Temperature-dependent electronic structure of Ni, Fe and Gd

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Abstract

We investigated the influence of electron correlations on the temperature dependence of the electronic structure of the prototypical ferromagnets Ni, Fe(bcc), and Gd using a many-body evaluation of a generalized model of magnetism. The single-particle energies are taken from an LSDA band structure calculation. The many-particle interactions are described by just two parameters, an intraband Coulomb interaction \( U \) and an interband exchange \( J \). The self-consistent model solution yields very realistic values for the \( T = 0 \) moments and the Curie temperatures. Typical differences as well as common features of the magnetism in Ni, Fe and Gd are worked out.

Fe, Ni on the one hand, and Gd on the other, are representatives of rather different classes of magnetism (band ferromagnets versus ‘local-moment’ Heisenberg ferromagnets). Nevertheless it should be possible to describe them by one and the same theoretical model. The single-particle part of our model Hamiltonian \( H = H_0 + H_1 \),

\[
H_0 = \sum_{i,m} T_i(m) c_{i,m}^+ c_{i,m} = \sum_{k,m} \varepsilon_m(k) c_{k,m}^+ c_{k,m},
\]

has the usual structure and is written in the conventional notation. The band index \( m \) runs over all those energy bands which are related to magnetism, either because they are producing it or because they are strongly influenced by it. In Fe, Ni only the 3\( d \) subbands are relevant, while in the case of Gd the extremely narrow 4\( f \) ‘bands’ and the broad (6\( s, 5d \)) conduction band must be taken into account. As to the Coulomb interaction \( H_1 \), essential aspects of ferromagnetism are sufficiently well accounted for by intra-atomic direct (\( U_{mn} \)) and exchange (\( J_{mn} \)) terms only. They can be cast in the following form:

\[
H_1 = H_U + H_J + H_U^{\text{eff}},
\]

(2)

where \( H_U \) is an intraband Coulomb interaction of Hubbard-type:

\[
H_U \equiv \frac{1}{2} \sum_{i,m} U_i n_{i,m} n_{i,m}^-, \quad (U_i = U_{mm} + J_{mm}).
\]

The second term is an interband exchange interaction,

which can be written as spin–spin interaction between electrons from different subbands:

\[
H_J = - \sum_{i,m} J_{mn} \sigma_{im} \cdot \sigma_{in},
\]

(4)

where \( \sigma_{im} \) is the electron spin operator. The third partial operator in (2) is a spin-independent direct Coulomb interaction between electrons from different subbands (\( U_{mn} = 0.5(U_{mm} - 0.5J_{mn}) \)):

\[
H_{U^{\text{eff}}} = \frac{1}{2} \sum_{i,m} U_{mn} n_{im} n_{in}; \quad n_{im} = \sum_{\sigma} n_{i,m,\sigma}.
\]

(5)

Because of the lack of any spin dependence, the operator \( H_{U^{\text{eff}}} \) will not exert a decisive influence on typical magnetic phenomena. We therefore neglect it in the cases of Fe, Ni and Gd. The interaction part \( H_1 \) contains only those interactions that are considered to be vital for reproducing the magnetic correlations and the temperature dependence of the electronic structure. For a quantitative comparison with experimental data, however, we have to incorporate the consequences of ‘all the other’ interactions by a proper renormalization of the single-particle energies. This is done by an LSDA band structure calculation, the results of which are taken for \( \varepsilon_m(k) \) in (1).

For an approximate solution of the complicated many-body problem we now exploit the decomposition (2) of the model Hamiltonian. The formal solution of the one-electron Green function,

\[
\langle \langle c_{k,m,\sigma}^+ c_{k,m,\sigma} \rangle \rangle_E = \hbar [ E - \varepsilon_m^{\text{LSDA}}(k) - \Sigma_{k,m,\sigma}(E) ]^{-1},
\]

(6)
is given in terms of the self-energy $\Sigma_{k\sigma}(E)$,

$$\Sigma_{k\sigma}(E) = M_{k\sigma}(E) + M_{k\sigma}^{(J)}(E),$$  

(7)

which decomposes into two partial contributions according to

$$\langle \langle [c_{k\sigma}, H_{U,J}]; c_{k\sigma}^+ \rangle \rangle_E = M_{k\sigma}^{(U,J)}(E) \langle \langle c_{k\sigma}, c_{k\sigma}^+ \rangle \rangle_E.$$  

(8)

We use these partial self-energies to define two ‘effective-medium’ Hamiltonians ($x, y = U, J$; $y \neq x$):

$$H_{\text{eff}}(x) = \sum_{k\sigma} \left( e_m^{\text{LSDA}}(k) + M_{k\sigma}^{(y)}(E) \right) c_{k\sigma}^+ c_{k\sigma} + H_x.$$  

(9)

These operators are energy-dependent and non-Hermitian, leading, however, to the same Green function (6) as the original Hamiltonian ($H$). For $x = U$ we have to solve an ‘effective Hubbard problem’. In the case of Gd the intraband Coulomb interaction can be neglected for the broad conduction bands. The coupling constants $U_m$ in (3) are therefore to be counted only when $m$ denotes one of the seven 4f levels. From the experiment [1] we read off $U_d(Gd) = 11.44$ eV. Because of the extreme flatness of the 4f dispersions the ‘Hubbard problem’ can be solved exactly. For the 3d subbands of Ni and Fe ($U_{2d}(Ni) = 6$ eV; $U_{2d}(Fe) = 1.8$ eV) the self-consistent moment method of Ref. [2] is used.

For $x = J$ one encounters the ‘sf problem’, which we approximate using the moment-conserving decoupling procedure of Ref. [3]. We assume for Ni and Fe that all pairs of 3d subbands have the same $J_{\text{max}}$ ($J(Ni) = 0.4$ eV; $J(Fe) = 0.2$ eV). In Gd only the interband exchange between 4f levels and conduction bands is important ($J_1 = 0.17$ eV for broad, ‘s-like’ subbands ($m = 1, 2$), $J_2 = 0.36$ eV for narrow, ‘d-like’ subbands ($m = 3-6$)).

Each of the self-energy parts, following from $H_{\text{eff}}(x)$ (9), depends on the respective other self-energy part, and furthermore, on certain thermodynamic expectation values, which, however, can be expressed by the ‘full’ Green function (6). For Fe and Ni the above model parameters have been fixed by the requirement that a self-consistent ferromagnetic solution is found with a phase transition of second order at $T_C$ and that $T = 0$ moment that is as realistic as possible. For Gd we fitted $J_1$ and $J_2$ to the itinerant electron contribution of 0.63 $\mu_B$ to the $T = 0$ moment.

The 3d spectrum of Fe and Ni consists of five non-degenerate subbands, which we denote $m = 1, 2, \ldots, 5$, as described in Ref. [4]. In Ni the first four are fully occupied, in Fe the first two. These bands are of course magnetically inactive. The Ni ferromagnetism is therefore exclusively due to the uppermost $m = 5$ subband, where the calculated $T_C = 635$ K [4] is very close to the experimental value (631 K). Correlation effects mediated mainly by $U$ provoke for the partially filled $m = 5$ band two different kinds of band splittings. One is of order $E$ and persists for all temperatures (‘Hubbard bands’!). Because $U = 6$ eV in Ni, the lower quasiparticle subband lies some 6 eV below the chemical potential $\mu$, being nothing other than the famous ‘Ni 6 eV satellite’. The other splitting leads to an additional spin asymmetry in the two resulting quasiparticle subbands, thus providing a finite magnetic moment. This spin asymmetry is removed when $T \to T_C$. In principle, the $m = 3, 4$ and 5 subbands of Fe behave like the Ni $m = 5$ subband. However, because of the smaller $U$, all the quasiparticle subbands overlap, thus avoiding a feature that deserves the notation ‘satellite’. On the other hand, a ‘non-collapsing’ exchange splitting appears above $T_C$ (Fig. 1). The theoretical $T_C = 1045$ K coincides with the experimental value.

The conduction bands of the rare-earth metal Gd are in principle magnetically inactive. The temperature dependence of the quasiparticle band structure is therefore induced by interband exchange with the ferromagnetic 4f system. The lower, relatively broad, ‘s-like’ ($m = 1, 2$) subbands exhibit a spin splitting that decreases with increasing temperature, collapsing for $T \to T_C$. Strikingly different behaviour is found for the flatter ‘d-like’ ($m = 3, 4, 5$) dispersions. They split even for $T > T_C$, and the spin depolarization with increasing temperature is due to a rearrangement of spectral weight rather than to a ‘Stoner-like’ collapse of the exchange splitting.

References