

5 Magnetism: From Stoner to Hubbard

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1 Introduction

We will discuss a realistic approach to magnetism and electronic structure of correlated materials which takes into account dynamical many-body effects. The scheme combines the features of the itinerant electron theory (Stoner) of magnetic crystals with the localized-moment description (Heisenberg) in a unified spin-fluctuations approach for a generalized multi-orbital Hubbard model. Moreover, we analyze the calculation of effective exchange interaction-parameters based on the realistic electronic structure of correlated magnetic crystals.

2 From Stoner to Hubbard

We start to discuss the different models of magnetic materials (Fig. 1) with the simplest one-band Stoner Hamiltonian

$$H_s = \sum_{\mathbf{k}\sigma} (\varepsilon_{\mathbf{k}} + I\langle n_{-\sigma} \rangle) c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma}, \quad (1)$$

where $\varepsilon_{\mathbf{k}}$ is the energy band spectrum and I is a Stoner interatomic exchange parameter. In this case the temperature-dependent magnetic properties are related to the so-called Stoner excitations from the occupied "spin-up" to the unoccupied "spin-down" band. They reduce the magnitude of the magnetization, so that finally at the Curie point the itinerant system becomes a nonmagnetic metal.

If we compare the Stoner model with a standard Hubbard approach with the Hamiltonian

$$H_h = \sum_{ij\sigma} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} + \sum_i U n_{i\uparrow} n_{i\downarrow}, \quad (2)$$

where t_{ij} are the hopping parameters and U the characteristic Hubbard Coulomb interaction, then one can easily realize that the Stoner model is just a mean-field approximation to the Hubbard model. In the weakly correlated case the only possible magnetic excitations are spin-flips, and the corresponding energy is of the order $I \cdot M$ with $M = \langle n_\uparrow - n_\downarrow \rangle$ which is much larger than realistic Curie temperatures. In the opposite limit, the strongly correlated Hubbard model at half-filling [1], one can derive an effective Heisenberg model

$$H_e = - \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j. \quad (3)$$

The kinetic exchange interactions $J_{ij} = -2t_{ij}t_{ji}/U$ are of the order of magnetic (Néel) transition temperatures. The Heisenberg model describes well the magnetism of localized $4f$ -materials. In the case of transition metals, where both longitudinal and transverse magnetic fluctuations are important, the most appropriate model is the Hubbard Hamiltonian, Eq. (2).

We can discuss the different approaches to estimate the effective Heisenberg interactions, presented in Figs. 2 to 4. In Fig. 2 a simple two-site spin-model for the Heisenberg interaction with the singlet and triplet states is compared with the so-called Slater one-electron model for antiferromagnetic states, which results in an additional factor of two in the definition of the effective

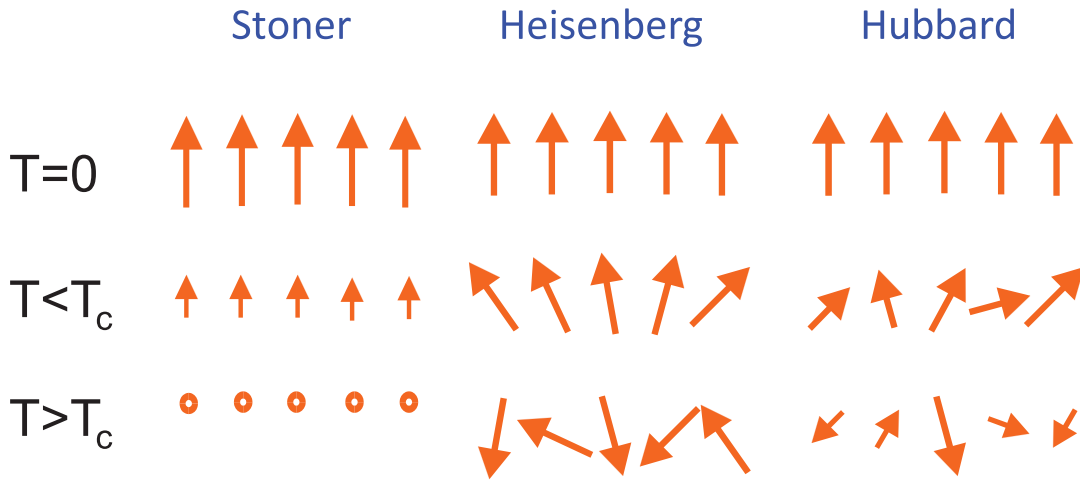


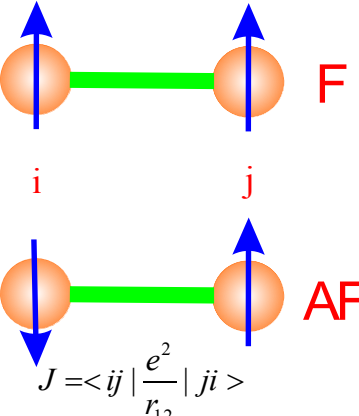
Fig. 1: Schematic view of different models of magnetism: Stoner model for itinerant weakly correlated electrons, Heisenberg model for localized magnetic moments and Hubbard model for the spin-fluctuations model of correlated electrons.

exchange interaction. In Fig. 3 the solution of the two-site Hubbard model for the many-body sector with one spin-up and one spin-down electron is shown, which results in the famous Anderson kinetic exchange interaction [1]. Finally, one can show that the mean-field solution of the Hubbard model with band energies modified via infinitesimal spin-rotations results in the same effective exchange interactions of a classical Heisenberg model (Fig. 4). In this case we used the so-called “local force theorem” which was originally formulated for density functional theory [2] and will be proven later (Sec. 6) using the Baym-Kadanoff approach. This theorem gives a simple recipe to obtain the total energy difference for a small perturbation of the charge- or spin-density as a change from the non-selfconsistent band energy for corresponding perturbation. In our case the energy of the infinitesimal spin-rotation in the two-site mean-field rotationally invariant Slater (spin-polarized LDA) model have been calculated and compared with the corresponding classical Heisenberg model (Fig. 4). We can see that both schemes give exactly the same effective exchange parameter.

3 From LDA to DMFT

The calculation of thermodynamic properties and excitation spectra of different magnetic materials is one of the most important problems of the microscopic theory of magnetism. We introduce a general functional approach which will cover density-functional theory (DFT), dynamical mean-field theory (DMFT) and Baym-Kadanoff (BK) theories [3]. Let us start from the full many-body Hamiltonian describing the electrons moving in the periodic external potential $V(\mathbf{r})$ of the ions with chemical potential μ and interacting via the Coulomb law $U(\mathbf{r}-\mathbf{r}') = 1/|\mathbf{r}-\mathbf{r}'|$. We use atomic units $\hbar = m = e = 1$. In the field-operator representation the Hamiltonian has

Magnetism of H₂: Heisenberg vs. Slater



Exchange interaction:

$$H_{ex} = -2J_{ij} \vec{S}_i \cdot \vec{S}_j$$

$$J = \frac{1}{2}(E_S - E_T) = E_{AF} - E_F$$

$$E_F = |\uparrow\uparrow\rangle = E_T$$

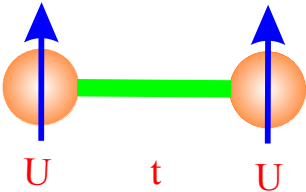
$$E_{AF} = |\uparrow\downarrow\rangle = \frac{1}{2}(E_S + E_T)$$

$$\psi_T = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle); \quad |\uparrow\uparrow\rangle; \quad |\downarrow\downarrow\rangle$$

$$\psi_S = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$$

Fig. 2: Exchange interaction in a two-site Heisenberg model.

Exchange interactions: Anderson



$$H = t \sum_{ij=1,2} c_{i\sigma}^\dagger c_{j\sigma} + U \sum_{i=1,2} n_{i\uparrow} n_{i\downarrow}$$

ED in subspace
 $N_- = 1, N_+ = 1$

$U \gg t$

Anderson kinetic exchange

$$E_T = 0$$

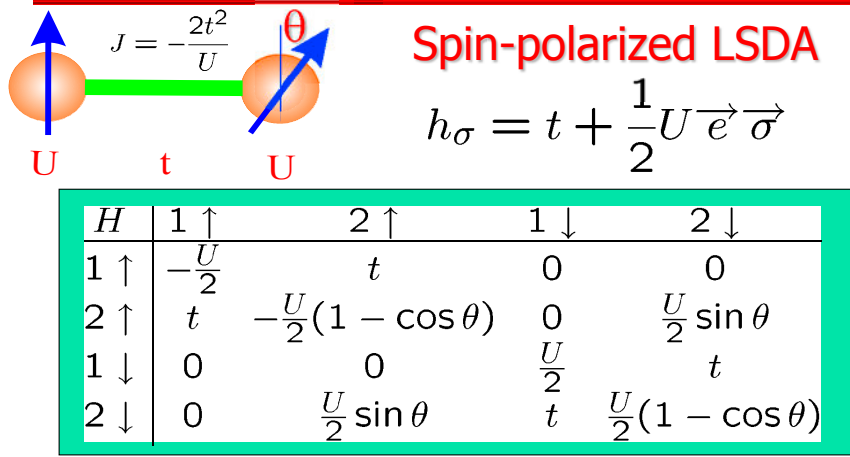
$$E_S = \frac{U}{2} \left(1 - \sqrt{1 + \frac{16t^2}{U}} \right) \approx -\frac{4t^2}{U}$$

$$J = -\frac{2t^2}{U}$$

N	i	j	H	1	2	3	4
1	↑	↓	1	0	0	t	t
2	↓	↑	2	0	0	t	t
3	↑↓	0	3	t	t	U	0
4	0	↑↓	4	t	t	0	U

Fig. 3: Exchange interaction in the two-site Hubbard model.

Exchange: Local force approach



Spectrum: $\epsilon_i = \pm \frac{1}{2} \sqrt{4t^2 + U^2 \pm Ut \sqrt{2(1 \mp \cos \theta)}}$

Exchange energy: $E_x(\theta) \approx \frac{t^2}{U} \cos \theta = -2JS^2 \vec{e}_i \cdot \vec{e}_j = -\frac{1}{2}J \cos \theta$

Fig. 4: Exchange interaction in the two-site mean-field rotationally invariant Slater model.

the form

$$\begin{aligned}
 H = & \sum_{\sigma} \int d\mathbf{r} \hat{\psi}_{\sigma}^{\dagger}(\mathbf{r}) \left[-\frac{1}{2} \nabla^2 + V(\mathbf{r}) - \mu \right] \hat{\psi}_{\sigma}(\mathbf{r}) \\
 & + \frac{1}{2} \sum_{\sigma\sigma'} \int d\mathbf{r} \int d\mathbf{r}' \hat{\psi}_{\sigma}^{\dagger}(\mathbf{r}) \hat{\psi}_{\sigma'}^{\dagger}(\mathbf{r}') U(\mathbf{r} - \mathbf{r}') \hat{\psi}_{\sigma'}(\mathbf{r}') \hat{\psi}_{\sigma}(\mathbf{r}).
 \end{aligned} \tag{4}$$

We can always use a single-particle orthonormal basis set $\phi_n(\mathbf{r})$, in solids for example Wannier orbitals with a full set of quantum numbers, e.g., site, orbital and spin index $n = (i, m, \sigma)$, and expand the fields in creation and annihilation operators

$$\begin{aligned}
 \hat{\psi}(\mathbf{r}) &= \sum_n \phi_n(\mathbf{r}) \hat{c}_n \\
 \hat{\psi}^{\dagger}(\mathbf{r}) &= \sum_n \phi_n^*(\mathbf{r}) \hat{c}_n^{\dagger}
 \end{aligned} \tag{5}$$

Going from fermionic operators to the Grassmann variables $\{c_n^*, c_n\}$ we can write the functional integral representation for the partition function of the many-body Hamiltonian in the imaginary time domain using the Euclidean action S

$$Z = \int \mathcal{D}[c^*, c] e^{-S} \tag{6}$$

$$S = \sum_{12} c_1^* (\partial_{\tau} + t_{12}) c_2 + \frac{1}{2} \sum_{1234} c_1^* c_2^* U_{1234} c_4 c_3, \tag{7}$$

where the one- and two-electron matrix elements are defined as

$$\begin{aligned} t_{12} &= \int d\mathbf{r} \phi_1^*(\mathbf{r}) \left[-\frac{1}{2} \nabla^2 + V(\mathbf{r}) - \mu \right] \phi_2(\mathbf{r}) \\ U_{1234} &= \int d\mathbf{r} \int d\mathbf{r}' \phi_1^*(\mathbf{r}) \phi_2^*(\mathbf{r}') U(\mathbf{r} - \mathbf{r}') \phi_3(\mathbf{r}) \phi_4(\mathbf{r}'). \end{aligned} \quad (8)$$

and we use the following short definition of the sum: $\sum_1 \dots \equiv \sum_{im} \int d\tau \dots$

The one-electron Green function is defined via the simplest non-zero correlation function for fermions

$$G_{12} = -\langle c_1 c_2^* \rangle_S = -\frac{1}{Z} \int \mathcal{D}[c^*, c] c_1 c_2^* \exp(-S) \quad (9)$$

and gives all information on the spin-dependent electronic structure of correlated materials.

The main difficulties of strongly interacting electronic systems are related to the fact that the higher-order correlation functions do not separate into a product of lower-order correlation functions. For example the two-particle Green function or generalized susceptibility (X) is defined in the following way [4]

$$X_{1234} = \langle c_1 c_2 c_3^* c_4^* \rangle_S = \frac{1}{Z} \int \mathcal{D}[c^*, c] c_1 c_2 c_3^* c_4^* \exp(-S), \quad (10)$$

and can be expressed as a simple "non-interacting" part and a connected correlated contribution through the exact Green function and the full vertex function Γ_{1234} [5]

$$X_{1234} = G_{14} G_{23} - G_{13} G_{24} + \sum_{1'2'3'4'} G_{11'} G_{22'} \Gamma_{1'2'3'4'} G_{3'3} G_{4'4}. \quad (11)$$

In principle, the spin-dependent part of the two-particle correlation function or generalized magnetic susceptibility contains all information on the magnetic properties of solids.

Modern computational material science is based on the density-functional (DFT) approach [6]. It is a common practice to use this scheme not only for the total energy calculations and related quantities such as charge and spin densities, but also for different spectral characteristics. Sometimes the agreement of the computational results with the experimental data is very impressive, despite the absence of a reliable theoretical background. In principle, the energies of Kohn-Sham quasiparticles [6] which are calculated in standard band theory are just auxiliary quantities for the total energy calculation.

The DFT functional is defined in the following way: the Kohn-Sham potential $V_{KS} = V_{ext} + V_H + V_{xc}$ plays the role of the effective one-electron potential, where all exchange-correlations effects V_{xc} are taken into account. In this case, V_{ext} is the external potential and V_H is the Hartree potential. In principle the exchange-correlation potential V_{xc} is known only for the homogeneous electron gas [8]. Therefore in practical applications one uses the so-called local-density approximation (LDA) to DFT. The total-energy functional reads

$$E_{tot}[n] = T_0[n] + V_{ext}[n] + V_H[n] + V_{xc}[n] \quad (12)$$

where T_0 is the kinetic energy of the non-interacting systems. Finally, if we define the total electron density as

$$n(\mathbf{r}) = \sum_i \phi_i^*(\mathbf{r}) \phi_i(\mathbf{r}) \quad (13)$$

the local-density approximation to the DFT reads

$$T_0[n] + V_{ext}[n] = \sum_i \int d\mathbf{r} \phi_i^*(\mathbf{r}) \left[-\frac{1}{2} \nabla^2 + V_{ext}(\mathbf{r}) - \mu \right] \phi_i(\mathbf{r}) \quad (14)$$

$$E_H[n] = \frac{1}{2} \int d\mathbf{r} n(\mathbf{r}) U(\mathbf{r} - \mathbf{r}') n(\mathbf{r}') \quad (15)$$

$$E_{xc}[n] = \int d\mathbf{r} n(\mathbf{r}) \varepsilon_{xc}(n(\mathbf{r})) \quad (16)$$

where $\varepsilon(n)$ is exchange correlation density for the homogeneous electron gas, which has been calculated with quantum Monte Carlo (QMC) [8]. The variational principle leads to the Kohn-Sham effective one-electron equation (in atomic units)

$$\left[-\frac{1}{2} \nabla^2 + V_{ext}(\mathbf{r}) + V_H + V_{xc} \right] \phi_k(\mathbf{r}) = \varepsilon_k \phi_k(\mathbf{r}). \quad (17)$$

In the one-band case the back Fourier transform of $\varepsilon_{\mathbf{k}}$ will give an effective Kohn-Sham hopping parameter $t(\mathbf{R})$ to t_{ij} , where i, j are the lattice indices. For the realistic multi-orbital case one can use the efficient first-principle Wannier-function parameterization of the energy bands to get the multi orbital (m) hopping matrix elements $t_{im,jm'}^{\text{LDA}}$, which will be used in the magnetic many-body formalism.

In the DFT scheme we lose information about the non-equal time Green function, which gives the single particle excitation spectrum as well as the $\mathbf{k}$ -dependence of the spectral function, and restrict ourselves only to the ground state energy of the many-electron system. Moreover, we also lose information about all collective excitations in solids, such as plasmons or magnons, which can be obtained from the generalized susceptibility.

Despite all achievements of the quantitative electronic structure theory, the list of difficulties and shortcomings is growing, especially when considering the magnetic d - and f -electron systems. In a number of cases the theory appears even *qualitatively* inadequate. First, the DFT scheme cannot describe correctly the phenomenon of “Mott insulators” [7], as was first observed by Terakura *et al.* [9] in their attempt of calculating the electronic structure of $3d$ -metal oxides. Later we faced similar problems in field of high- T_c superconductors [10] and other compounds [11]. The Ce- and U- based “heavy fermion” compounds such as CeCu_6 , UPt_3 , etc, are other “hot-spots”: normally the calculated effective masses are orders of magnitude smaller than what is experimentally observed [12]. Even for the pure $3d$ -metals some qualitative differences between theory and experiment exists. For example, there are at least three difficulties with the photoelectron spectra of ferromagnetic nickel [13]: (i) the measured width of the occupied part of the d -band is 30% narrower than calculated (ii) the spin-splitting is half of what is predicted by LDA and (iii) the band structure cannot describe the famous 6 eV satellite. Calculations for

paramagnetic spin-disordered states [14] lead to the conclusion that Ni has no local magnetic moments above the Curie temperature T_C , in clear contradiction with experimental results [15]. For iron, standard band theory cannot explain the data about spin polarization of thermionic electrons [16–18] and some features of angle-resolved photoemission spectra [18–20]. All these difficulties raise questions about the DFT approach: what is the “electron spectrum” that we really calculate and how can we improve the electronic theory for magnetic d - and f -systems? It was understood many years ago that all these problems are connected with the inadequate description of many-body effects in DFT calculations of the excitation spectra. Methods such as GW [21] and simplified $LDA+U$ [11] have been proposed to improve the situation. These methods are very useful for the description of antiferromagnetic transition-metal oxides as Mott insulators [11]. However, one should note that $LDA+U$ is just a mean-field approximation and cannot describe *correlation* effects which are, by definition, the many-body effects *beyond* Hartree-Fock. For example, in these approaches one needs spin- or orbital-ordering to describe the Mott insulator and it is impossible to describe correctly the electronic structure of NiO or MnO in the paramagnetic phase. At the same time, the magnetic ordering should not be important for the basic physics of Mott insulators [7]. All the “Hartree-Fock-like” approaches fail to describe the renormalization of the effective mass in the heavy fermion systems. There are also many problems concerning the electronic structure and itinerant magnetism of $3d$ metals as described above. Thus, one needs some practical ways of incorporating *correlation* effects in the electronic structure of solids.

In principle, there are two ways to include them into DFT calculations. The first uses a *time-dependent* DFT formalism which can guarantee, in principle, an opportunity to calculate exact response functions [22], in the same sense as the Hohenberg-Kohn theorem guarantees the total energy in usual “static” DFT [6]. However, all the expressions for this time-dependent non-local DFT in real calculations are based on RPA-like approximations which do not give a satisfactory description of really highly correlated systems. They are excellent for investigating the plasmon spectrum of aluminum, but not for understanding the nature of high- T_c superconductivity or heavy fermion behavior. Another way is to use an “alternative” many-body theory developed in the 50s by Gell-Mann and Brueckner, Galitskii and Migdal, Beliaev and many others in terms of the Green functions rather than the electron density [23]. We try to formulate such a computational approach as a generalization of $LDA+U$ scheme, the so-called “ $LDA+DMFT$ ” method. The main difference between $LDA+DMFT$ and $LDA+U$ is that in the former dynamical fluctuations, the real correlation effects, are accounted for by a local but energy dependent self-energy $\Sigma(\omega)$.

A comparison of the standard DFT theory in the local-density approximation (LDA) and the $LDA+DMFT$ approach is represented in table I. First of all, LDA is based on the Hohenberg-Kohn theorem stating that the total energy E_{tot} is a functional of charge (and spin) densities, while the $LDA+DMFT$ scheme considers the thermodynamic potential Ω as a functional of exact one-particle Green functions. This approach in many-particle theory has been introduced in the works by Luttinger and Ward [24] and Baym and Kadanoff [25]. The Green function in $LDA+DMFT$ plays the same role as the density matrix in LDA. We stress the dynamical

Table 1: *Comparison of LDA and realistic DMFT schemes*

LDA	LDA+DMFT
Density functional	Baym-Kadanoff functional
Density $\rho(\mathbf{r})$	Green-Function $G(\mathbf{r}, \mathbf{r}', \omega)$
Potential $V_{xc}(\mathbf{r})$	Self-energy $\Sigma_i(\omega)$
$E_{tot} = E_{sp} - E_{dc}$	$\Omega = \Omega_{sp} - \Omega_{dc}$
$E_{sp} = \sum_{k < k_F} \varepsilon_k$	$\Omega_{sp} = -\text{Tr} \ln[-G^{-1}]$
$E_{dc} = E_H + \int \rho V_{xc} d\mathbf{r} - E_{xc}$	$\Omega_{dc} = \text{Tr} \Sigma G - \Phi_{LW}$

nature of the correlation effects that are taken into account in the LDA+DMFT approach since the density in the LDA is just the static limit of the local Green function. Further, the self-energy Σ is treated analogously to the exchange-correlation potential, the local approximation for Σ , which is assumed to be energy-dependent but not momentum-dependent, corresponds to the local approximation for V_{xc} . In both formalisms the thermodynamic potential can be represented in a “single-particle” form, Ω_{sp} minus the contributions of the “doubly counted terms”, Ω_{dc} . This will be important for the consideration of the so-called “local force theorem” and the computation of magnetic interaction parameters. The single-particle contribution to the thermodynamic potential in the LDA+DMFT would have the same form as in the LDA if we were taking into account only the poles of the Green function and neglected the quasiparticle damping. However, even then the quasiparticle energies are not quite the same since the poles of the Green functions do not coincide, generally speaking, with the “Kohn-Sham” energies. The quantity Φ_{LW} is the Luttinger-Ward generating functional for the self energy, or the sum of all the skeleton diagrams without free legs [24].

The difficulty with finite temperature effects is one of the main shortcomings of a standard DFT formalism. In all realistic calculations the temperature is included in the Fermi distribution functions and in the lattice constants via the thermal expansion [26]. At the same time, for the itinerant electron magnets the temperature effects connected with the “Bose” degrees of freedom due to spin waves and paramagnons are much more important [27]. In principle, these effects could be taken into account in DFT via the temperature dependence of the exchange-correlation potential, the corresponding terms being nonlocal. It is not easy to propose an adequate expression for such temperature-dependent non-local potential. One of the first attempts in this direction is based on simple RPA-like considerations [28]. On the other hand, in LDA+DMFT-type scheme all calculations are naturally carried out for finite temperatures by using Matsubara frequencies, as is usual in many-body theory [23].

The main assumption of the LDA+DMFT approach is the importance of only intra-site “Hubbard correlations” with the local approximation for the self-energy. It is worthwhile to stress a

difference of this kind of locality from the locality in DFT theory. In the latter, the local approximation means that the exchange-correlation energy is calculated for the homogeneous electron gas [8]. It is known from exact QMC calculations that the correlation effects lead to some instabilities of the state of homogeneous electron gas (magnetism, charge ordering, etc) only for electron densities which are orders of magnitude smaller than ones typical for real metals (the critical values of the parameter r_s are of order of hundred in comparison with the “normal” range 2-6 for metals). At the same time, magnetism and charge ordering are rather usual for real compounds with the d - and f -elements. It thus seems that the “atomic-like” features of d - and f -states are of crucial importance to describe the correlation effects in real compounds. Only these features are taken into account in the Hubbard-like terms for the d - or f -states in LDA+DMFT approach. Therefore one can view the LDA+DMFT as the simplest way for quantitative considerations of correlation effects in transition metals and their compounds, based on the LDA description for all non-correlated electrons in the systems.

The investigation of correlation effects in the electronic structure and magnetism of iron-group metals is still far from having found the final picture and attracts continuous interest (see, e.g., [14, 29–31] and references therein). Despite many attempts, the situation is still unclear both theoretically and experimentally. For example, there is no agreement on the presence of a 5 eV satellite in the photoemission spectrum of iron [19, 20], and on the existence of local spin splitting above the Curie temperature in nickel [32]. From the theoretical point of view, different approaches such as second-order perturbation theory [30, 33], the T -matrix approximation [29, 34], the three-body Faddeev approximation [35], and the moment expansion method [36] were used. Unfortunately, the range applicability of these schemes is not clear. Here we present the LDA+DMFT approach [18, 37, 38] which is based on the combination of standard band-theory techniques (LDA) with dynamical mean-field theory (DMFT) [39].

4 Realistic DMFT scheme

In the LDA+DMFT approach we consider the renormalisation of the “bare” LDA energy or electron hopping due to correlation effects. Of course the t_{ij} contain already some part of the correlation effects but only those which may be considered in the local density approximation. The most important “rest” in strongly correlated system is the correlations of the Hubbard type [41] due to the intra-site Coulomb repulsion. Therefore we start from the general form of the LDA+DMFT Hamiltonian

$$H = \sum_{ij\sigma\{m\}} t_{im_1,jm_2}^{\text{LDA}} c_{im_1\sigma}^\dagger c_{jm_2\sigma} + \frac{1}{2} \sum_{i\{\sigma m\}} U_{m_1m_2m'_1m'_2}^i c_{im_1\sigma}^\dagger c_{im_2\sigma'}^\dagger c_{im'_2\sigma'} c_{im'_1\sigma} \quad (18)$$

where the (i, j) represents different crystal sites, $\{m\}$ label different orbitals and the $\{\sigma\}$ are spin indices. Coulomb matrix elements are defined in the usual way (see Eq. (8) with the screened Coulomb interactions in the basis of localized Wannier functions).

The simplified form of the LDA+DMFT Hamiltonian is related to the diagonal “density-density

approximation”

$$H = \sum_{\{im\sigma\}} t_{im,i'm'}^{\text{LDA}} c_{im\sigma}^\dagger c_{i'm'\sigma} + \frac{1}{2} \sum_{imm'\sigma} U_{mm'}^i n_{im\sigma} n_{im'-\sigma} + \frac{1}{2} \sum_{im \neq m'\sigma} (U_{mm'}^i - J_{mm'}^i) n_{im\sigma} n_{im'\sigma} \quad (19)$$

where i is the site index and m the orbital quantum numbers, $\sigma = \uparrow, \downarrow$ the spin projection, $c^\dagger, c$ the Fermi creation and annihilation operators ($n = c^\dagger c$), and t^{LDA} is the effective single-particle Hamiltonian obtained from the non-magnetic LDA with the correction for double counting of the average interactions among d -electrons. In the general case of a spin-polarized LDA Hamiltonian this correction is presented in Refs. [18, 37, 40]. In the magnetic LDA it is just a shift “back” of correlated d -states with respect to the s and p -states by the average Coulomb and exchange potential

$$E_{dc} = \frac{1}{2} \bar{U} n_d (1 - n_d) - \frac{1}{2} \bar{J} [n_{d\uparrow} (1 - n_{d\uparrow}) + n_{d\downarrow} (1 - n_{d\downarrow})]$$

with $\bar{U}$ and $\bar{J}$ being the average Coulomb and exchange interactions and $n_d = n_{d\uparrow} + n_{d\downarrow}$ the total number of correlated $d(f)$ -electrons.

The screened Coulomb and exchange vertices for the d -electrons

$$U_{mm'} = \langle mm' | U_{scr}(\mathbf{r} - \mathbf{r}') | mm' \rangle \quad (20)$$

$$J_{mm'} = \langle mm' | U_{scr}(\mathbf{r} - \mathbf{r}') | m'm \rangle \quad (21)$$

are expressed via the effective Slater integrals. We use the minimal spd -basis in the LMTO-TB formalism [42] and numerical orthogonalization for the $t^{\text{LDA}}(\mathbf{k})$ matrix [37]. The local density approximation [6] is used for the self-consistent electronic structure calculation.

In order to find the best local approximation for the self-energy we use the DMFT method [43] for real systems. This scheme becomes exact in the limit of infinite lattice coordination number [44]. The DMFT approach reduces the lattice many-body problem (Eq. (21)) to the self-consistent solution of an effective one-site Anderson model. In this case we need a local Green-function matrix which has the following form in the orthogonal Wannier representation

$$G(i\omega) = \sum_{\mathbf{k}} \{ (i\omega + \mu) \hat{1} - t^{\text{LDA}}(\mathbf{k}) - \Sigma(i\omega) \}^{-1}, \quad (22)$$

where μ is the chemical potential. Note that due to the cubic crystal symmetry of ferromagnetic bcc -iron the local Green function in the absence of spin-orbit interaction is diagonal both in the orbital and the spin indices. The so-called bath Green function that defines the effective Anderson model and preserves the double-counting of the local self-energy is obtained as a solution of the impurity model via [43]

$$\mathcal{G}^{-1}(i\omega) = G^{-1}(i\omega) + \Sigma(i\omega) \quad (23)$$

5 Solution of quantum impurity problem

5.1 Hirsch-Fye quantum Monte Carlo

As discussed above, 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 $n_{m\sigma} = c_{m\sigma}^\dagger c_{m\sigma}$ and $\mathbf{c}(\tau) = [c_{m\sigma}(\tau)]$ is a vector of Grassman variables) is given by:

$$S_{imp} = - \int_0^\beta d\tau \int_0^\beta d\tau' \text{Tr}[\mathbf{c}^\dagger(\tau) \mathcal{G}^{-1}(\tau, \tau') \mathbf{c}(\tau')] + \frac{1}{2} \sum_{m, m', \sigma} \int_0^\beta d\tau [U_{mm'} n_\sigma^m n_{-\sigma}^{m'} + (U_{mm'} - J_{mm'}) n_\sigma^m n_\sigma^{m'}]. \quad (24)$$

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

$$\mathbf{G}_\sigma(\tau - \tau') = -\frac{1}{Z} \int D[\mathbf{c}, \mathbf{c}^\dagger] e^{-S_{imp}} \mathbf{c}(\tau) \mathbf{c}^\dagger(\tau'), \quad (25)$$

where Z is the partition function.

The local Green functions for the imaginary time interval $[0, \beta]$ with the mesh $\tau_l = l\Delta\tau$, $l = 0, \dots, L-1$, and $\Delta\tau = \beta/L$, where $\beta = 1/T$ are calculated in the path-integral formalism [43,45]

$$G_m^{ll'} = \frac{1}{Z} \sum_{s_{mm'}^l} \det[O(s)] * G_m^{ll'}(s). \quad (26)$$

Here we redefined for simplicity $m \equiv \{m, \sigma\}$, and the so-called fermion-determinant $\det[O(s)]$ as well as the Green function for an arbitrary set of auxiliary fields $G(s) = O^{-1}(s)$ are obtained via the Dyson equation [46] for the imaginary-time matrix ($\mathbf{G}_m(s) \equiv G_m^{ll'}(s)$):

$$\mathbf{G}_m = [\mathbf{1} - (\mathbf{G}_m^0 - \mathbf{1})(e^{V_m} - \mathbf{1})]^{-1} \mathbf{G}_m^0,$$

where the effective fluctuation potential from the Ising fields $s_{mm'}^l = \pm 1$ is

$$V_m^l = \sum_{m'(\neq m)} \lambda_{mm'} s_{mm'}^l \sigma_{mm'}, \text{ where } \sigma_{mm'} = \begin{cases} 1, m < m' \\ -1, m > m' \end{cases}$$

and the discrete Hubbard-Stratonovich parameters are $\lambda_{mm'} = \text{arccosh}[\exp(\frac{1}{2}\Delta\tau U_{mm'})]$ [46]. Using the output local Green function from QMC and input bath Green functions the new self-energy is obtain via Eq. (23) and the self-consistent loop can be closed through Eq. (22). The main problem of the multiband QMC formalism is the large number of the auxiliary fields $s_{mm'}^l$. For each time slice l it is equal to $M(2M-1)$ where M is the total number of orbitals, giving 45 Ising fields for a d -shell. We computed the sum over these auxiliary fields in Eq.(26) using importance-sampling QMC, and performed a dozen self-consistent iterations over the

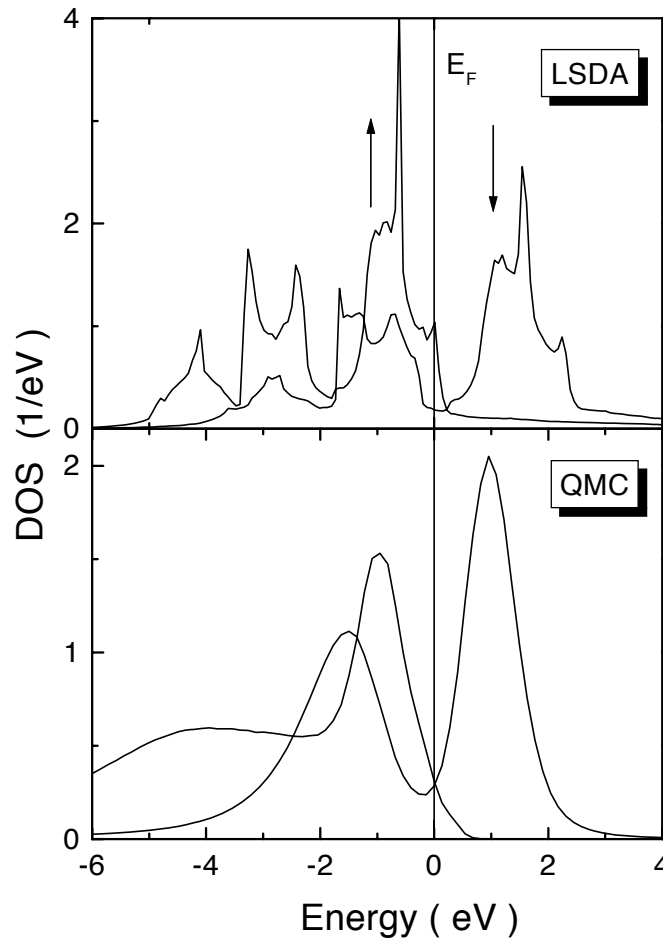


Fig. 5: Spin-resolved density of d -states and magnetic moments for ferromagnetic iron calculated in LDA (top) and LDA+QMC (bottom) for different average Coulomb interactions with $J=0.9$ eV and temperature $T=850$ K.

self-energy Eqs. (22,23,26). The number of QMC sweeps was of the order of 10^5 on the CRAY-T3e supercomputer. The final $G_m(\tau)$ has very little statistical noise. We use the maximum entropy method [47] for analytical continuations of the QMC Green functions to the real axis.

Comparison of the total density of states (DOS) with the results of LDA calculations (Fig. 5) shows reasonable agreement for the single-particle properties of the not “highly correlated” ferromagnetic iron. We calculate *bcc*-iron at its experimental lattice constant with 256 k -points in the irreducible part of Brillouin zone. The Matsubara summation corresponds to the temperature of about $T = 850$ K. The average magnetic moment is about $1.9 \mu_B$, which corresponds to a small reduction of the LDA-value of $2.2 \mu_B$ for such a high temperature. The DOS curves in the LDA+ Σ approach with the exact QMC solution of on-site multi-orbital problem is similar to that obtained within the simple perturbative fluctuation-exchange (FLEX) approximation described below. The discussion of the results and the comparison with experimental data will be given in Section 4.

5.2 Continuous-time quantum Monte Carlo

Even though DMFT reduces the extended lattice problem to a single-site problem, the solution of the underlying Anderson impurity model remains a formidable quantum many-body problem that requires accurate solvers. Recently a new class has emerged, the continuous-time quantum impurity solvers. These are based on stochastic, i.e., Monte-Carlo methods and mainly come in two different flavors: The weak and strong-coupling approach.

The weak-coupling or interaction expansion (CT-INT) continuous-time quantum Monte Carlo algorithm for fermions was originally introduced by Aleksey Rubtsov [48]. The power of the new CT-QMC scheme is that it performs the path integral without any transformation to effective non-interacting models and that it can be used for any complicated electron-electron vertex. We introduce the algorithm in the path integral formulation for the single-orbital Anderson impurity problem with a Hubbard-type interaction $Un_{\uparrow}n_{\downarrow}$. The generalization to the multiorbital case can be found in Ref. [49]. To start, the action for the Anderson impurity model, up to an irrelevant additive constant, is divided into a Gaussian S_0 and an interacting part S_U

$$S_0 = \sum_{\sigma} \int_0^{\beta} d\tau \int_0^{\beta} d\tau' c_{\sigma}^*(\tau) [\partial_{\tau} - \mu + \Delta(\tau - \tau') + U\alpha_{-\sigma}(\tau)\delta(\tau - \tau')] c_{\sigma}(\tau'), \quad (27)$$

$$S_U = U \int_0^{\beta} d\tau [c_{\uparrow}^*(\tau)c_{\uparrow}(\tau) - \alpha_{\uparrow}(\tau)][c_{\downarrow}^*(\tau)c_{\downarrow}(\tau) - \alpha_{\downarrow}(\tau)]. \quad (28)$$

The parameters α are introduced to control the sign problem. A formal series expansion for the partition function is obtained by expanding the exponential in the interaction term,

$$\mathcal{Z} = \int e^{-S_0[c^*, c]} \sum_{k=0}^{\infty} \frac{(-1)^k}{k!} U^k \int_0^{\beta} d\tau_1 \dots \int_0^{\beta} d\tau_k [c_{\uparrow}^*(\tau_1)c_{\uparrow}(\tau_1) - \alpha_{\uparrow}(\tau_1)][c_{\downarrow}^*(\tau_1)c_{\downarrow}(\tau_1) - \alpha_{\downarrow}(\tau_1)] \dots [c_{\uparrow}^*(\tau_k)c_{\uparrow}(\tau_k) - \alpha_{\uparrow}(\tau_k)][c_{\downarrow}^*(\tau_k)c_{\downarrow}(\tau_k) - \alpha_{\downarrow}(\tau_k)] \mathcal{D}[c^*, c]. \quad (29)$$

Using the definition of the average over the noninteracting action:

$$\langle \dots \rangle_0 = \frac{1}{\mathcal{Z}_0} \int \mathcal{D}[c^*, c] \dots \exp(-S_0), \quad (30)$$

the partition function can be expressed in the following form

$$\mathcal{Z} = \mathcal{Z}_0 \sum_{k=0}^{\infty} \int_0^{\beta} d\tau_1 \dots \int_{\tau_{k-1}}^{\beta} d\tau_k \text{sgn}(\Omega_k) |\Omega_k|, \quad (31)$$

where the integrand is given by

$$\Omega_k = (-1)^k U^k \langle [c_{\uparrow}^*(\tau_1)c_{\uparrow}(\tau_1) - \alpha_{\uparrow}(\tau_1)][c_{\downarrow}^*(\tau_1)c_{\downarrow}(\tau_1) - \alpha_{\downarrow}(\tau_1)] \dots [c_{\uparrow}^*(\tau_k)c_{\uparrow}(\tau_k) - \alpha_{\uparrow}(\tau_k)][c_{\downarrow}^*(\tau_k)c_{\downarrow}(\tau_k) - \alpha_{\downarrow}(\tau_k)] \rangle_0. \quad (32)$$

Note that here the range of time integration has been changed such that time ordering is explicit: $\tau_1 < \dots < \tau_{k-1} < \tau_k$. For a given set of times all $k!$ permutations of this sequence

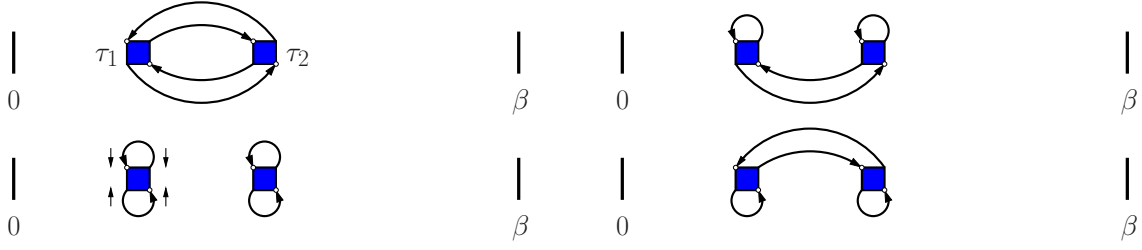


Fig. 6: The four contributions to the partition function for $k = 2$. The interaction vertices are depicted by squares. Bare Green functions are shown as lines. Vertical arrows indicate the spin direction. Connecting the vertices by Green functions in all possible ways is the interpretation of the determinant.

contribute to Eq. 29. These can be brought into the standard sequence by permuting quadruples of Grassmann numbers, and hence without gaining an additional sign. Since all terms are subject to time-ordering, their contribution to the integral is identical, so that the factor $1/k!$ in (29) cancels. A configuration can be fully characterized by specifying a perturbation order k and an (unnumbered) set of k times: $C_k = \{\tau_1, \dots, \tau_k\}$.

The Monte Carlo algorithm performs importance sampling over this configuration space. The weight of a configuration is thereby taken to be equal to the modulus of the integrand, Eq. 32. Since S_0 is Gaussian, the average over the noninteracting system can be evaluated using Wick's theorem. Hence the weight of a configuration is essentially given by a fermionic determinant of a matrix containing the bare Green functions

$$\Omega_k = (-1)^k U^k \prod_{\sigma} \det \hat{g}^{\sigma}, \quad (33)$$

where the local Green function in the α fields is equal to

$$(\hat{g}^{\sigma})_{ij} = g_0^{\sigma}(\tau_i - \tau_j) - \alpha_{\sigma}(\tau_i) \delta_{ij}. \quad (34)$$

Note that determinants for different spin orientations factorize since the Green function is diagonal in spin-space.

The hybridization expansion (CT-HYB) or strong-coupling algorithm was initially introduced by Philipp Werner *et al.* [50] and has been generalized to multi-orbital systems with general interactions [51, 52]. Here the algorithm is discussed in the segment representation, which allows for a very fast computation of the trace for density-density type interactions. The action is regrouped into the atomic part

$$S_{\text{at}} = \int_0^{\beta} d\tau \sum_{\sigma} c_{\sigma}^*(\tau) [\partial_{\tau} - \mu] c_{\sigma}(\tau) + U \int_0^{\beta} d\tau c_{\uparrow}^*(\tau) c_{\uparrow}(\tau) c_{\downarrow}^*(\tau) c_{\downarrow}(\tau) \quad (35)$$

and the part of the action S_{Δ} which contains the hybridization term:

$$S_{\Delta} = - \int_0^{\beta} d\tau' \int_0^{\beta} d\tau \sum_{\sigma} c_{\sigma}(\tau) \Delta(\tau - \tau') c_{\sigma}^*(\tau'). \quad (36)$$

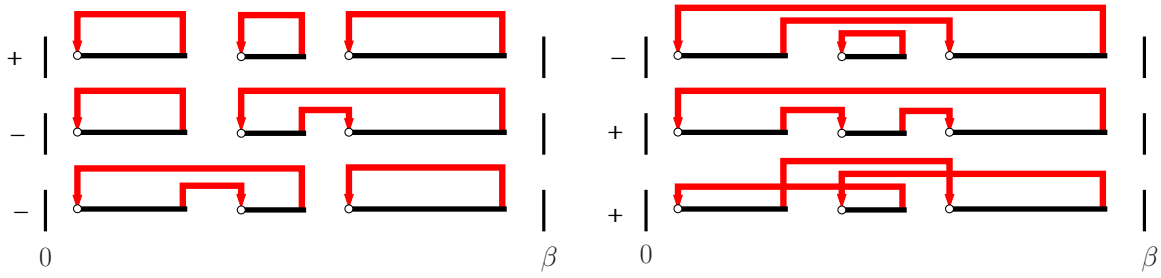


Fig. 7: Diagrammatic representation of the six contributions to the partition function for spinless fermions at $k = 3$. An electron is inserted at the start of a segment (marked by an open circle) and removed at the segment endpoint. The hybridization function lines $\Delta(\tau_i - \tau'_j)$ (shown in red) are connected to the segments in all possible ways. The sign of each diagram is given on the left. The diagrams can be collected into a determinant. Reproduced from Ref. [50].

Here the sign is taken out by reversing the original order of c and c^* to avoid an alternating sign in the expansion. To simplify the notation, consider first the spinless fermion model, which is obtained by disregarding the spin sums and interaction in Eqs. (35), (36). The series expansion for the partition function is generated by expanding in the hybridization term

$$\mathcal{Z} = \int \mathcal{D}[c^*, c] e^{-S_{\text{at}}} \sum_k \frac{1}{k!} \int_0^\beta d\tau'_1 \int_0^\beta d\tau_1 \dots \int_0^\beta d\tau'_k \int_0^\beta d\tau_k \times \\ \times c(\tau_k) c^*(\tau'_k) \dots c(\tau_1) c^*(\tau'_1) \Delta(\tau_1 - \tau'_1) \dots \Delta(\tau_k - \tau'_k). \quad (37)$$

The important observation now is that, at any order, the diagrams can be collected into a determinant of hybridization functions. The partition function then takes the form

$$\mathcal{Z} = \mathcal{Z}_{\text{at}} \sum_k \int_0^\beta d\tau'_1 \int_{\tau'_1}^\beta d\tau_1 \dots \int_{\tau_{k-1}}^\beta d\tau'_k \int_{\tau'_k}^{\circ\tau'_k} d\tau_k \langle c(\tau_k) c^*(\tau'_k) \dots c(\tau_1) c^*(\tau'_1) \rangle_{\text{at}} \det \hat{\Delta}^{(k)}, \quad (38)$$

where the average is over the states of the atomic problem described by S_{at} . Here $\det \hat{\Delta}^{(k)}$ denotes the determinant of the matrix of hybridizations $\hat{\Delta}_{ij} = \Delta(\tau_i - \tau'_j)$. The diagrams contributing to the partition function for $k = 3$ are shown in Fig. 7. A diagram is depicted by a collection of segments, where a segment is symbolic for the time interval where the impurity is occupied. The collection of diagrams obtained by connecting the hybridization lines in all possible ways corresponds to the determinant. Collecting the diagrams into a determinant is essential to alleviate or completely suppress the sign problem. Note that the imaginary time interval in Eq. (38) is viewed as a circle denoted by $\circ\tau'_k$. The trajectories in the path integral are subject to antiperiodic boundary conditions which is accommodated by an additional sign if a segment winds around the circle.

For the single-orbital Anderson impurity model with Hubbard interaction the segment picture still holds and gives a very intuitive picture of the imaginary time dynamics. A configuration is visualized by two separate timelines, one for each spin. The additional sum over spins, $\sum_{\sigma_1 \dots \sigma_k}$,

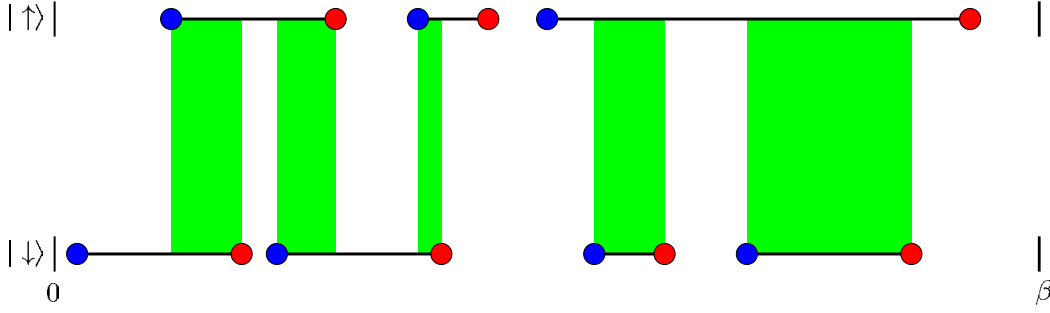


Fig. 8: Example one band CT-HYB: a segment picture: blue dots illustrate annihilation operators, red ones creation operators, and the black line represent the hybridization function $\Delta(\tau_i - \tau_j)$. The green regions represent the time interval at which two electrons are present on the impurity with the total time l_d for which the U price has to be paid.

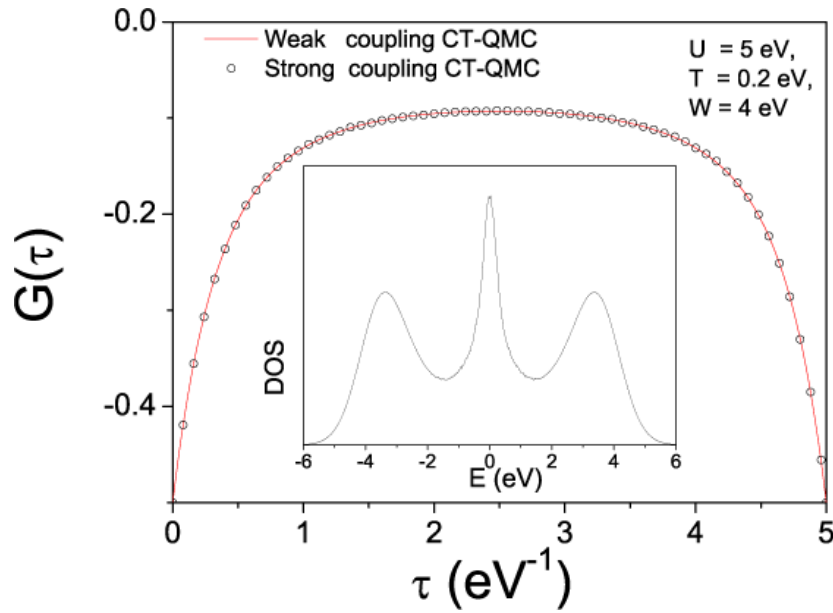


Fig. 9: Comparison of the weak coupling (CT-INT) and strong coupling (CT-HYB) CT-QMC impurity solvers for one-band semicircular model with $U \geq W$. In the insert the density of states obtained with maximum entropy scheme is shown

which enters in the first line of Eq. (38), generates contributions such as the one shown in Fig. 8. The only difference from the spinless fermion model is that when the impurity is doubly occupied, the energy U has to be paid and the trace is $e^{\mu(l_\uparrow + l_\downarrow) - U l_d}$, where l_σ is the time spent on the impurity for an electron with spin σ and l_d is the time the impurity is doubly occupied.

In Fig. 9 we show the comparison of CT-INT and CT-HYB calculations for a strongly coupled ($U \geq W$) single band model. The perfect agreement of these two complementary CT-QMC schemes supports the important conclusion about the possibility of the numerically exact solution of quantum impurity problems.

5.3 Fluctuation exchange approximation

The QMC method described above is probably the most rigorous practical way to solve an effective impurity problem in the framework of DMFT. However, it is rather time consuming. Besides that, in the previous section we did not work with the *complete* four-index Coulomb matrix

$$\langle 12 | v | 34 \rangle = \int d\mathbf{r} d\mathbf{r}' \psi_1^*(\mathbf{r}) \psi_2^*(\mathbf{r}') v_{scr}(\mathbf{r} - \mathbf{r}') \psi_3(\mathbf{r}) \psi_4(\mathbf{r}'), \quad (39)$$

where we define for simplicity $m_1 \equiv 1$.

For moderately strong correlations (which is the case of iron group metals) one can propose an approximate scheme which is more suitable for the calculations. It is based on the fluctuation exchange (FLEX) approximation by Bickers and Scalapino [53], generalized to the multiband spin-polarized case [18, 37, 54]. The electronic self-energy in the FLEX is equal to

$$\Sigma = \Sigma^{\text{HF}} + \Sigma^{(2)} + \Sigma^{(ph)} + \Sigma^{(pp)}, \quad (40)$$

where the Hartree-Fock contribution has a standard form

$$\Sigma_{12,\sigma}^{\text{HF}} = \sum_{34} \left[\langle 13 | v | 24 \rangle \sum_{\sigma'} n_{34}^{\sigma'} - \langle 13 | v | 42 \rangle n_{34}^{\sigma} \right], \quad (41)$$

with the occupation matrix $n_{12}^{\sigma} = G_{21}^{\sigma}(\tau \rightarrow -0)$; this contribution to Σ is equivalent to the spin-polarized “rotationally-invariant” LDA+ U method [40].

The second-order contribution in the spin-polarized case reads

$$\begin{aligned} \Sigma_{12,\sigma}^{(2)}(\tau) = & - \sum_{\{3-8\}} \langle 13 | v | 74 \rangle G_{78}^{\sigma}(\tau) \times \\ & \times \left[\langle 85 | v | 26 \rangle \sum_{\sigma'} G_{63}^{\sigma'}(\tau) G_{45}^{\sigma'}(-\tau) - \langle 85 | v | 62 \rangle G_{63}^{\sigma}(\tau) G_{45}^{\sigma}(-\tau) \right], \end{aligned} \quad (42)$$

and the higher-order particle-hole (or particle-particle) contribution

$$\Sigma_{12,\sigma}^{(ph)}(\tau) = \sum_{34,\sigma'} W_{13,42}^{\sigma\sigma'}(\tau) G_{34}^{\sigma'}(\tau), \quad (43)$$

with $p-h$ ($p-p$) fluctuation potential matrix

$$W^{\sigma\sigma'}(i\omega) = \begin{bmatrix} W^{\uparrow\uparrow}(i\omega) & W^{\uparrow\downarrow}(i\omega) \\ W^{\downarrow\uparrow}(i\omega) & W^{\downarrow\downarrow}(i\omega) \end{bmatrix}, \quad (44)$$

where the spin-dependent effective potentials have a generalized RPA-form and can be found in [18]. Note that for both the $p-h$ and $p-p$ channels the effective interactions, according to Eq. (44), are non-diagonal matrices in *spin space* as in the QMC-scheme, in sharp contrast to any mean-field approximation like LDA. This can be important for spin-dependent transport phenomena in transition metal multilayers.

We could further reduce the computational procedure by neglecting dynamical interactions in the p - p channel since the most important fluctuations in itinerant electron magnets are spin-fluctuations in the p - h channel. We take into account static (of T -matrix type) renormalizations of the effective interactions, replacing the bare matrix $U_{12,34} = \langle 12 | v | 34 \rangle$ in the FLEX-equations with the corresponding spin-dependent scattering T -matrix

$$\langle 12 | T^{\sigma\sigma'} | 34 \rangle = \langle 12 | v | 34 \rangle - \sum_{5678} \langle 12 | v | 56 \rangle \int_0^\beta d\tau G_{56}^\sigma(\tau) G_{78}^{\sigma'}(\tau) \langle 78 | T^{\sigma\sigma'} | 34 \rangle. \quad (45)$$

Similar approximation have been checked for the Hubbard model [55] and appear to be accurate enough for not too large U . Finally, in the spirit of the DMFT-approach $\Sigma = \Sigma[G_0]$, and all the Green functions in the self-consistent FLEX-equations are in fact the bath Green-functions G_0 .

6 Effective magnetic interactions in LDA+DMFT

A useful scheme for analyses of exchange interactions in the LDA approach is the so called “local force theorem”. In this case the calculation of small total energy changes reduces to variations of the one-particle density of states [56, 57]. First of all, let us prove the analog of the local force theorem in the LDA+DMFT approach. In contrast with standard density-functional theory, it deals with the real dynamical quasiparticles defined via Green functions for the correlated electrons rather than with Kohn-Sham “quasiparticles” which are, strictly speaking, only auxiliary states for the total energy calculations. Therefore, instead of working with the thermodynamic potential as a *density* functional we have to start from the general expression for Ω in terms of the exact Green function given in Table I. We have to also keep in mind the Dyson equation $G^{-1} = G_0^{-1} - \Sigma$ and the variational identity $\delta\Phi_{LW} = \text{Tr} \Sigma \delta G$. Here $\text{Tr} = \text{Tr}_{\omega i L \sigma}$ is the sum over Matsubara frequencies $\text{Tr}_\omega \dots = \sum_\omega \dots$, with $\omega = \pi T (2n + 1)$, $n = 0, \pm 1, \dots$, with T the temperature, $i L \sigma$ the numbers of sites (i), the orbital quantum numbers ($L = l, m$), and the spin projections σ , correspondingly. We represent the expression for Ω as a difference of “single particle” (sp) and “double counted” (dc) terms as is usual in density-functional theory. When neglecting the quasiparticle damping, Ω_{sp} will be nothing but the thermodynamic potential of “free” fermions but with exact quasiparticle energies. Suppose we change the external potential, for example, by small spin rotations. Then the variation of the thermodynamic potential can be written as

$$\delta\Omega = \delta^* \Omega_{sp} + \delta_1 \Omega_{sp} - \delta\Omega_{dc} \quad (46)$$

where δ^* is the variation without taking into account the change of the “self-consistent potential”, i.e. self energy, and δ_1 is the variation due to the change of Σ . To avoid a possible misunderstanding, note that we consider the variation of Ω in the general “non-equilibrium” case when the torques acting on spins are nonzero and therefore $\delta\Omega \neq 0$. In order to study the response of the system to general spin rotations one can consider either variations of the spin directions at fixed effective fields or, vice versa, rotations of the effective fields, i.e. variations

of Σ , at fixed magnetic moments. We use the second way. Taking into account the variational property of Φ , it can be easily shown (cf. Ref. [24]) that

$$\delta_1 \Omega_{sp} = \delta \Omega_{dc} = \text{Tr } G \delta \Sigma \quad (47)$$

and hence

$$\delta \Omega = \delta^* \Omega_{sp} = -\delta^* \text{Tr } \ln [\Sigma - G_0^{-1}] \quad (48)$$

which is an analog of the “local force theorem” in density-functional theory [57].

In the LDA+DMFT scheme, the self energy is local, i.e., is diagonal in the site indices. Let us write the spin-matrix structure of the self-energy and Green function in the following form

$$\Sigma_i = \Sigma_i^c + \mathbf{\Sigma}_i^s \cdot \boldsymbol{\sigma}, \quad G_{ij} = G_{ij}^c + \mathbf{G}_{ij}^s \cdot \boldsymbol{\sigma} \quad (49)$$

where $\Sigma_i^{(c,s)} = (\Sigma_i^\uparrow \pm \Sigma_i^\downarrow)/2$ and $\mathbf{\Sigma}_i^s = \Sigma_i^s \mathbf{e}_i$, with $\mathbf{e}_i$ being the unit vector in the direction of effective spin-dependent potential on site i , $\boldsymbol{\sigma} = (\sigma_x, \sigma_y, \sigma_z)$ are the Pauli matrices, $G_{ij}^c = \text{Tr}_\sigma(G_{ij})/2$ and $\mathbf{G}_{ij}^s = \text{Tr}_\sigma(G_{ij}\boldsymbol{\sigma})/2$. We assume that the bare Green function G^0 does not depend on spin directions and all the spin-dependent terms including the Hartree-Fock terms are incorporated into the self-energy. Spin excitations with low energies are connected with the rotations of vectors $\mathbf{e}_i$

$$\delta \mathbf{e}_i = \delta \boldsymbol{\varphi}_i \times \mathbf{e}_i \quad (50)$$

According to the “local force theorem” (48) the corresponding variation of the thermodynamic potential can be written as

$$\delta \Omega = \delta^* \Omega_{sp} = \mathbf{V}_i \delta \boldsymbol{\varphi}_i, \quad (51)$$

where the torque is equal to

$$\mathbf{V}_i = 2 \text{Tr}_{\omega L} [\mathbf{\Sigma}_i^s \times \mathbf{G}_{ii}^s]. \quad (52)$$

Using the spinor structure of the Dyson equation one can write the Green function in this expression in terms of pair contributions. As a result, we represent the total thermodynamic potential of spin rotations or the effective Hamiltonian in the form [38]

$$\Omega_{spin} = - \sum_{ij} \text{Tr}_{\omega L} \{ (\mathbf{G}_{ij}^s \cdot \mathbf{\Sigma}_j^s) (\mathbf{G}_{ji}^s \cdot \mathbf{\Sigma}_i^s) - \Sigma_i^s G_{ij}^c \Sigma_j^s G_{ji}^c - i (\mathbf{\Sigma}_i^s \times \mathbf{G}_{ij}^c \cdot \mathbf{\Sigma}_j^s) \mathbf{G}_{ji}^s \}. \quad (53)$$

One can show by direct calculation that

$$\left[\frac{\delta \Omega_{spin}}{\delta \boldsymbol{\varphi}_i} \right]_{G=\text{const}} = \mathbf{V}_i. \quad (54)$$

This means that $\Omega_{spin} \{ \mathbf{e}_i \}$ is the effective spin Hamiltonian. The last term in Eq. (53) is nothing but a Dzialoshinskii-Moriya interaction term. It is non-zero only in the relativistic case where $\mathbf{\Sigma}_j^s$ and $\mathbf{G}_{ji}^s$ can be, generally speaking, “non-parallel” and $G_{ij} \neq G_{ji}$ for crystals without inversion center.

In the non-relativistic case, one can rewrite the spin Hamiltonian for small spin deviations near collinear magnetic structures in the following form

$$\Omega_{spin} = - \sum_{ij} J_{ij} \mathbf{e}_i \cdot \mathbf{e}_j, \quad (55)$$

where

$$J_{ij} = -\text{Tr}_{\omega_L} \left(\Sigma_i^s G_{ij}^\uparrow \Sigma_j^s G_{ji}^\downarrow \right) \quad (56)$$

are the effective exchange parameters. This formula generalizes the LDA expressions of [57,58] to the case of correlated systems.

The spin-wave spectrum in ferromagnets can be considered both directly from the exchange parameters or from the energy of corresponding spiral structures (cf. Ref. [57]). In the non-relativistic case when anisotropy is absent one has

$$\omega_{\mathbf{q}} = \frac{4}{M} \sum_j J_{0j} (1 - \cos \mathbf{q} \cdot \mathbf{R}_j) \equiv \frac{4}{M} [J(\mathbf{0}) - J(\mathbf{q})], \quad (57)$$

where M is the magnetic moment (in Bohr magnetons) per magnetic ion.

It should be noted that the expression for the spin-stiffness tensor $D_{\alpha\beta}$ defined by the relation $\omega_{\mathbf{q}} = D_{\alpha\beta} q_\alpha q_\beta$ ($\mathbf{q} \rightarrow 0$) in terms of exchange parameters has to be exact as the consequence of the phenomenological Landau-Lifshitz equations, which are definitely correct in the long-wavelength limit. Direct calculation based on variation of the total energy under spiral spin rotations (cf. Ref. [57]) leads to the following expression

$$D_{\alpha\beta} = -\frac{2}{M} \text{Tr}_{\omega_L} \sum_{\mathbf{k}} \left(\Sigma^s \frac{\partial G^\uparrow(\mathbf{k})}{\partial k_\alpha} \Sigma^s \frac{\partial G^\downarrow(\mathbf{k})}{\partial k_\beta} \right), \quad (58)$$

where $\mathbf{k}$ is the quasi-momentum and the summation is over the Brillouin zone. The expressions Eqs. (56) and (57) are reminiscent of usual RKKY indirect exchange interactions in the s - d exchange model (with Σ^s instead of the s - d exchange integral). One can prove that this expression for the stiffness is exact within the local approximation [59]. At the same time, the exchange parameters themselves, generally speaking, differ from the exact response characteristics defined via static susceptibility since the latter contains vertex corrections. The derivation of approximate exchange parameters from the variations of the thermodynamic potential can be useful for the estimation of J_{ij} in different magnetic systems.

7 LDA+DMFT results for itinerant ferromagnetic metals

We have started from the spin-polarized LDA band structure of ferromagnetic iron within the LMTO method [42] in the minimal s, p, d basis set and used numerical orthogonalization to find the H_t part of our starting Hamiltonian. We take into account Coulomb interactions only between d -states. The correct parameterization of the H_U part is indeed a serious problem. For example, first-principle estimations of average Coulomb interactions (U) [30,60] in iron lead to unreasonably large values of order of 5–6 eV in comparison with experimental values of the U -parameter in the range of 1–2 eV [30]. A semi-empirical analysis of the appropriate interaction value [61] gives $U \simeq 2.3$ eV. The difficulties with choosing the correct value of U are connected with complicated screening problems, definitions of orthogonal orbitals in the crystal, and contributions of the inter-site interactions. In the quasi-atomic (spherical) approximation the full

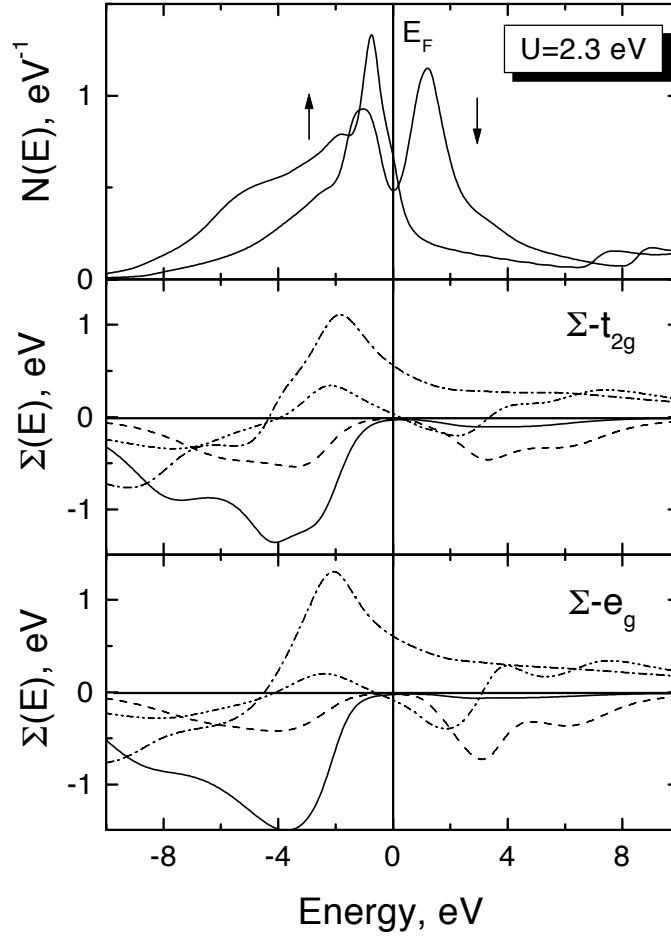


Fig. 10: Total spin-polarized density of states and d -part of the self-energy for iron with $U = 2.3$ eV and $J = 0.9$ eV for the temperature $T = 750$ K. Two different self-energies for t_{2g} and e_g d -states in the cubic crystal field symmetry are presented. The four different lines correspond to imaginary-part spin-up (full line) and spin-down (dashed line) as well as real-part spin-up (dashed-dot line) and spin-down (dashed-double-dot line).

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 [11,37]. 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 [62].

Note that the value of the intra-atomic (Hund) exchange interaction J is not sensitive to the screening and approximately equals 0.9 eV in different estimations [60]. For the most important parameter U , which defines the bare vertex matrix Eq. (39), we use the value $U = 2.3$ eV for Fe [61], $U = 3$ eV for Co and Mn and $U = 4$ eV for Ni and Cu. To calculate the spectral functions $A_\sigma(\mathbf{k}, E) = -\text{Tr}_L G_\sigma(\mathbf{k}, E + i0)/\pi$ and DOS as their sum over the Brillouin zone, we first performed analytical continuations for the matrix self-energy from Matsubara frequencies to the real axis using the Pade approximation [63], and then numerically inverted the Green-function matrix as in Eq. (22) for each $\mathbf{k}$ -point. In the self-consistent solution of the FLEX equations we used 1024 Matsubara frequencies and the FFT-scheme with an energy cut-off at 100 eV. The

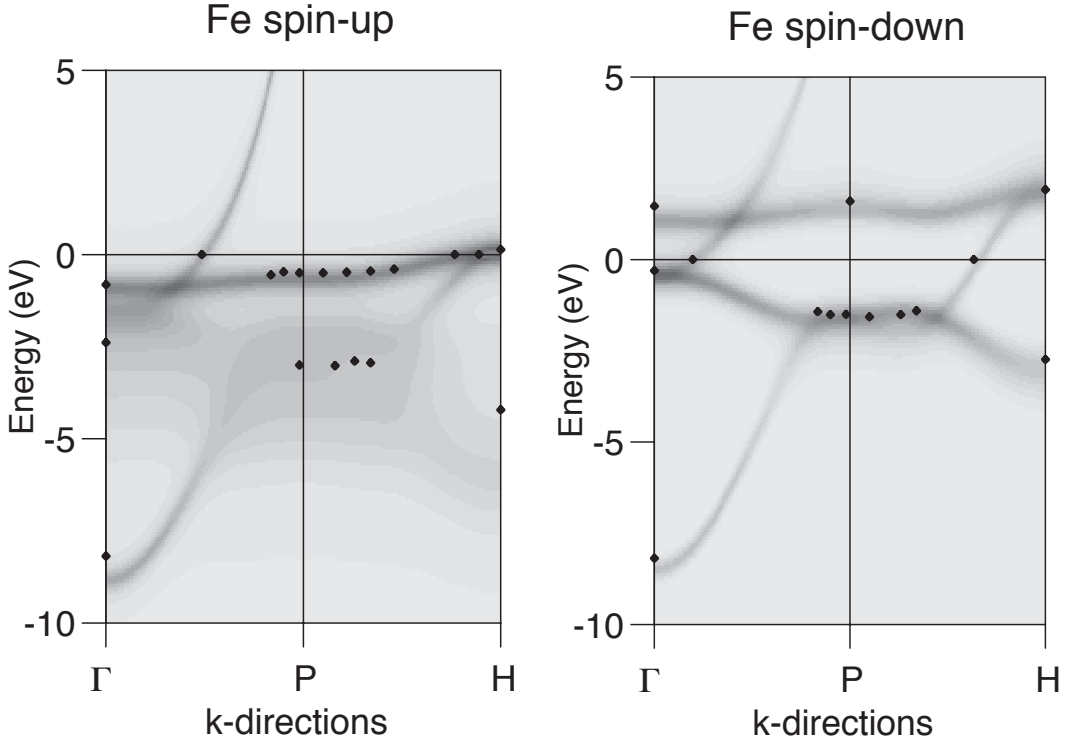


Fig. 11: *Spectral function of ferromagnetic iron for spin-up (a) and spin-down (b) and the two k -directions in the Brillouin zone compared with the experimental angle resolved photoemission and de Haas van Alphen (at the $E_F=0$) points.*

sums over the irreducible Brillouin zone have been made with 72 k -points for SCF-iterations and 1661 k -points for the final total density of states.

The depolarization of states near the Fermi level is another important correlation effect. The decrease of the ratio $P = [N_{\uparrow}(E_F) - N_{\downarrow}(E_F)] / [N_{\uparrow}(E_F) + N_{\downarrow}(E_F)]$ is a typical sign of spin-polaron effects [31, 64]. In our approach these effects are taken into account through the $W_{\uparrow\downarrow}^{(ph)}$ terms in the effective spin-polarized LDA+DMFT potential (Eq. (44)).

The energy dependence of self-energy in Fig. 10 shows the characteristic features of moderately correlated systems. At low energies $|E| < 1$ eV we see a typical Fermi-liquid behavior $\text{Im}\Sigma(E) \sim -E^2$, and $\partial \text{Re}\Sigma(E) / \partial E < 0$. At the same time, for the states beyond this interval within the d -bands the damping is rather large (of the order of 1 eV) so these states correspond to ill-defined quasiparticles, especially for the occupied states. This is probably one of the most important conclusions of our calculations. Qualitatively it was already pointed out in Ref. [33] on the basis of model second-order perturbation-theory calculations. We have shown that this is still the case for the realistic quasiparticle structure of iron with a reasonable value for the Coulomb interaction parameter.

Due to the noticeable broadening of the quasiparticle states, a description of the computational results in terms of effective band structure (determined, for example, from the maximum of the spectral density) would be incomplete. We present in Fig. 11 the *full* spectral density $A_{\sigma}(\mathbf{k}, E)$ including both coherent and incoherent parts as a function of $\mathbf{k}$ and E . We see that in general

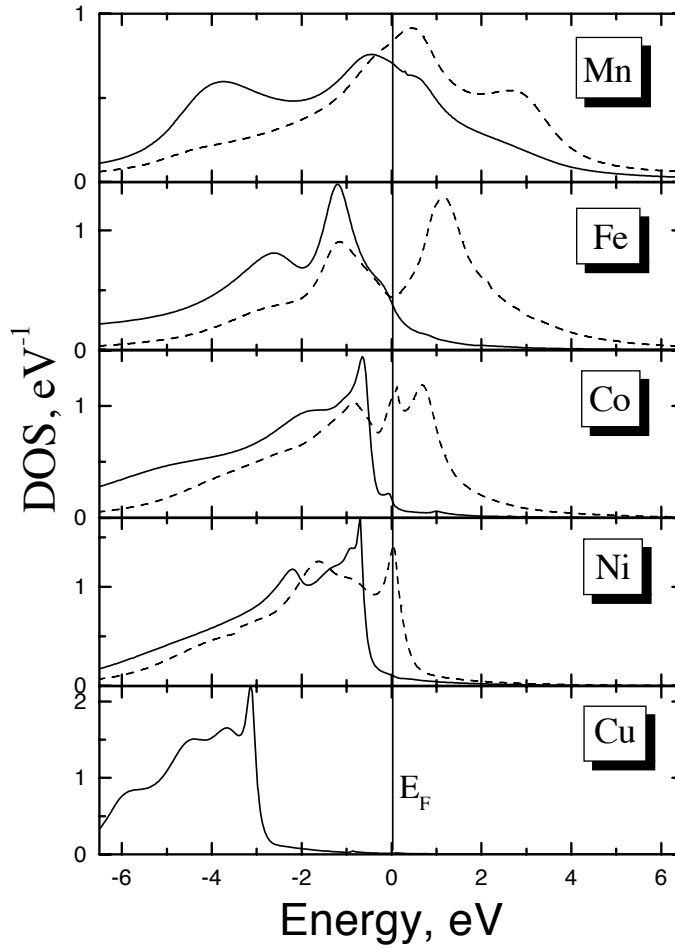


Fig. 12: Spin-polarized partial 3d density of states for different transition metals at temperature $T=750$ K. The full line is the spin-up, the dashed line the spin-down DOS.

the maxima of the spectral density (dark regions) coincide with the experimentally obtained band structure. However, for occupied majority spin states at about -3 eV the distribution of the spectral density is rather broad and the description of these states in terms of the quasi-particle dispersion is problematic. This conclusion is in complete quantitative agreement with raw experimental data on angle-resolved spin-polarized photoemission [65] with the broad non-dispersive second peak in the spin-up spectral function around -3 eV.

Comparison of the DOS for transition metals in the Fig. 12 shows interesting correlation effects. First of all, the most prominent difference from the LDA calculation is observed for antiferromagnetic *fcc*-Mn. There is the clear formation of lower and upper Hubbard bands around ± 3 eV. Such behavior is related with the half-filled Mn *d*-shell, which corresponds to a large phase space for particle-hole fluctuations. For ferromagnetic *bcc*-Fe, the *p-h* excitations are suppressed by the large exchange splitting and a *bcc* structural minimum in the DOS near the Fermi level. In the case of ferromagnetic *fcc*-Co and Ni, correlation effects are more important than for Fe since there is no structural *bcc*-dip in the density of states. One can see the formation of a "three-peak" structure for the spin-down DOS for Co and Ni and satellite formation around

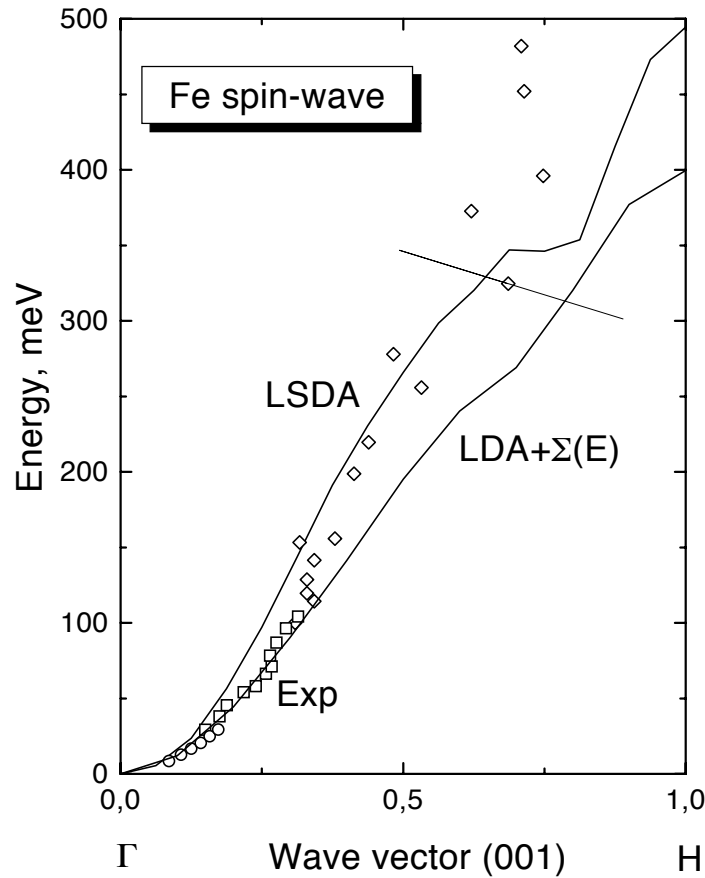


Fig. 13: The spin-wave spectrum for ferromagnetic iron in LDA and LDA+ Σ compared with different experiments (circles [16], squares [17], and diamonds [18]) (a); The corresponding spin-wave spectrum from the LDA+ Σ scheme in the (110) plane (b).

-5 eV. In order to describe the satellite formation more carefully one needs to include T -matrix effects [29, 34] or use the QMC scheme in LDA+DMFT calculations. Finally, there are no big correlation effects in non-magnetic *fcc*-Cu since the *d*-states are located well below the Fermi level.

Using the self-consistent values for $\Sigma_m(i\omega)$ computed by QMC, we calculate the exchange interactions (Eq. 56) and spin-wave spectrum (Eq. 57) using the four-dimensional fast Fourier transform (FFT) method [66] in $(\mathbf{k}, i\omega)$ space with a mesh of $20^3 \times 320$. The spin-wave spectrum for ferromagnetic iron is presented in Fig. 13 with comparison to the results of LDA-exchange calculations [57] and with different experimental data [67–69]. The room-temperature neutron scattering experiments have a sample dependence (Fe-12%Si in Ref. [67, 69] and Fe-4%Si in Ref. [68]) due to problems with the *bcc*-Fe crystal growth. Note that for high-energy spin-waves the experimental data [69] has large error-bars due to Stoner damping (we show one experimental point with the uncertainties in $\mathbf{q}$ space). On the other hand, the expression of magnon frequency in terms of exchange parameters itself becomes problematic in that region due to the breakdown of the adiabatic approximation. Therefore we think that the comparison of our theoretical results with the experimental spin-wave spectrum for large energy needs addi-

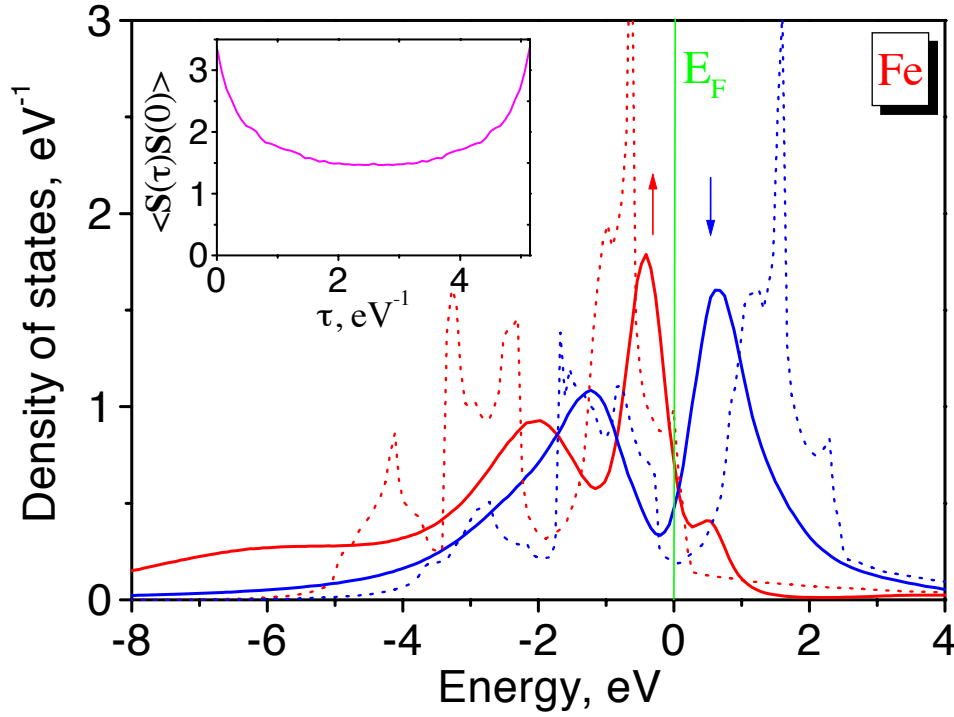


Fig. 14: LDA+DMFT results for ferromagnetic iron ($T = 0.8 T_C$). The partial densities of the d -states (full lines) are compared with the corresponding LDA results at zero temperature (dashed lines) for the spin-up (red lines, arrow-up) and spin-down (blue lines, arrow-down) states. The insert shows the spin-spin autocorrelation function for $T=1.2 T_C$.

tional investigation of Stoner excitations and calculations of the dynamical susceptibility in the LDA+DMFT approach [43]. Within the LDA scheme one could use the linear-response formalism [70] to calculate the spin-wave spectrum with the Stoner renormalizations, which should give in principle the same spin-wave stiffness as our LDA calculations. Our LDA spin-wave spectrum agrees well with the results of frozen magnon calculations [71, 72].

At the lower-energy, where the present adiabatic theory is reliable, the LDA+DMFT spin-wave spectrum agrees better with experiments than the result of the LDA calculations. Experimental values of the spin-wave stiffness $D = 280 \text{ meV/\AA}^2$ [68] agree well with the theoretical LDA+DMFT estimate of $260 \text{ meV/\AA}^2$ [38].

Self-consistent LDA+DMFT results for the local spectral function of iron and nickel are shown in Figs. 14 and 15, respectively. The LDA+DMFT approach describes well all the qualitative features of the density of states (DOS), which is especially non-trivial for nickel. Our QMC results reproduce well the three main correlation effects on the one particle spectra below T_C [75–77]: the presence of the famous 6 eV satellite, the 30% narrowing of the occupied part of d -band and the 50% decrease of exchange splitting compared to the LDA results. Note that the satellite in Ni has substantially more spin-up contributions, in agreement with photoemission spectra [77]. The exchange splitting of the d -band depends very weakly on temperature from $T = 0.6 T_C$ to $T = 0.9 T_C$. Correlation effects in Fe are less pronounced than in Ni, due to its large spin-splitting and the characteristic bcc -structural dip in the density of states for spin-down

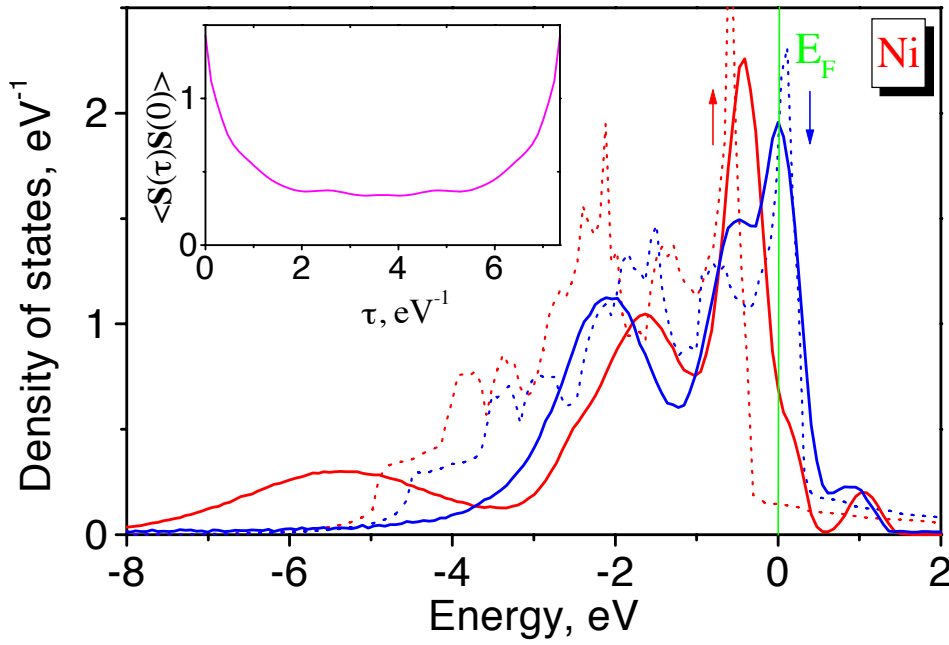


Fig. 15: LDA+DMFT results for ferromagnetic nickel ($T = 0.9 T_C$). The partial densities of the d -states (full lines) is compared with the corresponding LDA results at zero temperature (dashed lines) for the spin-up (red lines, arrow-up) and spin-down (blue lines, arrow-down) states. The insert shows the spin-spin autocorrelation function for $T = 1.8 T_C$.

states near the Fermi level that reduces the density of states for particle hole excitations.

Now we discuss the application of the LDA+DMFT approach to the description of finite-temperature magnetic properties of iron and nickel. While density functional theory can, in principle, provide a rigorous description of the thermodynamic properties, at present there is no accurate practical implementation available. As a result the finite-temperature properties of magnetic materials are estimated following a simple suggestion [57], whereby constrained DFT at $T = 0$ is used to extract exchange constants for a *classical* Heisenberg model, which in turn is solved using approximation methods (e.g., RPA, mean field) from classical statistical mechanics of spin systems [57, 78–80]. The most recent implementation of this approach gives good values for the transition temperature of iron but not of nickel [81]. While these localized spin models give, by construction, at high temperatures a Curie-Weiss like magnetic susceptibility, as observed experimentally in Fe and Ni, they encountered difficulties in predicting the correct values of the Curie constants [82].

The uniform spin susceptibility in the paramagnetic state: $\chi_{q=0} = dM/dH$ was extracted from QMC simulations by measuring the induced magnetic moment in a small external magnetic field. The dynamical mean-field results account for the Curie-Weiss law which is observed experimentally in Fe and Ni. As the temperature increases above T_C , the atomic character of the system is partially restored resulting in an atomic-like susceptibility with effective moment

$$\chi_{q=0} = \frac{\mu_{\text{eff}}^2}{3(T - T_C)} \quad (59)$$

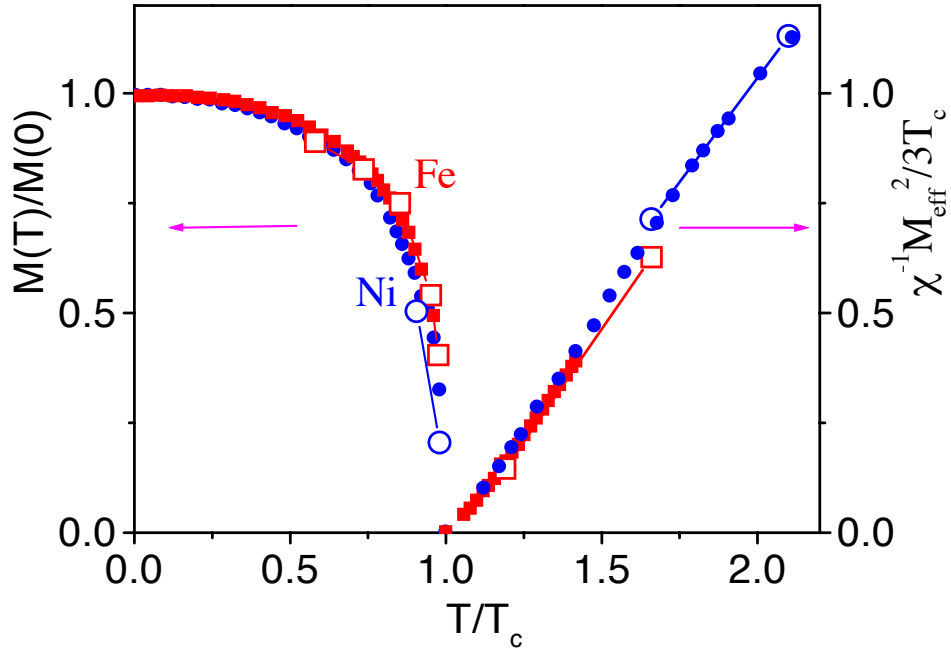


Fig. 16: Temperature dependence of ordered moment and inverse ferromagnetic susceptibility for Fe (open squares) and Ni (open circles) compared with experimental results (filled symbols).

The temperature dependence of the ordered magnetic moment below the Curie temperature and the inverse of the uniform susceptibility above the Curie point are plotted in Fig. 16 together with the corresponding experimental data for iron and nickel [83]. The LDA+DMFT calculations describe the magnetization-curve and the slope of the high-temperature Curie-Weiss susceptibility remarkably well. The calculated values of high-temperature magnetic moments extracted from the uniform spin susceptibility are $\mu_{\text{eff}} = 3.09$ (1.50) μ_B for Fe (Ni), in good agreement with the experimental data $\mu_{\text{eff}} = 3.13$ (1.62) μ_B for Fe (Ni) [83].

One can estimate the value of the Curie temperature of Fe and Ni from the disappearance of spin polarization in the self-consistent DMFT solution and from the Curie-Weiss law in Eq. (59). Our estimates $T_C = 1900$ (700) K are in reasonable agreement with experimental values of 1043 (631) K for Fe (Ni) respectively [83], considering the single-site nature of the DMFT approach, which is not able to capture the reduction of T_C due to long wavelength spin waves. These effects are governed by the spin wave stiffness. Since the ratio T_C/a^2D of the spin wave stiffness D to T_C is nearly a factor of 3 larger for Fe than for Ni [83] (a is the lattice spacing), we expect the Curie temperature from DMFT to be much higher than the observed T_C in Fe than in Ni. Note that this is a consequence of the long-range oscillating character of exchange interactions in iron compared to short-range ferromagnetic exchange interactions in nickel [81]. Quantitative calculations demonstrating the sizable reduction of T_C due to spin waves in Fe in the framework of a Heisenberg model were performed in Ref [81]. Moreover including additional spin-flip terms in the rotationally invariant Coulomb matrix will drastically reduce the effective Curie temperature for the case of iron with its approximately two unpaired electrons [88].

Within dynamical mean-field theory one can also compute the local spin susceptibility

$$\chi_{\text{loc}} = \frac{g_s^2}{3} \int_0^\beta d\tau \langle \mathbf{S}(\tau) \mathbf{S}(0) \rangle, \quad (60)$$

where $g_s = 2$ is the gyromagnetic ratio, $\mathbf{S} = \frac{1}{2} \sum_{m,\sigma,\sigma'} c_{m\sigma}^\dagger \vec{\sigma}_{\sigma\sigma'} c_{m\sigma'}$ the single-site spin operator, and $\vec{\sigma} = (\sigma_x, \sigma_y, \sigma_z)$ are the Pauli matrices. It differs from the $\mathbf{q} = \mathbf{0}$ susceptibility by the absence of spin polarization in the Weiss field of the impurity model. Eq. (60) cannot be probed directly in experiments but it is easily computed in DMFT-QMC. Its behavior as a function of temperature gives a very intuitive picture of the degree of correlations in the system. In a weakly correlated system we expect Eq. (60) to be nearly temperature-independent, while in a strongly correlated system we expect a leading Curie-Weiss behavior at high temperatures $\chi_{\text{loc}} = \mu_{\text{loc}}^2 / (3T + C)$, where μ_{loc} is an effective local magnetic moment. In the Heisenberg model with spin S , $\mu_{\text{loc}}^2 = S(S+1)g_s^2$ and for well-defined local magnetic moments (e.g., for rare-earth magnets) this quantity should be temperature-independent. For the itinerant electron magnets μ_{loc} is temperature-dependent, due to a variety of competing many body effects such as Kondo screening and the reduction of local magnetic moment by temperature [27]. All these effects are included in the DMFT calculations. The τ -dependence of the correlation function $\langle \mathbf{S}(\tau) \mathbf{S}(0) \rangle$ results in the temperature dependence of μ_{loc} and is displayed in the inserts on the Figs. 14,15. Iron can be considered as a magnet with very well-defined local moments above T_C (the τ -dependence of the correlation function is relatively weak), whereas nickel is a more itinerant electron magnet (stronger τ -dependence of the local spin-spin autocorrelation function).

The comparison of the values of the local and the $\mathbf{q} = \mathbf{0}$ susceptibility gives a crude measure of the degree of short range order which is present above T_C . As expected, the moments extracted from the local susceptibility, Eq. (60), are a bit smaller ($2.8 \mu_B$ for iron and $1.3 \mu_B$ for nickel) than those extracted from the uniform magnetic susceptibility. This reflects the small degree of short-range correlations that remains well above T_C [85]. The high-temperature LDA+DMFT data clearly show the presence of a local-moment above T_C . This moment is correlated with the presence of high energy features (of the order of the Coulomb energies) in the photoemission. This is also true below T_C , where the spin dependence of the spectra is more pronounced for the satellite region in nickel than for the quasiparticle bands near the Fermi level (Fig. 15). This can explain the apparent discrepancies between different experimental determinations of the high-temperature magnetic splittings [84, 86, 87] as being the result of probing different energy regions. The resonant photoemission experiments [86] reflect the presence of local-moment polarization in the high-energy spectrum above T_C in nickel, while the low-energy ARPES investigations [87] result in non-magnetic bands near the Fermi level. This is exactly the DMFT view on the electronic structure of transition metals above T_C . Fluctuating moments and atomic-like configurations are large at short times, which results in correlation effects in the high-energy spectra such as spin-multiplet splittings. The moment is reduced at longer time scales, corresponding to a more band-like, less correlated electronic structure near the Fermi level.

8 Conclusions

We have discussed a general scheme for investigating the magnetic properties for correlated itinerant-electron systems. This approach is based on the combination of the first-principle LDA scheme with dynamical mean-field theory. The application of the LDA+DMFT method gives an adequate description of the quasiparticle electronic structure of ferromagnetic transition metals. The main correlation effects in the electron energy spectrum are a strong damping of the occupied states more than 1 eV from the Fermi level E_F and essentially a depolarization of the states in the vicinity of E_F . We obtain a reasonable agreement with different experimental spectral data (spin-polarized photo- and thermo-emission). The method is quite universal and can be applied for other correlated d - and f -electron magnetic systems.

We discussed as well a general method for the investigation of magnetic interactions in correlated electron systems. Our general expressions are also valid in the relativistic case and can be used for the calculation of both exchange and Dzyaloshinskii-Moriya interactions, and magnetic anisotropy [38]. The illustrative example of ferromagnetic iron shows that correlation effects in the exchange interactions may be noticeable even in such moderately correlated systems. For rare-earth metals and their compounds, colossal magnetoresistance materials, or high- T_c systems, this effect may be even more important.

This work demonstrates an essential difference between the spin density functional and the LDA+DMFT approach. The latter deals with the thermodynamic potential as a functional of the local Green function rather than the electron density. Nevertheless, there is a close connection between the two techniques (the self-energy corresponds to the exchange-correlation potential). In particular, an analog of the local force theorem can be proved within the LDA+DMFT approach. It may be useful not only for the calculation of magnetic interactions but also for elastic stresses, in particular pressure, and other physical properties of correlated magnetic systems.

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