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Band model for the understanding of ferromagnetism in semiconductors and insulators

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ABSTRACT

To investigate ferromagnetic semiconductors and insulators, such as the famous EuO, EuS, or $CrBr_3$, we propose a hybridized Kondo-lattice model, where, in addition to the conduction electrons, localized moments (e.g., the 4*f*-electrons) are modeled as a strongly correlated band system. The quasi-empty conduction band is weakly filled due to the hybridization term. This activates the intraatomic exchange coupling between conduction and localized electrons. Temperature-dependent phase diagrams and quasiparticle densities of states are presented for various coupling and hybridization strengths. Moreover, the influence of the one-particle energy of the localized electrons E_f is discussed. A comparison with mean field calculations is given at the end of this work.

1. Introduction

Collective magnetism requires the existence of permanent magnetic moments, which arise from either localized or itinerant electrons. The distinction of the magnetic moment's type is of enormous conceptual importance: While itinerant electrons cause band magnetism, particularly known from the classical ferromagnets Fe, Co and Ni, and usually examined within the Hubbard model [1–3], local moment magnetism can be found in insulators and semiconductors (such as EuO, EuS and $CrBr_3$ [4–6]), in the huge class of the heavy fermion systems, in diluted magnetic semiconductors (which are promising candidates for spintronic applications [7–10]), and also in some metallic systems like Gd. Such local moment systems are usually described by spin models (e.g. Heisenberg or Ising model [11,12]) with coupling constants due to direct exchange or any type of superexchange. It remains an important aspect of modern magnetism fundamental research to find a unified theory which depicts the variety of magnetic phenomena. A small step along this path will be presented in this paper.

Since collective magnetism is exclusively realized in solid state bodies, whose electronic structure is represented by energy bands and gaps that consequently determine the magnetic properties, it seems to be the more natural choice to use band models instead of pure spin models. In the present work, we propose a band model that describes semiconductors and insulators, i.e., local moment systems. For our purposes, the well-known Kondo-lattice model [13], with local spins replaced by correlated *f*-orbitals, is a formidable starting point as the interaction between subsystems (in our case: localized and itinerant electrons) may lead to magnetic ordering. Note, however, that the interaction is of indirect nature (e.g. Ruderman–Kittel–Kasuya–Yosida interaction [14–16], hereafter referred to as RKKY), thus, in search of spin-ordering effects, requiring at least a minimum of electrons in each subsystem. The problem one is now confronted with is that semiconductors, and insulators likewise, have quasi-empty conduction bands, making indirect electronic exchange impossible. In our work, we therefore extend the Kondo-lattice model by a hybridization term, which gives localized electrons the opportunity to virtually transform into conduction band electrons and vice versa. This solves the problem of beforehand absent conduction electrons, hence allowing a virtual RKKY interaction, but also destabilizes the magnetic moments in the different subsystems. As we can read off from our results, the last fact is of great importance for the understanding of the magnetism found with our model proposed.

The setup of this paper is as follows: In Section 2 we present the Hamiltonian, which characterizes the many-body problem that we solve by Green's functions methods in Section 3. Within this solution, a set of self-energies is required, but so far undetermined. Approximations to compute the self-energies are presented in Section 4, finalizing the theoretical framework. The results and a corresponding discussion are presented in Section 5. A summary is given in Section 6.

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2. Model

For the above described systems we introduce the following Hamiltonian H:

$$H = H_s + H_f + H_{f(U)} + H_{sf} + H_V. \tag{1}$$

The kinetic energy of the conduction band electrons is concerned by H_s:

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

where $c_{i\sigma}^\dagger$ ($c_{i\sigma}$) are creation (annihilation) operators of conduction band (s-)electrons at lattice site $\boldsymbol{R}_i$ with spin σ, respectively. The chemical potential is denoted by μ, and T_{ij} are the usual hopping matrix elements, which are related to the Bloch energies $\epsilon(\boldsymbol{k})$ via

$$T_{ij} = \frac{1}{N} \sum_{\boldsymbol{k}} \epsilon(\boldsymbol{k}) e^{-i\boldsymbol{k}(\boldsymbol{R}_i - \boldsymbol{R}_j)}. \tag{3}$$

For low band occupations, a Coulomb interaction between s-electrons can be neglected safely.

Assuming a non-degenerate f-level E_f, the one-particle energy of the localized f-electrons with creation (annihilation) operators $f_{i\sigma}^\dagger$ ($f_{i\sigma}$) is represented by

$$H_f = \sum_{i,\sigma} (E_f - \mu) f_{i\sigma}^\dagger f_{i\sigma}. \tag{4}$$

In order to disfavor double occupancies, a large intraatomic Coulomb interaction is taken into account by

$$H_{f(U)} = \frac{U_f}{2} \sum_{i,\sigma} n_{fi\sigma} n_{fi-\sigma}, \tag{5}$$

where $n_{fi\sigma} = f_{i\sigma}^\dagger f_{i\sigma}$ is the number operator. Since the 4f-wave functions' overlap is negligible, a direct f-electron exchange is fairly small and hence not part of the Hamiltonian (1). Note, moreover, that Eqs. (4) and (5) are only valid for systems with a total spin of 1/2, thus being an approximation in modeling the 4f-levels. This simplification is done in order to keep mathematics on a tractable level, whereas the underlying physical effects are not expected to be affected significantly.

To allow for magnetic ordering, interactions between conduction band and f-electrons must be considered. The s–f-exchange is commonly described by an intraatomic spin–spin-coupling:

$$H_{sf} = -J \sum_i \boldsymbol{\sigma}_i \cdot \boldsymbol{S}_i, \tag{6}$$

where $\boldsymbol{\sigma}_i$ and $\boldsymbol{S}_i$ represent the conduction and f-electron spin operators at lattice site $\boldsymbol{R}_i$, respectively. The coupling strength is given by J, where positive (negative) values of J stand for a preferred (anti)parallel alignment of s- and f-electron magnetic moments.

The last part of the Hamiltonian in Eq. (1):

$$H_V = V \sum_{i,\sigma} (c_{i\sigma}^\dagger f_{i\sigma} + f_{i\sigma}^\dagger c_{i\sigma}), \tag{7}$$

allows for (virtual) electronic transitions from the 4f-level into the conduction band and vice versa, where V is the hybridization strength. While the s–f-exchange, Eq. (6), originates from the non-classical part of the Coulomb interaction between the conduction electrons and the localized f-electrons, the hybridization term, Eq. (7), is a one-particle scattering term which mimics, in the most simple way, the hybridization between the respective bands. While concrete values might be obtained by standard self-consistent band-structure or constraint RPA calculations, there is a priori no reason why those couplings should not co-exist.

As a consequence of the hybridization, the respective average occupation numbers

$$n_{s\sigma} = \langle c_{i\sigma}^\dagger c_{i\sigma} \rangle, \quad n_{f\sigma} = \langle f_{i\sigma}^\dagger f_{i\sigma} \rangle,$$
$$n_s = \sum_\sigma n_{s\sigma}, \quad n_f = \sum_\sigma n_{f\sigma}, \tag{8}$$

are not necessarily constants when varying a model parameter. Contrary, the total occupation number

$$n = n_s + n_f \tag{9}$$

is kept fixed at a constant value by proper adjustment of the chemical potential μ. Since needed for later purposes, we also introduce the dimensionless magnetizations $m_{s,f}$:

$$m_s = \sum_\sigma z_\sigma n_{s\sigma},$$
$$m_f = \sum_\sigma z_\sigma n_{f\sigma}, \tag{10}$$

using $z_\sigma = \delta_{\sigma\uparrow} - \delta_{\sigma\downarrow}$.

To fix some of the model parameters, we set the on-site energies of the conduction electrons to $T_{ii} = 0$ eV. Hence, the conduction band center of gravity defines the energy zero. Energy units are essentially fixed by choosing $W = 1$ eV for the width of the conduction band. Furthermore, we assume a strong Hubbard interaction $U_f \gg W$.

The one-particle energy of the 4f-levels, E_f, is a parameter that decisively affects the physics of the model. A stable local magnetic moment on the f-levels is formed in the limit $E_f \ll 0$ and $E_f + U_f \gg 0$ where each f-level is exactly occupied by one electron. Here, however, we also consider a parameter regime where E_f comes close to the lower edge of the conduction band. This implies the presence of charge fluctuations and thereby the hybridization term, Eq. (7) is activated which eventually generates an effective f–f magnetic exchange. Note that this implies that the Schrieffer–Wolff transformation [17] does not apply to this parameter regime. Consequently, $H_f + H_{f(U)} + H_V$ cannot be replaced by a local antiferromagnetic exchange which would trivially compete (or cooperate) with H_{sf}.

It is worth mentioning that the model (1) reduces to the conventional periodic Anderson model if the s–f-exchange, Eq. (6), is neglected ($J = 0$). On the other hand, for $V = 0$, the model essentially reduces to a fermionized variant of the famous Kondo-lattice model.

3. Theory

In order to determine $n_{s,f}$ and $m_{s,f}$ self-consistently, we attack the many-body problem posed by the Hamiltonian (1) using Green's function methods. With the definition of the pure one-particle s- and f-Green's functions:

$$G_{\boldsymbol{k}\sigma}(E) = \frac{1}{N} \sum_{i,j} e^{i\boldsymbol{k}(\boldsymbol{R}_i - \boldsymbol{R}_j)} \left\langle\!\left\langle c_{i\sigma}; c_{j\sigma}^\dagger \right\rangle\!\right\rangle_E,$$
$$F_{\boldsymbol{k}\sigma}(E) = \frac{1}{N} \sum_{i,j} e^{i\boldsymbol{k}(\boldsymbol{R}_i - \boldsymbol{R}_j)} \left\langle\!\left\langle f_{i\sigma}; f_{j\sigma}^\dagger \right\rangle\!\right\rangle_E, \tag{11}$$

as well as with two mixed Green's functions:

$$P_{\boldsymbol{k}\sigma}^{(1)}(E) = \frac{1}{N} \sum_{i,j} e^{i\boldsymbol{k}(\boldsymbol{R}_i - \boldsymbol{R}_j)} \left\langle\!\left\langle c_{i\sigma}; f_{j\sigma}^\dagger \right\rangle\!\right\rangle_E,$$
$$P_{\boldsymbol{k}\sigma}^{(2)}(E) = \frac{1}{N} \sum_{i,j} e^{i\boldsymbol{k}(\boldsymbol{R}_i - \boldsymbol{R}_j)} \left\langle\!\left\langle f_{i\sigma}; c_{j\sigma}^\dagger \right\rangle\!\right\rangle_E, \tag{12}$$

all given in $\boldsymbol{k}$-space with N being the number of lattice sites, one easily obtains the following equation of motion: