperature and exhibits a remarkable k -dependence.

In conclusion, we observe for an s - f coupling $J/W = 0.2$ strong correlation effects in the band electron spectrum due to the exchange interaction with the localized f spin system (figures 1, 2). Normally each (k, σ) excitation splits into two peaks, which may be classified as the polaron and scattering peaks. The spectral weights of these two peaks are

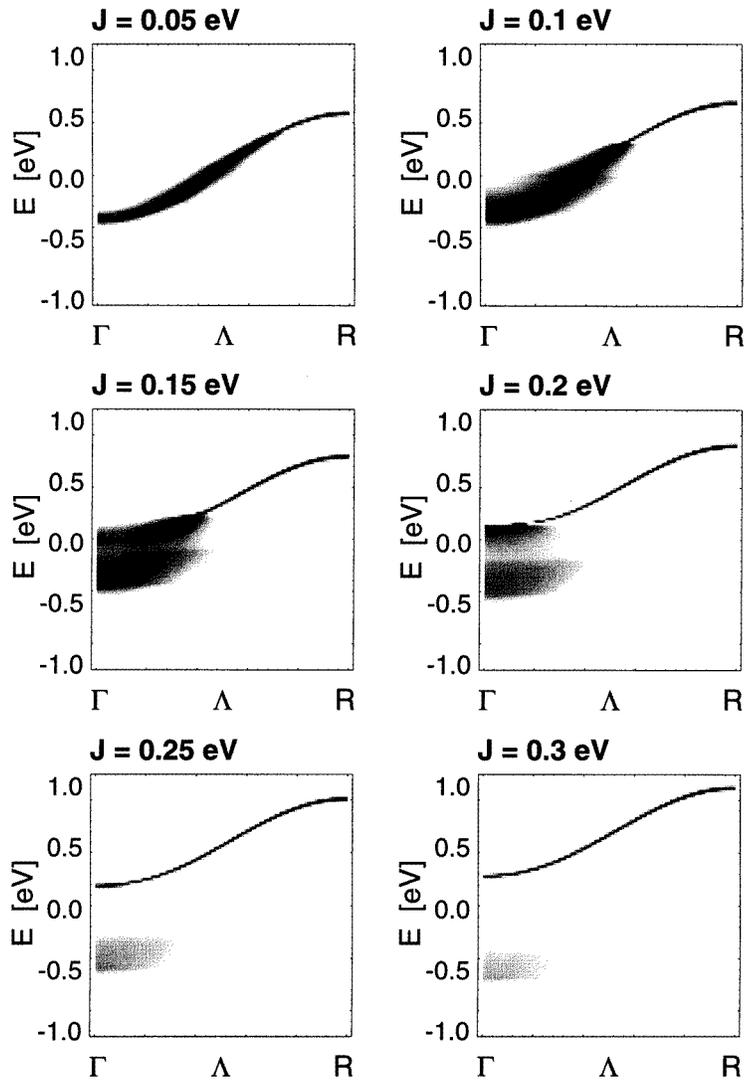


Figure 4. The spin-down quasiparticle dispersion, at $T = 0$, represented as a density plot of the spectral density (cf. figure 3), for various values of the s - f exchange coupling J in the low-density limit. Parameters: $W = 1$ eV, $S = 7/2$; sc lattice.

strongly k - and T -dependent, where the T -dependence comes into play almost exclusively via the magnetic state of the f spins. Since our theory models local moment ferromagnets such as Gd, it suggests thinking about the consequences of the correlation effects in the electronic quasiparticle spectrum for a corresponding angle- and spin-resolved direct or inverse photoemission experiment. Such an experiment obviously requires a careful interpretation beyond the usual one-electron picture. To clarify the situation we use in figure 3 a density plot for the spectral density by taking the degree of shading as a measure of the intensity, thereby mapping a ‘weighted’ quasiparticle band-structure. The \downarrow spectrum

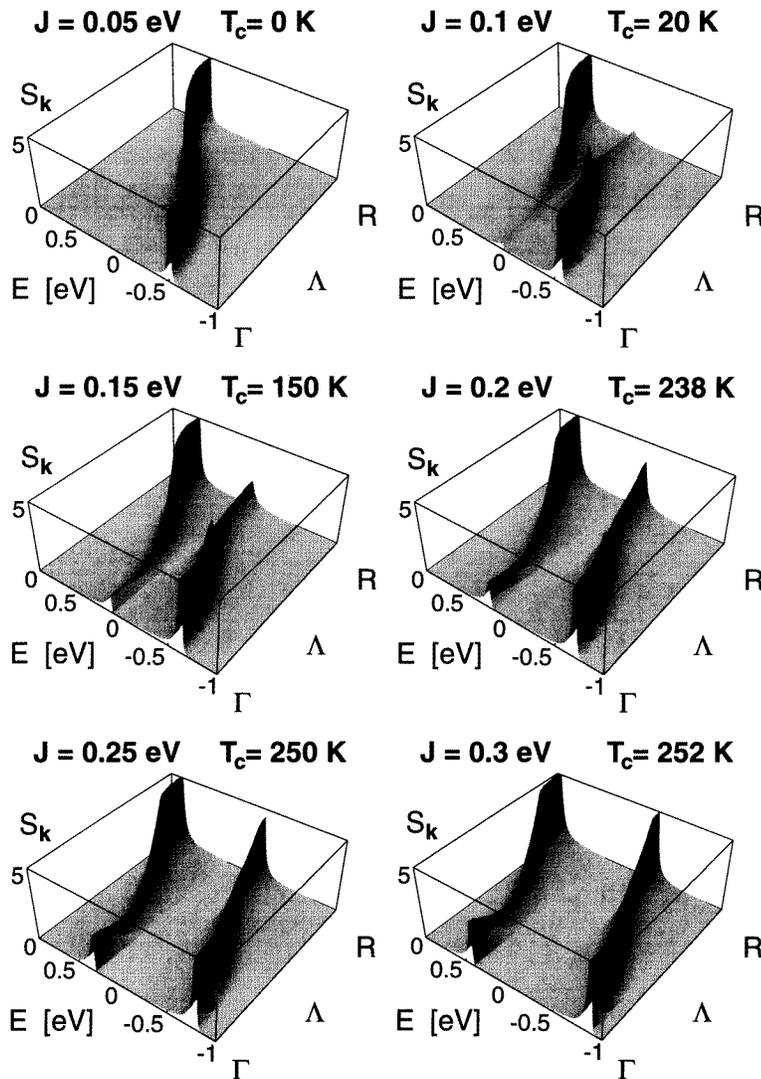


Figure 5. The spin-down spectral density at $T = T_c$ (paramagnetic phase) as function of energy for wave vectors from the Λ direction and for various values of the exchange coupling J . The band occupation $n = 0.2$. The T_c -values change with J .

at very low temperatures is dominated by the polaron peak being, however, accompanied by a broad scattering spectrum for wave vectors not too far away from the Γ point. With increasing temperature the polaron peak survives but the scattering spectrum (the low-energy part) is more and more bunched to a prominent peak. In the \uparrow spectrum it is the polaron part that gains weight with increasing temperature. The photoemission will show for each k -vector two excitations per spin with strongly wave-vector- and temperature-dependent spectral weights.

It is interesting to compare these results with those for an only weakly coupled system. No splitting appears, and the spectrum can be understood in a one-electron picture. The

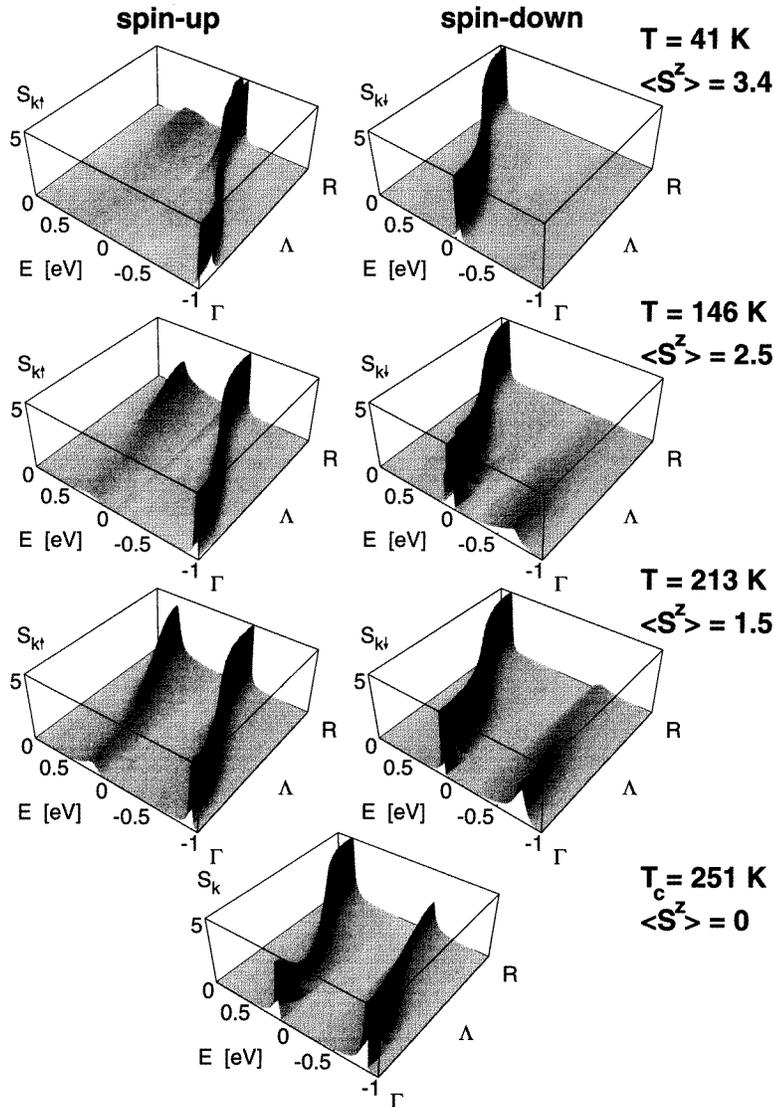


Figure 6. The spectral density of a band that is more than half-filled ($n = 1.8$) as a function of the energy E for wave vectors from the Λ direction and for four different temperatures. $T_c = 251$ K is the self-consistently calculated Curie temperature for the following parameters: $J = 0.25$ eV, $W = 1$ eV, $S = 7/2$; sc lattice.

electronic self-energy (3.33) is well approximated by the linear term $-(1/2)Jz_\sigma \langle S^z \rangle$, which is responsible for a ‘Stoner-like’ temperature shift of the dispersions. The quasiparticle density of states shows only slight deviations from the ‘free’ Bloch density of states. On a plausible level the strikingly different results for intermediate and for weak exchange couplings J/W can easily be interpreted. For weak couplings the conduction electron possesses a relatively high mobility. Therefore, it ‘sees’ the total magnetization of the f spin system, which builds up something like an internal magnetic field causing the ‘Stoner-

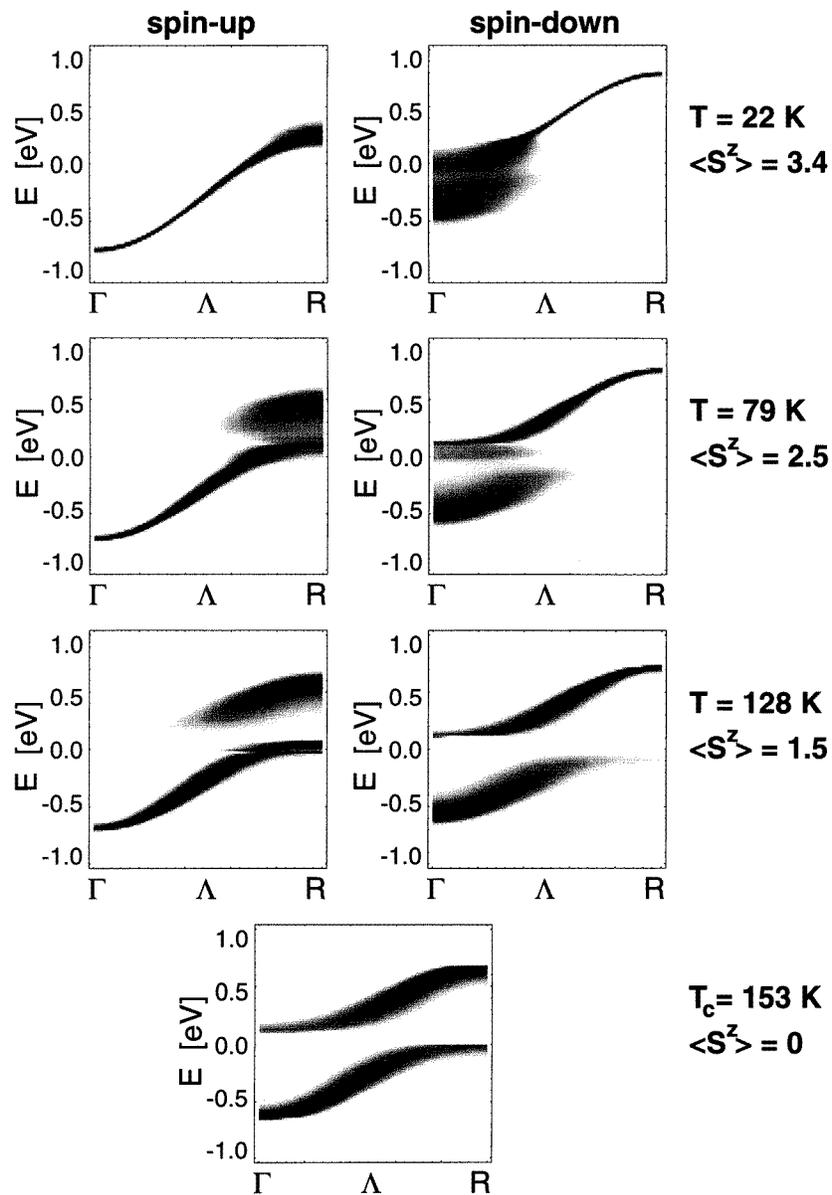


Figure 7. The quasiparticle dispersion, represented as density plot of the spectral density, for wave vectors from the Λ direction and for four different temperatures. Parameters: $J = 0.15$ eV, $W = 1$ eV, $n = 0.1$, $S = 7/2$; sc lattice. The degree of shading is a measure of the spectral weight of the quasiparticle excitation.

like' shift proportional to $\langle S^z \rangle$. For stronger exchange couplings J/W the local frame becomes decisive. The conduction electron spin can fix its position parallel or antiparallel to the local f spin. The two arrangements require two different energies which do not change very much with temperature; only the probabilities that the electron enters the local frame as an \uparrow or a \downarrow electron become distinctly T -dependent. This leads to temperature-dependent

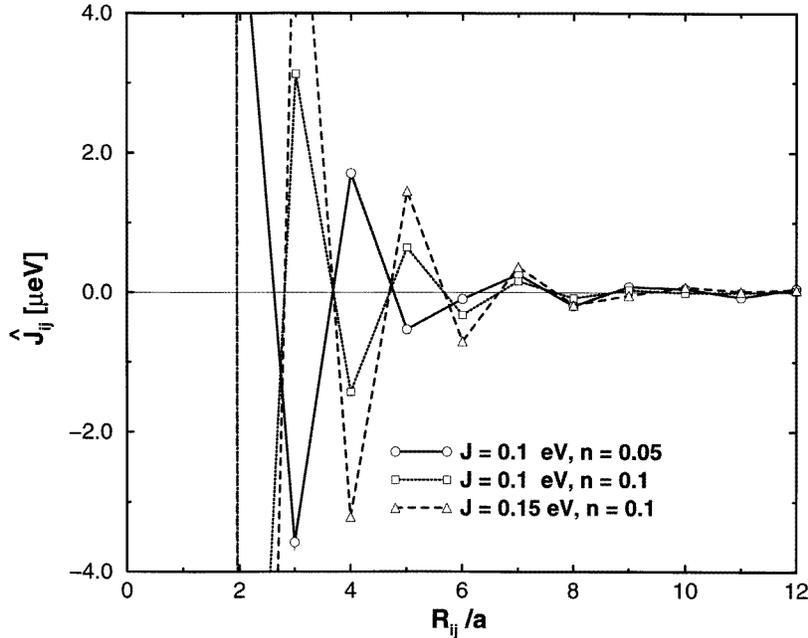


Figure 8. The effective exchange integral \hat{J}_{ij} as a function of the distance R_{ij} in the (100) direction of the sc lattice for non-interacting band electrons (RKKY). Parameters: $J = 0.2$ eV, $W = 1$ eV. Solid line: $J = 0.1$ eV, $n = 0.05$; dotted line: $J = 0.1$ eV, $n = 0.1$; broken line: $J = 0.15$ eV, $n = 0.1$.

spectral weights of the quasiparticle peaks, the positions of which, however, remain more or less unaffected (figure 1).

The correlation-caused splitting of the single-electron spectral density sets in when the s-f coupling strength J/W exceeds a critical value of about 0.1. Figure 4 demonstrates the J -dependence of the down-spin quasiparticle dispersion for a ferromagnetically saturated f system and for a very small band occupation n . The non-trivial, rigorously solvable limiting case of a single electron in an otherwise empty conduction band [23–25, 31] is exactly reproduced by our theory. Therefore, the results for $T = 0$ and $n = 0.01$ in figure 4 should be highly reliable, proving that the splitting of the down-spin spectral density into a polaronic and a scattering portion is not at all an artefact of unavoidable approximations used in the calculations. It is a fundamental feature of the underlying physical system. The polaronic part is always a sharp structure, representing thereby a quasiparticle with a sufficiently long lifetime. The scattering region is rather broad with a remarkable k -dependence. Near the Γ point the scattering states have especially high spectral weights at the cost of the magnetic polaron. For strongly coupled systems, $J/W > 0.2$, the polaron and scattering parts are split, i.e. the decay of the polaron via magnon emission becomes impossible. For weaker coupling the polaron peak dips for certain wave vectors into the scattering region, gaining thereby the possibility of flipping the electron spin by emitting a magnon. It is interesting to recognise that at the Curie point T_c (figures 1, 5) the two structures in the paramagnetic single-electron spectral density both appear as prominent quasiparticle peaks. The splitting sets in for $J/W \geq 0.1$. The relative shift of the two peaks increases with J , approximately according to $(1/2)J(2S + 1)$. In the zero-bandwidth

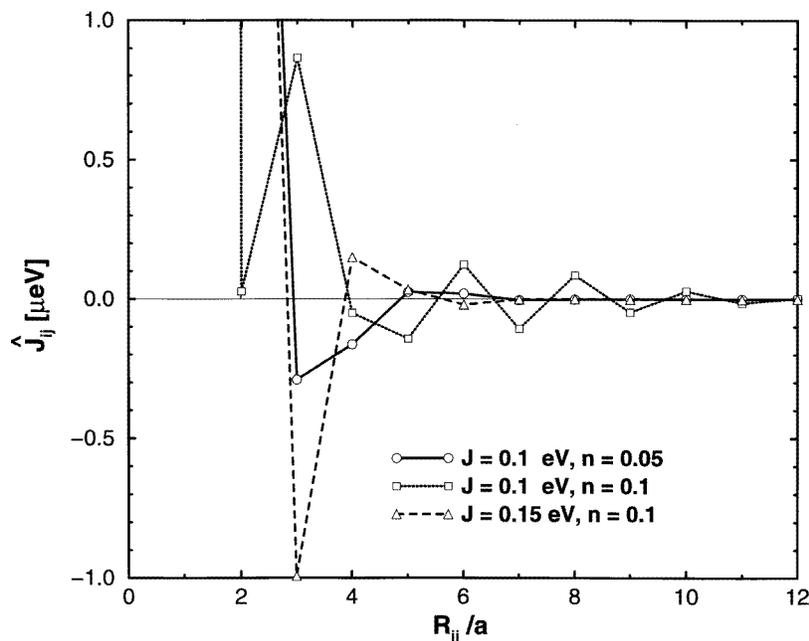


Figure 9. The effective exchange integral \hat{J}_{ij} as a function of the distance in the (100) direction calculated with the full theory (4.16). Solid line: $J = 0.1$ eV, $n = 0.05$; dotted line: $J = 0.1$ eV, $n = 0.1$; broken line: $J = 0.15$ eV, $n = 0.1$. The temperature is chosen so that the magnetization amounts to $\langle S^z \rangle = 3.0$. Parameters: $W = 1$ eV, $S = 7/2$; sc lattice.

limit $W \rightarrow 0$, this is an exact result [28]. The T_c -values, used in figure 5, are self-consistently calculated (see the next subsection) and of course strongly change with the coupling strength J . The correlation effects for more-than-half-filled bands ($n > 1$) are compatible with those that we just discussed for less-than-half-filled bands ($n < 1$). We have only to apply the particle (σ)-hole ($-\sigma$) symmetry. An example is given in figure 6. At $T = 0$ the down-spin spectrum is now simple while the up-spin spectrum consists of a low-energy polaron peak and high-energy scattering states. The critical value of the exchange coupling J/W , for which the gap between the polaron and scattering parts of the spectrum opens, turns out to be temperature dependent. Figure 7 shows an example for a moderate coupling $J/W = 0.15$ and a less-than-half-filled energy band. For low temperatures the two quasiparticle subbands are overlapping, while a gap appears when the temperature approaches the Curie point. This is accompanied by a substantial quasiparticle band narrowing.

5.2. Magnetic properties

The spin polarization of the conduction electrons induced by the exchange coupling with the local f-moment system leads to an indirect coupling between the localized spins (4.14). This type of exchange coupling is referred to in the literature as RKKY interaction [16–18]. The original derivation is performed within perturbation theory of second order leading for the free-electron gas to the well-known oscillatory behaviour of the effective exchange integral (figure 8). The band occupation n ($0 \leq n \leq 2$) determines the Fermi edge and

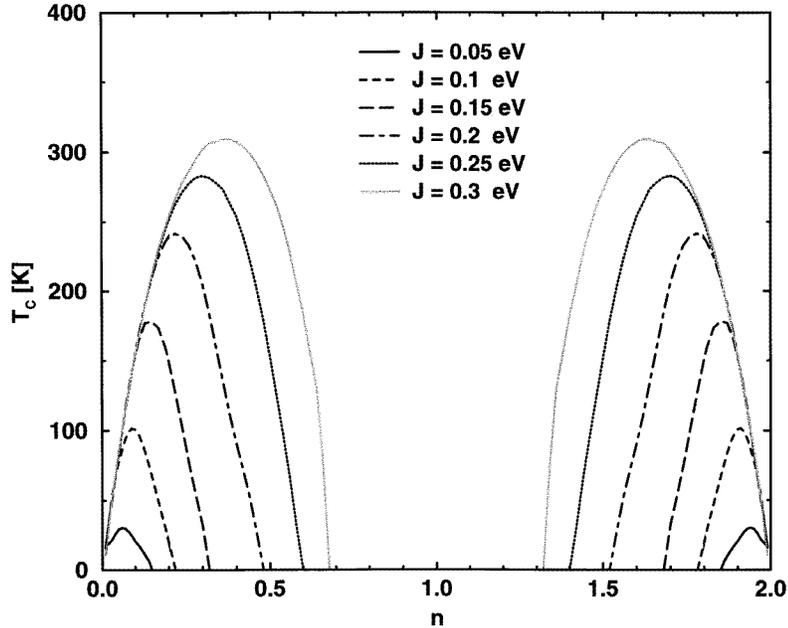


Figure 10. The Curie temperature as a function of the band occupation n ($0 \leq n \leq 2$) for various values of the s-f exchange coupling J . Other parameters: $W = 1$ eV, $S = 7/2$; sc lattice.

thereby the wavelength of the oscillation (4.18). The perturbation theory does not lead to any temperature dependence of the indirect exchange coupling. That becomes different in our extended theory. The effective exchange integrals (4.15) are decisively influenced by the conduction electron self-energy. That implies in particular a remarkable temperature dependence. The example plotted in figure 9 demonstrates that the oscillations of the exchange integral are far less regular and are substantially damped compared to those of the ‘conventional’ RKKY theory for the free electron. The shape of the distance-dependent coupling \hat{J}_{ij} differs strongly for different symmetry directions. The influence of temperature on the electronic self-energy is mainly due to the magnetization $\langle S^z \rangle$ of the f system and that feeds back to the indirect f-f interaction and the effective magnon energies $E(q)$ of (4.21).

One of the key quantities of a ferromagnetic material is the Curie temperature of (4.29). We stress once more that we did not consider a direct exchange between the localized spins, so a ferromagnetic order must come out self-consistently as a consequence of the mutual influence of the conduction electrons and the localized spins. The various results are gathered together in figure 10. No ferromagnetism is possible around the half-filled band ($n = 1$). The Curie temperature has a maximum for the quarter-filled (three-quarter-filled) band ($n = 0.5$ and 1.5) and increases with the s-f coupling J . The RPA treatment of the effective spin Hamiltonian in section 4 certainly overestimates the magnetism somewhat, but, nevertheless, realistic values of the transition temperatures are found.

The finiteness of the Curie temperature is exclusively due to the s-f exchange J between the local moment system and the conduction electrons. For small J the conventional RKKY behaviour (4.18) is valid. From perturbation theory of second order, it leads to a J^2 -dependence of the transition temperature. This behaviour is correctly reproduced by our

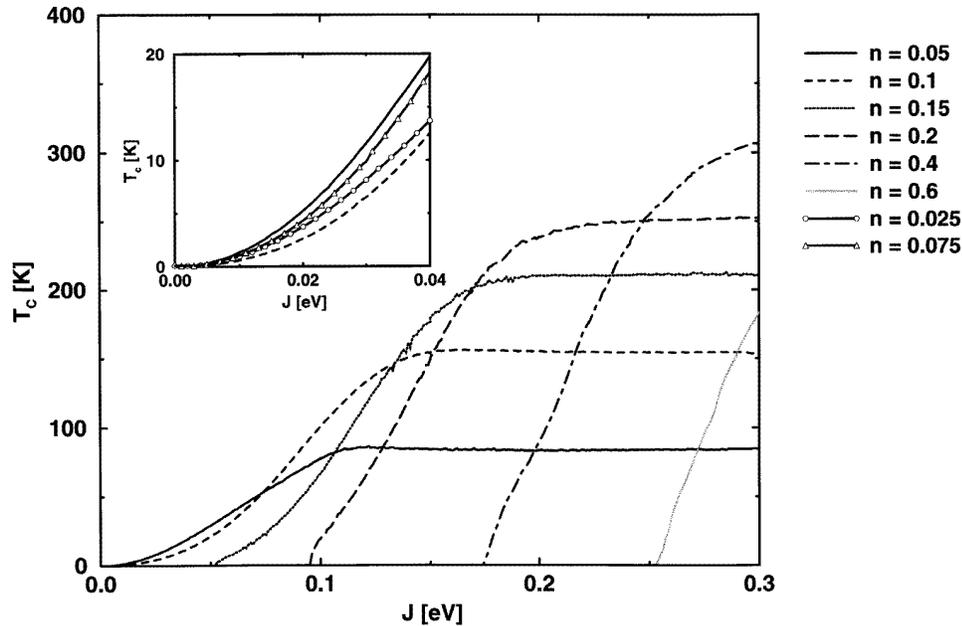


Figure 11. The Curie temperature as a function of the s-f exchange coupling J for various values of the band occupation n . Other parameters: $W = 1$ eV, $S = 7/2$; sc lattice.

theory as can be seen in figure 11. For higher band occupations ($n \geq 0.2$), however, J must exceed a critical value to allow ferromagnetism (figure 10). With increasing exchange coupling the Curie temperature deviates more and more from the RKKY prediction, which finally leads to saturation. This saturation is reached for smaller J -values for lower band occupations n . This feature is certainly a physically reasonable result. As soon as the splitting of the energy band into the two quasiparticle subbands occurs (figure 2) a further increasing exchange interaction J will no longer change the magnetic behaviour of the local moment system very much. A perturbational treatment of the problem is restricted to rather weakly coupled systems.

The f spin correlation functions (figure 12) exhibit the temperature behaviour typical of Heisenberg ferromagnets. The magnetization curve deviates to a certain degree from the $S = 7/2$ Brillouin function. All of the spin correlations enter the electronic self-energy, being therefore responsible for the temperature dependence. According to the results (4.23) to (4.27) the correlation functions are fully determined by the quantity $\varphi(S)$ that can be interpreted as an average magnon number. Consequently it disappears for $T = 0$ (the 'magnon vacuum') and increases rapidly with increasing temperature. Our theory correctly reproduces the exact limiting value of $(1/3)S(S+1)$ for $\langle(S^z)^2\rangle$ in the paramagnetic region $T \geq T_c$. The temperature dependence of the induced conduction band polarization $m = \langle n_\uparrow \rangle - \langle n_\downarrow \rangle$ is very similar to that of the f magnetization (figure 12). The correlation functions γ_\uparrow and γ_\downarrow may be considered as rates of spin fluctuation between itinerant and localized spin systems. Assuming that they are real quantities, they must be identical. The small deviations to be seen in figure 13 are due to our approximate procedure. The mixed spin correlation $\langle \mathbf{S} \cdot \boldsymbol{\sigma} \rangle = (1/2)(\Delta_\uparrow - \Delta_\downarrow + \gamma_\uparrow + \gamma_\downarrow)$ is always positive, indicating a parallel alignment of the f spin and conduction electron spin.

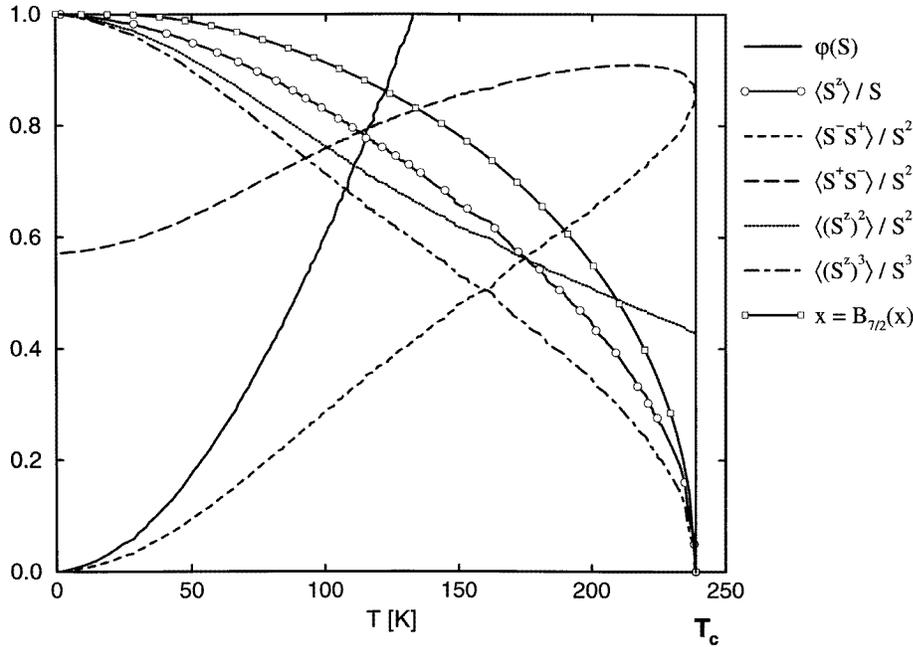


Figure 12. Correlation functions of the localized f spin system as functions of the temperature. Parameters: $J = 0.2$ eV, $W = 1$ eV, $n = 0.2$, $S = 7/2$; sc lattice.

6. Conclusions

We have presented an approximate but self-consistent theory for a metallic local moment ferromagnet described within the framework of the s-f model. The physical properties are determined by an exchange interaction between itinerant conduction electrons and localized f electrons which build up Hund's-rule-coupled localized spins ($S = 7/2$). The decisive model parameters and features are the coupling strength J , the lattice structure, the band occupation n ($0 \leq n \leq 2$) and the temperature T . The model simulates metallic ferromagnets such as the rare earth gadolinium, and will be applied in a forthcoming paper to such real substances. The exchange interaction induces characteristic quasiparticle effects on the conduction band which manifest themselves in the form of striking temperature dependencies, band deformations and splittings. The observed features can be understood as consequences of three basic elementary processes. The excited electron may hop over lattice sites where it orientates its spin parallel to the local f spin, or it may first emit or absorb a magnon thereby flipping its own spin and then hopping with parallel spin orientation over respective lattice sites. The third process can be interpreted as a repeated emission and reabsorption of a magnon by the electron which gives rise to a polaronic quasiparticle (a 'magnetic polaron'). In a certain sense the electron moves through the lattice accompanied by a cloud of virtual magnons.

The f spins polarize the conduction electrons and that feeds back as an indirect coupling between the f spins. In second-order perturbation theory one gets the well-known RKKY mechanism with oscillating sign of the effective exchange integral. The RKKY mechanism is valid of course only for small s-f coupling J , giving rise then to a J^2 -dependence of the resulting Curie temperature. For stronger coupling the effective exchange integrals become

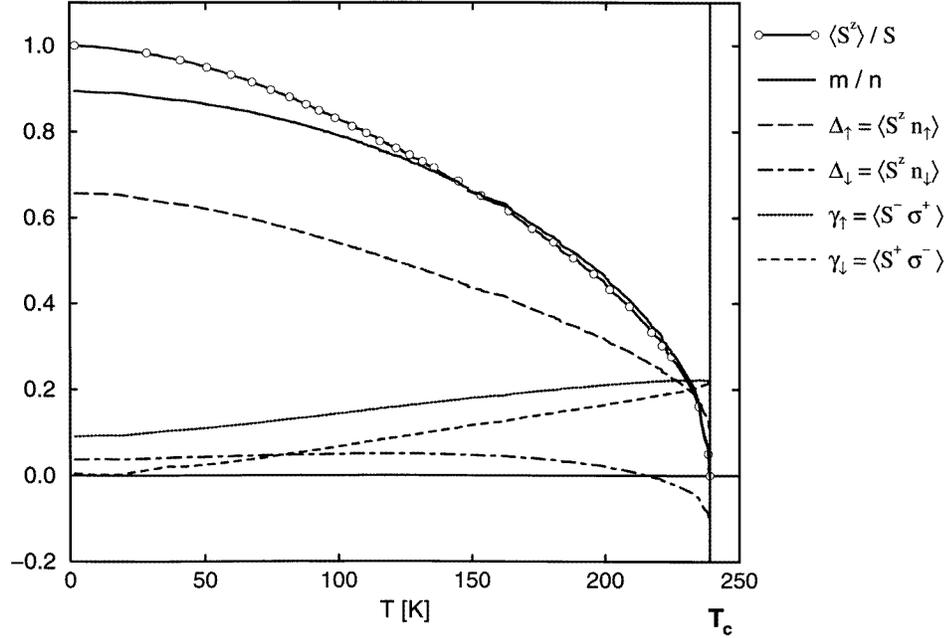


Figure 13. The magnetization-normalized electron spin polarization m/n , the ‘Ising correlation’ Δ_s , and the spin exchange rate γ_s as functions of the temperature. Parameters: $J = 0.2$ eV, $W = 1$ eV, $n = 0.2$, $S = 7/2$; sc lattice.

complicated functionals of the electronic self-energy, deviating substantially then from the RKKY picture. The oscillations are no longer so regular as in the perturbational treatment, but still take care of the fact that ferromagnetism is possible only in restricted regions of the band occupation. Furthermore one finds that T_c saturates with increasing J , where the saturation value is strongly dependent on the band occupation n .

It is intended to apply the above-presented theory to real substances such as Gd, Dy, and Tb. For this purpose we have to combine our many-body model calculation presented in this paper with a one-electron band-structure calculation based on density functional theory. This procedure has previously been performed for the ferromagnetic Heisenberg insulators EuO and EuS [3–5].

Acknowledgment

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Appendix A

A straightforward evaluation yields the following expressions:

$$\begin{aligned} & \langle \langle S_i^z [c_{i\sigma}, H_{sf}]_-; c_{j\sigma}^+ \rangle \rangle_E + z_\sigma \langle \langle S_i^{-\sigma} [c_{i-\sigma}, H_{sf}]_-; c_{j\sigma}^+ \rangle \rangle \\ & = -\frac{1}{2} J z_\sigma (S(S+1) G_{ij\sigma}(E) - z_\sigma \Gamma_{ii,j\sigma}(E) - F_{ii,j\sigma}(E)) \end{aligned} \quad (\text{A.1})$$

$$\langle\langle [S_i^z, H_{sf}] c_{i\sigma}; c_{j\sigma}^+ \rangle\rangle = -\frac{1}{2} J z_\sigma F_{ii,j\sigma}^{(3)}(E) \quad (\text{A.2})$$

which prove that the ‘higher Ising functions’ on the right-hand side of (3.5) do not require the introduction of further Green functions. The ‘Ising functions’ can all be expressed using terms already introduced.

Appendix B

For fixing the coefficients $\alpha_{i\sigma}, \beta_{i\sigma}$ ($i = 1, \dots, 4$) in the *ansätze* (3.24) to (3.27) we need the first two spectral moments (3.30) for the Green functions $G_{ij\sigma}(E)$ (equation (2.7)), $F_{ii,j\sigma}(E)$ (equation (3.2)), $\Gamma_{ii,j\sigma}(E)$ (equation (3.3)), and $F_{ii,j\sigma}^{(n)}(E)$ for $n = 1, \dots, 4$ (equations (3.16) to (3.19)). Straightforward but somewhat tedious evaluations according to (3.28) yield

$$M_{ij\sigma}^{(0)}(G) = \delta_{ij} \quad (\text{B.1})$$

$$M_{ij\sigma}^{(1)}(G) = T_{ij} - \delta_{ij} \frac{1}{2} J z_\sigma \langle S^z \rangle \quad (\text{B.2})$$

$$M_{ij\sigma}^{(0)}(F) = 0 \quad (\text{B.3})$$

$$M_{ij\sigma}^{(1)}(F) = -\frac{1}{2} J \delta_{ij} (\langle S_i^{-\sigma} S_i^\sigma \rangle - \gamma_\sigma + 2z_\sigma \Delta_{-\sigma}) \quad (\text{B.4})$$

(where γ_σ and Δ_σ are defined in (2.17)),

$$M_{ij\sigma}^{(0)}(\Gamma) = \delta_{ij} \langle S^z \rangle \quad (\text{B.5})$$

$$M_{ij\sigma}^{(1)}(\Gamma) = T_{ij} \langle S^z \rangle + \delta_{ij} \frac{1}{2} J z_\sigma (\gamma_\sigma - \langle (S^z)^2 \rangle) \quad (\text{B.6})$$

$$M_{ij\sigma}^{(0)}(F^{(1)}) = 0 \quad (\text{B.7})$$

$$M_{ij\sigma}^{(1)}(F^{(1)}) = -\delta_{ij} \frac{1}{2} J \{ 3z_\sigma \langle S_i^\sigma S_i^{-\sigma} \rangle - 4 \langle S^z \rangle + \langle S_i^\sigma S_i^{-\sigma} S_i^z \rangle - 2z_\sigma S(S+1)(1 - \langle n_{-\sigma} \rangle) + 4\Delta_{-\sigma} - 3z_\sigma \mu_{-\sigma} - \eta_\sigma \} \quad (\text{B.8})$$

(where the ‘higher’ correlation functions μ_σ, η_σ are given in (2.18)),

$$M_{ij\sigma}^{(0)}(F^{(2)}) = \delta_{ij} \langle S_i^{-\sigma} S_i^\sigma \rangle \quad (\text{B.9})$$

$$M_{ij\sigma}^{(1)}(F^{(2)}) = T_{ij} \langle S_i^{-\sigma} S_i^\sigma \rangle - \delta_{ij} \frac{1}{2} J z_\sigma (\langle S_i^{-\sigma} S_i^\sigma S_i^z \rangle + 2\eta_\sigma) \quad (\text{B.10})$$

$$M_{ij\sigma}^{(0)}(F^{(3)}) = -\delta_{ij} \gamma_\sigma \quad (\text{B.11})$$

$$M_{ij\sigma}^{(1)}(F^{(3)}) = -T_{ij} \gamma_\sigma + \delta_{ij} \frac{1}{2} J (z_\sigma \eta_\sigma - \mu_\sigma - 2z_\sigma \vartheta) \quad (\text{B.12})$$

(where the spin-independent expectation value ϑ is defined in (2.18)), and

$$M_{ij\sigma}^{(0)}(F^{(4)}) = -\delta_{ij} \Delta_{-\sigma} \quad (\text{B.13})$$

$$M_{ij\sigma}^{(1)}(F^{(4)}) = -T_{ij} \Delta_{-\sigma} + \delta_{ij} \frac{1}{2} J (z_\sigma S(S+1) \langle n_{-\sigma} \rangle - z_\sigma \mu_{-\sigma} + \Delta_{-\sigma} - \eta_\sigma). \quad (\text{B.14})$$

These moments are used to fix, by the use of (3.29), the unknown coefficients $\alpha_{i\sigma}, \beta_{i\sigma}$:

$$\alpha_{1\sigma} = 0 \quad (\text{B.15})$$

$$\beta_{1\sigma} = \frac{M_{ij\sigma}^{(1)}(F^{(1)})}{M_{ij\sigma}^{(1)}(F)} \quad (\text{B.16})$$

$$\alpha_{2\sigma} = \langle S_i^{-\sigma} S_i^\sigma \rangle - \beta_{2\sigma} \langle S^z \rangle \quad (\text{B.17})$$

$$\beta_{2\sigma} = \frac{\langle (S^z)^3 \rangle + \langle (S^z)^2 \rangle (z_\sigma - z_\sigma \langle S^z \rangle) - \langle S^z \rangle^2 - 2\gamma_\sigma \beta_{1\sigma}}{\gamma_\sigma - \langle (S^z)^2 \rangle + \langle S^z \rangle^2} \quad (\text{B.18})$$

$$\alpha_{3\sigma} = -\gamma_\sigma \quad (\text{B.19})$$

$$\beta_{3\sigma} = \langle n_\sigma \rangle \quad (\text{B.20})$$

$$\alpha_{4\sigma} = -\Delta_{-\sigma} - \beta_{4\sigma} \langle S^z \rangle \quad (\text{B.21})$$

$$\beta_{4\sigma} = \frac{S(S+1)\langle n_{-\sigma} \rangle - \mu_{-\sigma} + \Delta_{-\sigma} (z_\sigma - \langle S^z \rangle) - z_\sigma \eta_\sigma}{\gamma_\sigma - \langle (S^z)^2 \rangle + \langle S^z \rangle^2}. \quad (\text{B.22})$$

Appendix C

We present an explicit solution for the hierarchy of Green functions introduced in the main text. If we agree upon the following abbreviations:

$$a_\sigma = -\alpha_{1\sigma} + z_\sigma \alpha_{2\sigma} - z_\sigma \alpha_{3\sigma} - z_\sigma S(S+1) \quad (\text{C.1})$$

$$b_\sigma = 1 + z_\sigma \beta_{2\sigma} \quad (\text{C.2})$$

$$c_\sigma = z_\sigma - \beta_{1\sigma} - z_\sigma \beta_{3\sigma} \quad (\text{C.3})$$

then we are left with the following approximate equation of motion of the ‘Ising’ function (3.3):

$$\sum_r \{E \delta_{pr} - T_{pr} - M_{pr\sigma}(E)\} \Gamma_{ir,j\sigma}(E) = \hbar \langle S^z \rangle \delta_{pj} + \delta_{ip} \left\{ \frac{1}{2} J(a_\sigma G_{ij\sigma}(E) + b_\sigma \Gamma_{ii,j\sigma}(E) + c_\sigma F_{ii,j\sigma}(E)) - \sum_r M_{ir\sigma}(E) \Gamma_{ir,j\sigma}(E) \right\}. \quad (\text{C.4})$$

Similarly we use the abbreviations

$$A_\sigma = z_\sigma \alpha_{1\sigma} - \alpha_{2\sigma} - \alpha_{3\sigma} + 2z_\sigma \alpha_{4\sigma} \quad (\text{C.5})$$

$$B_\sigma = -\beta_{2\sigma} + 2z_\sigma \beta_{4\sigma} \quad (\text{C.6})$$

$$C_\sigma = z_\sigma \beta_{1\sigma} - \beta_{3\sigma} \quad (\text{C.7})$$

for the equation of motion of the ‘spin-flip’ function (3.2):

$$\begin{aligned} \sum_r \{E \delta_{pr} - T_{pr} - M_{pr-\sigma}(E)\} F_{ir,j\sigma}(E) + \sum_m z_\sigma \hat{E}_{im} F_{mp,j\sigma}(E) \\ = \delta_{ip} \left\{ \frac{1}{2} J(A_\sigma G_{ij\sigma}(E) + B_\sigma \Gamma_{ii,j\sigma}(E) + C_\sigma F_{ii,j\sigma}(E)) \right. \\ \left. + \sum_m z_\sigma \hat{E}_{im} F_{mi,j\sigma}(E) - \sum_r M_{ir-\sigma}(E) F_{ir,j\sigma}(E) \right\}. \end{aligned} \quad (\text{C.8})$$

The \hat{E}_{im} are the site-dependent Fourier transforms of the ‘dressed’ magnon energies $\hat{E}(\mathbf{q})$, to be derived in equation (4.21):

$$\hat{E}_{im} = 2\langle S^z \rangle (\hat{J}_0 \delta_{im} - \hat{J}_{im}). \quad (\text{C.9})$$

The coupled system (3.1), (C.4), and (C.8) is solved in the wave-vector representation, for the single-electron Green function already explained by equation (2.9) and analogously defined for the ‘higher’ functions:

$$A_{kp,q\sigma}(E) = \frac{1}{N^2} \sum_{ipj} A_{ip,j\sigma}(E) e^{-i(\mathbf{k} \cdot \mathbf{R}_i + \mathbf{p} \cdot \mathbf{R}_p - \mathbf{q} \cdot \mathbf{R}_j)} \quad (A = F, \Gamma). \quad (\text{C.10})$$

In addition we can exploit translational symmetry:

$$A_{kp,q\sigma}(E) = \delta_{k+p,q} A_{q-pp,q\sigma}(E).$$

With the shorthand notation

$$\hat{A}_{q\sigma}(E) = \frac{1}{N} \sum_p A_{q-pp,q\sigma}(E) \quad (A = F, \Gamma) \quad (\text{C.11})$$

we first transform the ‘basic’ equation of motion (3.1):

$$(E - \varepsilon(\mathbf{q}))G_{q\sigma}(E) = \hbar - \frac{1}{2}J(z_\sigma \hat{\Gamma}_{q\sigma}(E) + \hat{F}_{q\sigma}(E)) \stackrel{!}{=} \hbar + M_{q\sigma}(E)G_{q\sigma}(E). \quad (\text{C.12})$$

This means that for the self-energy

$$M_{q\sigma}(E) = -\frac{1}{2}J(z_\sigma \hat{\Gamma}_{q\sigma}(E) + \hat{F}_{q\sigma}(E))G_{q\sigma}^{-1}(E). \quad (\text{C.13})$$

For the ‘higher’ functions on the right-hand side we use the equations of motion (C.4) and (C.8) of, respectively, the Ising and the spin-flip function which, after Fourier transformation, build together with (C.12) a closed system of equations.

Appendix D

We derive the needed correlation functions of the localized spin system. The starting point is the correlation function

$$f_{\mathbf{q}}(a) = \frac{1}{N} \sum_{i,j} e^{i\mathbf{q}\cdot(\mathbf{R}_i - \mathbf{R}_j)} \langle e^{aS_i^z} S_j^- S_i^+ \rangle \quad (\text{D.1})$$

that can be expressed in terms of $P_{\mathbf{q}}^{(a)}(E)$ according to the spectral theorem

$$f_{\mathbf{q}}(a) = \langle A^{(a)} \rangle [\exp(\beta E(\mathbf{q})) - 1]^{-1}. \quad (\text{D.2})$$

A wave-vector summation yields

$$\hat{f}(a) = \frac{1}{N} \sum_{\mathbf{q}} f_{\mathbf{q}}(a) = \langle e^{aS^z} S^- S^+ \rangle = \langle A^{(a)} \rangle \varphi(S). \quad (\text{D.3})$$

Here we have introduced $\varphi(S)$ from (4.24). Trying to find a determining equation for $\langle S^z \rangle$ we first derive a differential equation for

$$\Omega(a) = \langle e^{aS^z} \rangle \quad (\text{D.4})$$

whose solution allows us to fix $\langle S^z \rangle$:

$$\langle S^z \rangle = \frac{d}{da} \Omega(a) \Big|_{a=0}. \quad (\text{D.5})$$

Evaluating the commutator in (4.20) and exploiting the identity

$$S_i^+ S_i^- = \hbar^2 S(S+1) + \hbar S_i^z - (S_i^z)^2 \quad (\text{D.6})$$

one finds

$$\langle A^{(a)} \rangle = \hbar^2 S(S+1)(e^{-\hbar a} - 1)\Omega(a) + \hbar(e^{-\hbar a} + 1) \frac{d}{da} \Omega(a) - (e^{-\hbar a} - 1) \frac{d^2}{da^2} \Omega(a). \quad (\text{D.7})$$

Furthermore, the following obviously holds because of (5.9) and (5.11):

$$\hat{f}(a) = \hbar^2 S(S+1)\Omega(a) - \hbar \frac{d}{da} \Omega(a) - \frac{d^2}{da^2} \Omega(a). \quad (\text{D.8})$$

Inserting (D.7) and (D.8) into (D.3) leads to a differential equation of second order for $\Omega(a)$:

$$\frac{d^2}{da^2}\Omega(a) + \hbar \frac{1 + \varphi(S)(e^{-\hbar a} + 1)}{1 - \varphi(S)(e^{-\hbar a} - 1)} \frac{d}{da}\Omega(a) - \hbar^2 S(S + 1)\Omega(a) = 0. \quad (D.9)$$

A first boundary condition is

$$\Omega(0) = 1. \quad (D.10)$$

As the second condition we use

$$0 = \left\langle \prod_{m_s=-S}^{+S} (S^z - \hbar m_s) \right\rangle = \left[\prod_{m_s=-S}^{+S} \left(\frac{d}{da} - \hbar m_s \right) \right] \Omega(a)|_{a=0} = 0. \quad (D.11)$$

After some tedious manipulations we get as the solution of (D.9) [26]

$$\Omega(a) = \frac{e^{-\hbar a S} \varphi^{2S+1} - e^{\hbar a (S+1)} (1 + \varphi)^{2S+1}}{((1 + \varphi)e^{\hbar a} - \varphi)(\varphi^{2S+1} - (1 + \varphi)^{2S+1})}. \quad (D.12)$$

From this equation we can derive all of the f spin correlation functions in equation (2.16) which enter the expression (3.33) for the electronic self-energy. The terms which are important for our theory are listed as equations (4.23) to (4.27).

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