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Electronic structure and magnetic properties of correlated metals

A local self-consistent perturbation scheme

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Abstract. In the framework of LDA+DMFT (dynamical mean field theory) approach for realistic electronic structure calculations, a new perturbation scheme which combines the T-matrix and fluctuating exchange approximations is presented. This method is less computationally expensive than the numerically exact quantum Monte Carlo technique and give an adequate description of the electronic structure and exchange interactions in magnetic metals. We present a simple expression for the exchange interactions corresponding to the neglect of the vertex, corrections which becomes exact for the spin-wave stiffness in the local approximation. Electronic structure, correlation effects and exchange interactions for ferromagnetic nickel are discussed.

PACS. 71.10.-w Theories and models of many-electron systems – 71.15.-m Methods of electronic structure calculations

1 Introduction

Electronic structure and magnetic properties of iron-group metals have been a subject of great interest for a very long period (for review of early theories see [1–3]). Density functional (DF) theory in the form of the local spin density approximation (LSDA) or generalized gradient approximation (GGA), which which forms the basis of modern microscopic theory of solids, is faced with a series of difficulties when describing photoemission, thermoemission and other spectra of Fe and Ni as well as their finite-temperature magnetic properties (see [4–8] and Refs. therein). The electron correlation effects should be taken into account to solve these problems. Many attempts to include these effects in band structure calculations of transition metals are found in the literature [9–14]. Probably the most accurate and successful approach is the use of the dynamical mean-filed theory (DMFT, [15,16]) in the framework of so called LDA+DMFT approach [17,18] (for review see [19]), which adds many-body effects as an effective quantum impurity problem. The DMFT method has been applied to the magnetism of transition metals in references [8,20] within the numerically exact quantum Monte Carlo (QMC) scheme. Unfortunately, the using of the QMC technique is very cumbersome and expensive computationally; besides, the QMC method deals with the "truncated" two-indices interaction matrix instead of the complete four-indices one (see [20]). An alternative scheme was proposed in reference [7] based on a multiband spin-polarized generalization of the "fluctuating exchange" (FLEX) approximation by Bickers and Scalapino [21]. The original formulation of the FLEX approximation treats in an equal way both particle-hole (PH) and particle-particle (PP) channels. However, their roles in magnetism are completely different: the interaction of electrons with spin fluctuations in PH channel leads to the most relevant correlation effects [3] whereas PP processes are important for the renormalizations of the effective interactions in spirit of the T-matrix approach ("ladder approximation") by Galitskii [22] and Kanamori [23]. Therefore we used in reference [7] a "two-step" procedure when, at first, the bare matrix vertex is replaced by the T-matrix, and, secondly, PH channel processes with this effective interaction are taken into account explicitly. Note that the first attempt to combine the T-matrix and particle-hole correlations in relation with the problem of the magnetism of transition metals has been done by Liebsch [9]) but in a different way (introducing the particle-hole renormalization into the T-matrix which is opposite in this sense to the approach of Ref. [7]).

The latter ("two-step-FLEX") approximation gives, in general, qualitatively stasfactory description for the Hubbard model [24], as well as for ferromagnetic iron iron [7]; one can assume that it is reliable enough for systems with moderate correlations $U < W/2$ where U is the Hubbard

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on-site repulsion energy and W is the bandwidth. Replacing the bare Coulomb interaction by the T-matrix can be justified accurately, at least for spin-wave temperature region, both for the Hubbard model [25] and for the s-d exchange (spin-fermion) model [26]. However, the specific form of the approximation used in [7] can be improved further by taking into account the spin-dependence of the T-matrix. Here we present the formulation of the spin-polarized T-matrix-FLEX (SPTF) approximation and its application to the electronic structure of ferromagnetic nickel. It is worthwhile to stress that we use this perturbational approach *within* the framework of the DMFT scheme which is superior to the local approximation (neglecting the momentum dependence) in corresponding diagrams. We find a satisfactory description of correlation effects in nickel similar to in our previous QMC calculations [8] but with an essential gain in computational resources needed for the calculations.

2 Computational approach

We start with the general many-body Hamiltonian in the LDA+U scheme [27]:

$$\begin{aligned} H &= H_t + H_U \\ H_t &= \sum_{\lambda\lambda'\sigma} t_{\lambda\lambda'} c^+_{\lambda\sigma} c_{\lambda'\sigma} \\ H_U &= \frac{1}{2} \sum_{\{\lambda_i\}\sigma\sigma'} \langle \lambda_1 \lambda_2 \left| v \right| \lambda_1' \lambda_2' \rangle \, c^+_{\lambda_1\sigma} c^+_{\lambda_2\sigma'} c_{\lambda_2'\sigma'} c_{\lambda_1'\sigma}, \end{aligned} \tag{1}$$

where $\lambda = im$ are the site number (i) and orbital (m) quantum numbers, $\sigma = \uparrow, \downarrow$ is the spin projection, c^+, c are the Fermion creation and annihilation operators, H_t is the effective single-particle Hamiltonian from the LDA, corrected for the double-counting of average interactions among correlated electrons as will be described below, and the Coulomb matrix elements are defined in the standard way

$$\langle 12 \left| v \right| 34 \rangle = \int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{r}' \psi_1^*(\mathbf{r}) \psi_2^*(\mathbf{r}') v\left(\mathbf{r} - \mathbf{r}'\right) \psi_3(\mathbf{r}) \psi_4(\mathbf{r}'), \tag{2}$$

where we define for briefness $\lambda_1 \equiv 1$ etc. Following reference [22] we take into account the ladder (T-matrix) renormalization of the effective interaction:

$$\begin{aligned} \left\langle 13 \left| T^{\sigma\sigma'}(\mathrm{i}\Omega) \right| 24 \right\rangle &= \langle 13 \left| v \right| 24 \rangle - \frac{1}{\beta} \sum_{\omega} \sum_{5678} \langle 13 \left| v \right| 57 \rangle \\ &* G^{\sigma}_{56}(\mathrm{i}\omega) \, G^{\sigma'}_{78}(\mathrm{i}\Omega - \mathrm{i}\omega) \left\langle 68 \left| T^{\sigma\sigma'}(\mathrm{i}\Omega) \right| 24 \right\rangle, \end{aligned} \tag{3}$$

where $\omega = (2n+1)\pi T$ are the Matsubara frequencies for temperature $T \equiv \beta^{-1}$ ($n = 0, \pm 1, ...$). Further we rewrite the perturbation theory in terms of this effective interaction matrix.

At first, we take into account the "Hartree" and "Fock" diagrams with the replacement of the bare interaction by the T-matrix

$$\begin{aligned} \Sigma^{(TH)}_{12,\sigma}(\mathrm{i}\omega) &= \frac{1}{\beta} \sum_{\Omega} \sum_{34\sigma'} \left\langle 13 \left| T^{\sigma\sigma'}(\mathrm{i}\Omega) \right| 24 \right\rangle G^{\sigma'}_{43}(\mathrm{i}\Omega - \mathrm{i}\omega) \\ \Sigma^{(TF)}_{12,\sigma}(\mathrm{i}\omega) &= -\frac{1}{\beta} \sum_{\Omega} \sum_{34} \langle 14 \left| T^{\sigma\sigma}(\mathrm{i}\Omega) \right| 32 \rangle \, G^{\sigma}_{34}(\mathrm{i}\Omega - \mathrm{i}\omega). \end{aligned} \tag{4}$$

Note that $\Sigma^{(TH)} + \Sigma^{(TF)}$ contains exactly all the second-order contributions as can be easily seen from the corresponding Feynman diagrams. Now we have to consider the contribution of particle-hole excitations to the self-energy. Similar to [7] we will replace in the corresponding diagrams the bare interaction by the static limit of the T-matrix (as was already mentioned, it can be justified by the explicit calculation of the electron and magnon Green functions of a ferromagnet, at least, for spin-wave temperature region [25,26]). We improve the approximation [7] by taking into account the T-matrix spin dependence. When considering the particle-hole channel we replace in the Hamiltonian (1) $v \to T^{\sigma\sigma'}$ which is the solution of equation (3) at $\Omega = 0$. equation (4) is exact in the limit of low electron (or hole) density which is important for the criterion of magnetism, *e.g.*, in the case of nickel with its almost completely filled d-band.

Now we rewrite the effective Hamiltonian (1) with the replacement $\langle 12 \left| v \right| 34 \rangle$ by $\left\langle 12 \left| T^{\sigma\sigma'} \right| 34 \right\rangle$ in H_U. To consider the correlation effects due to PH channel we have to separate density (d) and magnetic (m) channels as in reference [21]

$$\begin{aligned} d_{12} &= \frac{1}{\sqrt{2}} \left(c^+_{1\uparrow} c_{2\uparrow} + c^+_{1\downarrow} c_{2\downarrow} \right) \\ m^0_{12} &= \frac{1}{\sqrt{2}} \left(c^+_{1\uparrow} c_{2\uparrow} - c^+_{1\downarrow} c_{2\downarrow} \right) \\ m^+_{12} &= c^+_{1\uparrow} c_{2\downarrow} \\ m^-_{12} &= c^+_{1\downarrow} c_{2\uparrow}. \end{aligned} \tag{5}$$

Then the interaction Hamiltonian can be rewritten in the following matrix form

$$\begin{aligned} H_U = \frac{1}{2} \mathrm{Tr} \Big(& D^+ * V^{\parallel} * D + m^+ * V^{\perp}_m * m^- \\ & + m^- * V^{\perp}_m * m^+ \Big) \end{aligned} \tag{6}$$

where $*$ means matrix multiplication with respect to the pairs of orbital indices, *e.g.*

$$\left(V^{\perp}_m * m^+ \right)_{11'} = \sum_{34} \left(V^{\perp}_m \right)_{11',22'} m^+_{22'},$$

the supervector D defined as

$$D = \left(d, m^0 \right), D^+ = \begin{pmatrix} d^+ \\ m^+_0 \end{pmatrix},$$

and the effective interactions have the following form:

$$\begin{aligned}
&\left(V_m^{\perp}\right)_{11',22'} = -\left\langle 12\left|T^{\uparrow\downarrow}\right|2'1'\right\rangle \\
&V^{\|} = \begin{pmatrix} V^{dd} & V^{dm} \\ V^{md} & V^{dd} \end{pmatrix} \\
&V^{dd}_{11',22'} = \frac{1}{2}\sum_{\sigma\sigma'}\left\langle 12\left|T^{\sigma\sigma'}\right|1'2'\right\rangle - \frac{1}{2}\sum_{\sigma}\left\langle 12\left|T^{\sigma\sigma}\right|2'1'\right\rangle \\
&V^{mm}_{11',22'} = \frac{1}{2}\sum_{\sigma\sigma'}\sigma\sigma'\left\langle 12\left|T^{\sigma\sigma'}\right|1'2'\right\rangle \\
&\quad -\frac{1}{2}\sum_{\sigma}\left\langle 12\left|T^{\sigma\sigma}\right|2'1'\right\rangle V^{dm}_{11',22'} = V^{md}_{22',11'} \\
&= \frac{1}{2}\Bigg[\left\langle 12\left|T^{\uparrow\uparrow}\right|1'2'\right\rangle - \left\langle 12\left|T^{\downarrow\downarrow}\right|1'2'\right\rangle - \left\langle 12\left|T^{\uparrow\downarrow}\right|1'2'\right\rangle \\
&+ \left\langle 12\left|T^{\downarrow\uparrow}\right|1'2'\right\rangle - \left\langle 12\left|T^{\uparrow\uparrow}\right|2'1'\right\rangle + \left\langle 12\left|T^{\downarrow\downarrow}\right|2'1'\right\rangle\Bigg]. \qquad (7)
\end{aligned}$$

To calculate the particle-hole (P-H) contribution to the electron self-energy we first have to write the expressions for the generalized susceptibilities, both transverse $\chi^{\perp}$ and longitudinal $\chi^{\|}$. The corresponding expressions are the same as in reference [7] but with another definition of the interaction vertices. One has

$$\chi^{+-}(\mathrm{i}\omega) = \left[1 + V_m^{\perp} * \Gamma^{\uparrow\downarrow}(\mathrm{i}\omega)\right]^{-1} * \Gamma^{\uparrow\downarrow}(\mathrm{i}\omega), \qquad (8)$$

where

$$\Gamma^{\sigma\sigma'}_{12,34}(\tau) = -G^{\sigma}_{23}(\tau)\, G^{\sigma'}_{41}(-\tau) \qquad (9)$$

is an "empty loop" susceptibility and $\Gamma(\mathrm{i}\omega)$ is its Fourier transform, τ is the imaginary time. The corresponding longitudinal susceptibility matrix has a more complicated form:

$$\chi^{\|}(\mathrm{i}\omega) = \left[1 + V^{\|} * \chi_0^{\|}(\mathrm{i}\omega)\right]^{-1} * \chi_0^{\|}(\mathrm{i}\omega), \qquad (10)$$

and the matrix of bare longitudinal susceptibility is

$$\chi_0^{\|} = \frac{1}{2}\begin{pmatrix} \Gamma^{\uparrow\uparrow} + \Gamma^{\downarrow\downarrow} & \Gamma^{\uparrow\uparrow} - \Gamma^{\downarrow\downarrow} \\ \Gamma^{\uparrow\uparrow} - \Gamma^{\downarrow\downarrow} & \Gamma^{\uparrow\uparrow} + \Gamma^{\downarrow\downarrow} \end{pmatrix}, \qquad (11)$$

in the dd-, dm^0-, m^0d-, and m^0m^0-channels ($d, m^0 = 1, 2$ in the supermatrix indices). An important feature of these equations is the coupling of longitudinal magnetic fluctuations and of density fluctuations. It is not present in the one-band Hubbard model due to the absence of the interaction of electrons with parallel spins. For this case equations (8, 10) coincides with the well-known result [28].

Now we can write the particle-hole contribution to the self-energy. Similar to reference [7] one has

$$\Sigma^{(ph)}_{12,\sigma}(\tau) = \sum_{34,\sigma'} W^{\sigma\sigma'}_{13,42}(\tau)\, G^{\sigma'}_{34}(\tau), \qquad (12)$$

with the P-H fluctuation potential matrix:

$$W^{\sigma\sigma'}(\mathrm{i}\omega) = \begin{bmatrix} W^{\uparrow\uparrow}(\mathrm{i}\omega) & W^{\perp}(\mathrm{i}\omega) \\ W^{\perp}(\mathrm{i}\omega) & W^{\downarrow\downarrow}(\mathrm{i}\omega) \end{bmatrix}, \qquad (13)$$

where the spin-dependent effective potentials are defined as

$$\begin{aligned}
W^{\uparrow\uparrow} &= \frac{1}{2}V^{\|} * \left[\chi^{\|} - \chi_0^{\|}\right] * V^{\|} \\
W^{\downarrow\downarrow} &= \frac{1}{2}V^{\|} * \left[\widetilde{\chi}^{\|} - \widetilde{\chi}_0^{\|}\right] * V^{\|} \\
W^{\uparrow\downarrow} &= V_m^{\perp} * \left[\chi^{+-} - \chi_0^{+-}\right] * V_m^{\perp} \\
W^{\downarrow\uparrow} &= V_m^{\perp} * \left[\chi^{-+} - \chi_0^{-+}\right] * V_m^{\perp} \qquad (14)
\end{aligned}$$

where $\widetilde{\chi}^{\|}, \widetilde{\chi}_0^{\|}$ differ from $\chi^{\|}, \chi_0^{\|}$ by the replacement of $\Gamma^{\uparrow\uparrow} \Leftrightarrow \Gamma^{\downarrow\downarrow}$ in equation (11). We have subtracted the second-order contributions since they have already been taken into account in equation (4).

Our final expression for the self energy is

$$\Sigma = \Sigma^{(TH)} + \Sigma^{(TF)} + \Sigma^{(PH)}. \qquad (15)$$

This formulation accurately takes into account accurately spin-polaron effects because of the interaction with magnetic fluctuations [25,29]. The energy dependence of the T-matrix which is important for the description of satellite effects in Ni [9], contains exact second-order terms in v and is rigorous (because of the first term) for almost filled or almost empty bands.

The FLEX approximation can in principle be directly applied to the crystal taking into account the momentum dependence of the self-energy, which would lead to very cumbersome calculations. To overcome this computational problem, we use as in reference [7] a local approximation to the self-energy, corresponding to combination of the SPTF approach presented above with the DMFT theory. The latter is considered as an *optimal* local approximation [15,16], therefore, this combined approach should be essentially more accurate than simple neglecting of momentum dependence in the corresponding diagrams of SPTF. The formal difference is that in a "canonical" version of FLEX-like approximations [21] the self-energy is written as a functional of exact Green functions, in the perturbation theory – as a functional of bare Green functions, and in the DMFT – as a functional of bare Green functions in an effective correlated medium.

The DMFT maps the many-body system onto a multi-orbital quantum impurity, *i.e.* a set of local degrees of freedom in a bath described by the Weiss field function $\mathcal{G}$. The impurity action (here $\mathbf{c}(\tau) = [c_{m\sigma}(\tau)]$ is a vector of Grassman variables) is given by:

$$\begin{aligned}
S_{\mathit{eff}} = &\int_0^{\beta} \mathrm{d}\tau \int_0^{\beta} \mathrm{d}\tau' \mathrm{Tr}[\mathbf{c}^{+}(\tau)\mathcal{G}^{-1}(\tau,\tau')\mathbf{c}(\tau')] \\
&+ \int_0^{\beta} \mathrm{d}\tau H_U\left[\mathbf{c}^{+}(\tau), \mathbf{c}(\tau)\right]. \qquad (16)
\end{aligned}$$

It describes the spin, orbital, energy and temperature dependent interactions of particular magnetic $3d$-atom with the rest of the crystal and is used to compute the local Greens function matrix:

$$\mathbf{G}_\sigma(\tau-\tau') = -\frac{1}{Z}\int D[\mathbf{c},\mathbf{c}^+]\mathrm{e}^{-S_{eff}}\mathbf{c}(\tau)\mathbf{c}^+(\tau') \qquad (17)$$

(Z is the partition function) and the impurity self energy $\mathcal{G}_\sigma^{-1}(\mathrm{i}\omega) - \mathbf{G}_\sigma^{-1}(\mathrm{i}\omega) = \Sigma_\sigma(\mathrm{i}\omega)$.

The Weiss field function $\mathcal{G}$ is required to obey the self consistency condition, which restores translational invariance to the impurity model description:

$$\mathbf{G}_\sigma(\mathrm{i}\omega) = \sum_{\mathbf{k}}\left[(\mathrm{i}\omega+\mu)\mathbf{1} - H(\mathbf{k}) - \Sigma_\sigma^{dc}(\mathrm{i}\omega)\right]^{-1} \qquad (18)$$

where μ is the chemical potential, $H(\mathbf{k})$ is the LDA Hamiltonian in an orthogonal basis. The local matrix Σ_σ^{dc} is the sum of two terms, the impurity self energy and a so-called "double counting" correction, E_{dc} which is meant to subtract the average electron-electron interactions already included in the LDA Hamiltonian. For metallic systems we propose the general form of dc-correction: $\Sigma_\sigma^{dc}(\mathrm{i}\omega) = \Sigma_\sigma(\mathrm{i}\omega) - \frac{1}{2}\mathrm{Tr}_\sigma\Sigma_\sigma(0)$ for the non-magnetic LDA Hamiltonian [8] and $\Sigma_\sigma^{dc}(\mathrm{i}\omega) = \Sigma_\sigma(\mathrm{i}\omega) - \Sigma_\sigma(0)$ for the magnetic LSDA Hamiltonian. This is motivated by the fact that the static part of the correlation effects are already well described in the density functional theory. Only the d-part of the self-energy is present in our calculations, therefore $\Sigma_\sigma^{dc} = 0$ for s- and p-states as well as for non-diagonal $d-s$, p contributions.

In spirit of the DMFT approach we have to use the Weiss function $\mathcal{G}_\sigma$ instead of G_σ in all expressions when calculating the self-energy on a given site. Similar to the one-band DMFT-perturbation scheme [30,24] we keep the static mean-field term in the bath Green functions: $\mathcal{G}_\sigma^{-1}(\mathrm{i}\omega) = \mathbf{G}_\sigma^{-1}(\mathrm{i}\omega) + \Sigma_\sigma(\mathrm{i}\omega) - \Sigma_\sigma(0)$.

3 Electronic structure of nickel

We have started from the non spin-polarized LDA or spin-polarized LSDA band structure of nickel within the TB-LMTO method [31] using a minimal s, p, d basis set and used numerical orthogonalization to find the $H(\mathbf{k})$ Hamiltonian in equation (18). We take into account of the Coulomb interactions between d-states only. The correct parameterization of the H_U part is indeed a serious problem. For example, first-principles estimations of average Coulomb interactions (U) [13,32] lead to an unreasonably large value of order of 5–6 eV in comparison with the value of the U-parameter in the range of 1–2 eV for iron [13] consistent with experiment. Semi-empirical analysis of the appropriate value [33] gives $U \simeq 3$ eV. It is shown in references [7,8] that an adequate description of a broad circle of properties of Fe and Ni in the LDA+DMFT scheme is possible when choosing $U \simeq 2-3$ eV. The difficulties with an *ab initio* determination of the correct value of U are connected with complicated screening problems, definitions of orthogonal orbitals in the crystal, and contributions of the intersite interactions. In the quasi-atomic (spherical) approximation the full U-matrix for the d-shell is determined by the three parameters U, J and δJ or equivalently by effective Slater integrals F^0, F^2 and F^4 [27]. For example, $U = F^0$, $J = (F^2 + F^4)/14$ and we use the simplest way of estimating δJ or F^4 keeping the ratio F^2/F^4 equal to its atomic value 0.625 [27].

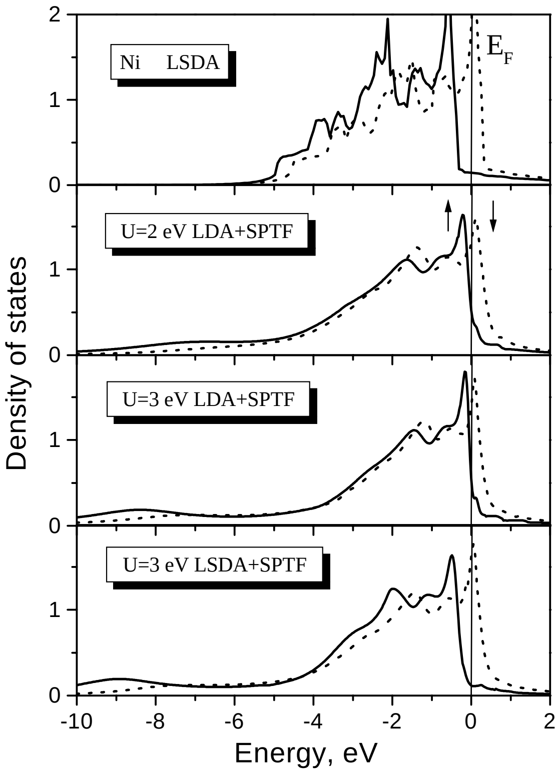

Fig. 1. Spin-up (full lines) and spin-down (dashed lines) density of d-states for ferromagnetic nickel in the LSDA and the LDA+SPTF (LSDA+SPTF) calculations for different average Coulomb interaction U with $J = 1$ eV and temperature $T = 200$ K.

Note that the value of intra-atomic (Hund) exchange interaction J is not sensitive to the screening and approximately equals 1 eV in different estimations [32]; here we have chosen $J = 1$ eV. For the most important parameter U, which defines the bare vertex matrix (Eq. (2)), we took the values $U = 2$ and 3 eV to check the dependence of the density of states (DOS) on U. To find DOS we applied a Pade approximant [34] for the analytical continuation of the Greens function from the Matsubara frequencies to the real energy axis. To find the self-consistent solution of the SPTF equations we used 1024 Matsubara frequencies and the FFT-scheme with the energy cut-off at 25 eV and temperature around 200 K. The sum over irreducible Brillouin zone has been made with 256 **k**-points.

Comparison of the LDA density of states and the SPTF calculation with DMFT self-consistency for the local self-energy matrix (Fig. 1) shows that the latter does reproduce the three most important characteristic features of correlation effects for nickel: 6 eV satellite, 30% narrowing of the d-bandwidth and 50% reduction of exchange splittings in comparison with the LSDA band structure [35–38]. For $U = 2$ eV the position of satellite is reproduced quite well, while for $U = 3$ eV it is shifted to the lower energies. Note that the LDA+DMFT consideration with the QMC solution of the effective impurity problem gives an adequate description of the electronic structure of Ni for the choice $U = 3$ eV [8]. In the SPTF approximation for the solution of impurity problem the choice $U = 2$ eV looks like the best one. The narrowing of the d-bandwidth in our calculations is reasonable for the both U-values. The non-magnetic LDA starting Hamiltonian is better than the LSDA one for correct description of the 50% reduction of the spin-splittings in nickel, while for magnetic LSDA Hamiltonian the the spin-splitting in the quasiparticle DOS remains approximately the same like in the LSDA results (Fig. 1). One can conclude that the LDA is better starting point than the LSDA for the account of the correlation effects; probably, it is important that exchange and correlation effects in magnetic splitting should be considered on an equal footing, in the same approximation. The local magnetic moment on the nickel atom is not very sensitive to U and is equal to 0.56 μ_B for $U = 2$ eV LDA+SPTF and 0.58 μ_B for $U = 3$ eV LSDA+SPTF calculations.

Another important correlation effect is an essential reduction of the spin polarization near the Fermi level in comparison with the LSDA calculations. This is connected with the spin-polaron effects because of the mixing of the spin-up and spin-down states [29]. They are taken into account in our scheme due to presence of the off-diagonal terms in the effective potential equation (13).

4 Exchange interactions in nickel

Calculating the variation of the thermodynamic potential with respect to small spin rotations with the use of the "local force theorem" an effective exchange interaction parameters can be found in the following form [20]

$$J_{ij} = -\mathrm{Tr}_{\omega L}\left(\Sigma_i^s G_{ij}^{\uparrow} \Sigma_j^s G_{ji}^{\downarrow}\right) \tag{19}$$

where $\Sigma_i^s = \frac{1}{2}\left(\Sigma_i^{\uparrow} - \Sigma_i^{\downarrow}\right)$. Correspondingly, the magnon dispersion relation $\omega_{\mathbf{q}}$ for a ferromagnet is defined by the formula

$$\omega_{\mathbf{q}} = \frac{4}{M}\left[J(0) - J(\mathbf{q})\right] \tag{20}$$

where M is the magnetic moment per unit cell, $J(\mathbf{q})$ is the Fourier transform of the exchange integrals defined by equation (19). The expression for the stiffness tensor $D_{\alpha\beta}$,

$$\omega_{\mathbf{q}} = D_{\alpha\beta} q_\alpha q_\beta, \ \ \mathbf{q} \to 0, \tag{21}$$

reads

$$D_{\alpha\beta} = -\frac{2}{M}\mathrm{Tr}_{\omega L}\sum_{\mathbf{k}}\left(\Sigma^s \frac{\partial G^{\uparrow}(\mathbf{k})}{\partial k_\alpha} \Sigma^s \frac{\partial G^{\downarrow}(\mathbf{k})}{\partial k_\beta}\right). \tag{22}$$

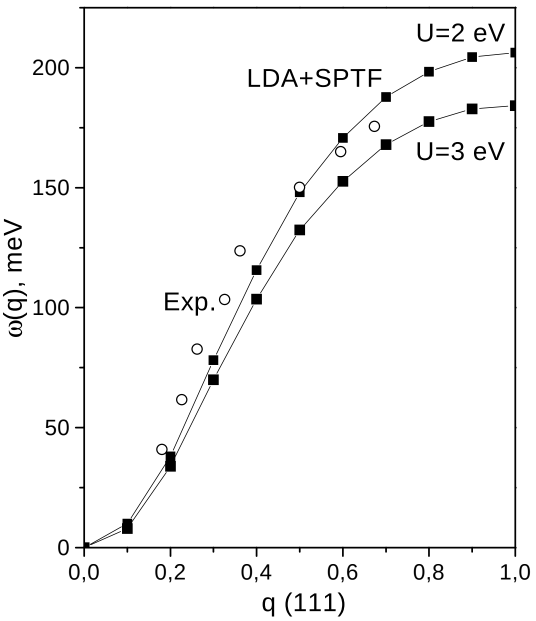

Fig. 2. Spin-wave spectrum for ferromagnetic nickel in LDA+SPTF scheme with different U and $J = 1$ eV in comparison with experimental magnon spectrum (Ref. [36]) in $\Gamma - L$ direction.

These results generalize the LSDA expressions of reference [39] to the case of correlated systems. One can show (see Appendix) that they can be derived using a standard diagrammatic approach under two assumptions: (i) the locality of the self-energy Σ (which is fulfilled in the DMFT) and (ii) the neglect of the vertex corrections. The expression (22) for the stiffness constant turns out to be exact in the framework of DMFT.

One should stress that it would be dangerous to justify the expressions (19, 22) by the local force theorem since the latter can be proven only for conserving (φ-derivable) approximations [20]. Unfortunately, we cannot demonstrate the φ-derivability of the SPTF approximation. However, as it is shown in the Appendix, this is not really necessary to obtain the expressions for exchange integrals and stiffness constant; it is neglecting of the vertex corrections that is used here essentially.

We have calculated the magnon spectrum for the optimal choice $U = 2$ eV and $J = 1$ eV using SPTF calculations with the non-magnetic LDA as a starting point. This is the choice of parameters which is optimal for the description of electron energy spectrum (see the previous section). The computational results for magnon dispersion are shown in Figure 2; the calculated spin-wave stiffness constant for Ni is found to be $D = 450$ meV/A^2 for $U = 2$ eV in an excellent agreement with the experimental value of 455 meV/A^2 [40]. It is interesting to note that the spin-wave spectrum for larger values of local Coulomb interactions: for $U = 3$ eV the exchange interactions become becomes softer (Fig. 2) in contrast with naive perturbative arguments. Generally, U should be compared with the width of the DOS peak near the Fermi level rather than with the total bandwidth [4] and, therefore, $U = 2 - 3$ eV corresponds already to the strongly correlated case. The simple approximation equation (19) for exchange interactions does not allow us to investigate the optical mode in magnon spectrum of nickel [43].

5 Conclusions

Here we have presented the results of new SPTF approximation in the framework of first-principle dynamical mean field theory (LDA+DMFT) for magnetic transition metals. This approximation combining the T-matrix and FLEX schemes gives a satisfactory description of both electronic and magnon spectra of Ni. In contrast with QMC method for the solution of effective impurity problem, this approach, being less rigorous, is not so time- and resource-consuming and allows to work with the most general rotationally invariant form of the Coulomb on-site interaction.

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Appendix: Exchange interactions and vertex corrections

In order to elucidate the approximation behind the expression for the exchange parameters (Eq. (19)), we consider the energy of a spiral magnetic configuration with the rigid rotation of the spinor-electron operators on site i by the polar angles θ_i and φ_i:

$$c_{im} \to U\left(\theta_i, \varphi_i\right) c_{im}$$

where

$$U\left(\theta, \varphi\right) = \begin{pmatrix} \cos\theta/2 & \sin\theta/2 \exp\left(-\mathrm{i}\varphi\right) \\ -\sin\theta/2 \exp\left(\mathrm{i}\varphi\right) & \cos\theta/2 \end{pmatrix},$$

assuming that $\theta_i = \text{const.}$ and $\varphi_i = \mathbf{q}\mathbf{R}_i$ where $\mathbf{R}_i$ is the site lattice vector. Since we take into account only on-site correlation effects the interaction term in the Hamiltonian is invariant under that transformation, and the change of the Hamiltonian is

$$\begin{aligned} \delta H &= \sum_{ij} \mathrm{Tr}_{L\sigma}\left[t_{ij} c_i^{+} \left(U_i^{+} U_j - 1\right) c_j\right] = \delta_1 H + \delta_2 H \\ \delta_1 H &= \sin^2 \frac{\theta}{2} \sum_{k} \mathrm{Tr}_{L\sigma}\left[\left(t\left(\mathbf{k}{+}\mathbf{q}\right) - t\left(\mathbf{k}\right)\right) c_{\mathbf{k}}^{+} c_{\mathbf{k}}\right] \\ \delta_2 H &= \frac{1}{2} \sin\theta \sum_{ij} \mathrm{Tr}_L\left[t_{ij} c_{i\downarrow}^{+} c_{j\uparrow}\right] \\ &\quad \times \left(\exp\left(\mathrm{i}\mathbf{q}\mathbf{R}_i\right) - \exp\left(\mathrm{i}\mathbf{q}\mathbf{R}_j\right)\right). \end{aligned} \tag{23}$$

Consider further the case of small θ, we can calculate the variation of the total energy to lowest order in θ which corresponds to the first order in $\delta_1 H$ and the second order in $\delta_2 H$:

$$\begin{aligned} \delta E = \frac{\theta^2}{4} \mathrm{Tr}_L \sum_{\mathbf{k}} &[t(\mathbf{k}+\mathbf{q}) - t(\mathbf{k})]\{n_{\mathbf{k}} \\ &+ \mathrm{Tr}_\omega[\gamma(k,q) G_\downarrow(k+q)[t(\mathbf{k}+\mathbf{q}) - t(\mathbf{k})] G_\uparrow(k)]\}, \end{aligned} \tag{24}$$

where $n_{\mathbf{k}} = \mathrm{Tr}_{L\sigma}\langle c_{\mathbf{k}}^{+} c_{\mathbf{k}}\rangle$, q, k are four-vectors with component $(\mathbf{q}, 0)$ and $(\mathbf{k}, \mathrm{i}\omega)$, γ is the three-leg vertex. Our main approximation is to neglect the vertex corrections ($\gamma = 1$). In this case the previous equation takes the following form:

$$\begin{aligned} \delta E = \frac{\theta^2}{4} &\mathrm{Tr}_{L\omega} \sum_{\mathbf{k}} [t(\mathbf{k}+\mathbf{q}) - t(\mathbf{k})] \\ * G_\downarrow(k+q)&[G_\downarrow^{-1}(k+q) - G_\uparrow^{-1}(k) + t(\mathbf{k}+\mathbf{q}) - t(\mathbf{k})] G_\uparrow(k). \end{aligned} \tag{25}$$

Using the following consequence of the Dyson equation:

$$\begin{aligned} t\left(\mathbf{k}{+}\mathbf{q}\right) - t\left(\mathbf{k}\right) = G_\uparrow^{-1}\left(k\right) - G_\downarrow^{-1}\left(k+q\right) \\ + \Sigma_\uparrow\left(E\right) - \Sigma_\downarrow\left(E\right) \end{aligned}$$

one can rewrite equation (25) in the form: $\delta E = \frac{\theta^2}{4}[J(0) - J(\mathbf{q})]$ with the exchange integrals corresponding to equation (19). We conclude that the expression for J_{ij} is accurate if the vertex corrections can be neglected. Note that the limit of small $\mathbf{q}$ this neglecting can be justified rigorously, provided that the self-energy and three-leg scalar vertex are local. This can be proven, *e.g.*, using the Ward-Takahashi identities [41]. Therefore, the expression for the stiffness constant of the ferromagnet (Eq. (22)) appears to be exact in the framework of DMFT [42].

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