Spin and orbital exchange interactions from Dynamical Mean Field Theory

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ABSTRACT

We derive a set of equations expressing the parameters of the magnetic interactions characterizing a strongly correlated electronic system in terms of single-electron Green's functions and self-energies. This allows to establish a mapping between the initial electronic system and a spin model including up to quadratic interactions between the effective spins, with a general interaction (exchange) tensor that accounts for anisotropic exchange, Dzyaloshinskii–Moriya interaction and other symmetric terms such as dipole–dipole interaction. We present the formulas in a format that can be used for computations via Dynamical Mean Field Theory algorithms.

1. Introduction

Describing a solid in terms of its magnetic properties requires the knowledge of an effective spin model which displays the same interesting physical properties as the many-electron Hamiltonian whose exact solution would give the complete description of the system. The determination of the form of the effective spin model and of the strength of the interactions between the constituent spins starting from the initial electronic model is, in general, a complicated many-body problem [1–12].

We have recently derived expressions for the parameters of the magnetic interactions within an extended (multi-orbital) Hubbard model [12], in the presence of arbitrary relativistic couplings affecting the electronic degrees of freedom (such as spin–orbit, magnetic anisotropy, Zeeman coupling with an external magnetic field). The formulas presented in Ref. [12], after neglecting the vertices of two-electron Green's functions, are expressed in terms of single-electron (but fully interacting) Green's functions G and the single-electron (hopping) Hamiltonian T. The use of the representation via T [9,12] for computations related to real materials requires the additional step of a tight-binding parametrization, which is implemented only in some methods of electronic structure calculations. On the other hand, a presentation of the formulas in

terms of Green's functions G and self-energies Σ would make them more suitable for implementation via Dynamical Mean Field Theory (DMFT) [13–15], since *any* DMFT calculation deals with G and Σ. Writing the parameters in a way that explicitly exhibits self-energies, analogous to what was done in Refs. [4,5,10], also allows to explicitly include the approximation of local self-energy, which is the key assumption of DMFT. We here present the adaptation of the formulas for the exchange tensor to this scheme.

2. Method and discussion

We consider the extended multi-orbital Hubbard Hamiltonian [16–20,12],

$$\hat{H} = \sum_{o,\sigma,m} \sum_{o',\sigma',m'} \hat{\phi}_{o,\sigma,m}^{\dagger} T_{o',\sigma',m'}^{o,\sigma,m} \hat{\phi}^{o',\sigma',m'} + \hat{H}_V, \tag{1}$$

where the field operator $\hat{\phi}_{o,\sigma,m}^{\dagger}$ creates an electron with quantum numbers $\{o, \sigma, m\}$: o refers to a set of the orbital indices (for a basis of localized Wannier wave functions,[1] these are the atom index a,

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[1] A different choice of the basis set of single-electron wave functions is possible, but our formalism is directly applicable only if the interaction term is rotationally invariant under spin rotations performed in the new basis (otherwise, the rotationally variant terms of the interaction should be included in the computation of the magnetic parameters).

the principal atomic quantum number n and the angular momentum quantum number l: $o \equiv \{a, n, l\}$), while $\sigma \in \{\uparrow, \downarrow\}$ and $m \in \{-l, -l+1, ..., l\}$ are the third components of the intrinsic-spin and orbital angular momenta, respectively. Local angular momenta are measured with respect to local reference frames, which depend on o and might not be collinear [12]. The single-particle Hamiltonian matrix $T_{o',\sigma',m'}^{o,\sigma,m}$ is completely arbitrary, so it can include any relativistic single-electron terms (Zeeman coupling, spin-orbit, magnetic anisotropies). The interaction Hamiltonian $\hat{H}_V$ is assumed to be rotationally invariant [12].

The goal in Ref. [12] was to map the model given by Eq. (1) onto an effective model of classical spins $\boldsymbol{e}_o$ including up to (arbitrary) quadratic interactions, with Hamiltonian

$$H_{\text{spin}} = \sum_o \boldsymbol{e}_o \cdot \mathcal{B}_o + \frac{1}{2} \sum_{o,o'} \sum_{\alpha,\alpha'} e_{o,\alpha} e_{o',\alpha'} \mathcal{H}_{oo'}^{\alpha\alpha'}, \tag{2}$$

determined by the exchange tensor $\mathcal{H}_{oo'}^{\alpha\alpha'} = \mathcal{H}_{o'o}^{\alpha'\alpha}$ (here and in the following α and α' are used to denote the space coordinates, e.g. x, y, z) and the effective magnetic field $\mathcal{B}_o$. It is convenient to decompose the exchange tensor into the three vectors $\mathcal{J}_{oo'} = \mathcal{J}_{o'o}$ (anisotropic exchange), $\mathcal{D}_{oo'} = -\mathcal{D}_{o'o}$ (Dzyaloshinskii–Moriya interaction), and $C_{oo'} = C_{o'o}$ (symmetric non-diagonal exchange), defined as

$$\mathcal{J}_{oo'}^{\alpha} \equiv \mathcal{H}_{oo'}^{\alpha\alpha}, \quad \mathcal{D}_{oo'}^{\alpha} \equiv \frac{1}{2} \sum_{\alpha'\alpha''} \varepsilon^{\alpha\alpha'\alpha''} \mathcal{H}_{oo'}^{\alpha'\alpha''}, \quad C_{oo'}^{\alpha} \equiv \frac{1}{2} \sum_{\alpha'\alpha''} \left| \varepsilon^{\alpha\alpha'\alpha''} \right| \mathcal{H}_{oo'}^{\alpha'\alpha''}, \tag{3}$$

where $\varepsilon^{\alpha\alpha'\alpha''}$ is the completely anti-symmetric tensor of rank 3. The Heisenberg model is obtained as the particular case in which $\mathcal{H}_{oo'}^{\alpha\alpha'} \equiv \delta^{\alpha\alpha'} \mathcal{J}_{oo'}$.

To perform the mapping, in Ref. [12] we have derived the response of the thermodynamic potential of the electronic system under small spatially dependent rotations of the spin quantization axes associated with each orbital spinor denoted by o, up to second order in the rotation angles. The derivation of such response involves path integration over the fermionic fields after the introduction of auxiliary bosonic degrees of freedom which express the amplitudes of rotations from an initial spin configuration; the coefficients of the interactions between the remaining bosons are put in correspondence with the parameters of the spin model (2) by imposing that the thermodynamic potential of the spin system after the spin rotations is equal to that of the electrons. Excluding the vertex contributions, the parameters of the spin model are expressed in terms of single-electron Green's functions (which of course include interaction effects) and the single-particle part of the electronic Hamiltonian, T.

This procedure is similar to the one previously adopted in Refs. [4,5] for the case of quenched orbital moments, but in Ref. [12] we have considered rotations of the local total spins $\hat{\boldsymbol{s}}_o = \hat{\boldsymbol{l}}_o + \hat{\boldsymbol{s}}_o$, where $\hat{\boldsymbol{l}}_o$ and $\hat{\boldsymbol{s}}_o$ are, respectively, the orbital and intrinsic angular momenta associated with the states o. More precisely, we have considered rotations in the space of the single-particle eigenfunctions of $\hat{\boldsymbol{s}}_o^2$ and $\hat{s}_o^z$, analogous to Ref. [9], while in Refs. [4,5] the rotations affected the space of eigenfunctions of $\hat{\boldsymbol{s}}_o^2$ and $\hat{s}_o^z$. This allowed us to obtain formulas for the exchange tensor that can be separated into contributions coming from the interactions between spin–spin, orbital–orbital, or spin–orbital degrees of freedom of the electrons; for example, for the anisotropic exchange parameters we have

$$\mathcal{J}_{oo'} \equiv \mathcal{J}_{oo'}^{\text{spin–spin}} + \mathcal{J}_{oo'}^{\text{orb–orb}} + \mathcal{J}_{oo'}^{\text{spin–orb}}. \tag{4}$$

The terms labelled as "spin–spin" or "spin" are those contributions to magnetic interactions that would arise if we kept the orbital

magnetic moments quenched, i.e., if we rotated only the intrinsic-spin spinors. Analogously, the terms labelled as "orb" or "orb–orb" arise if we rotate only the orbital magnetic moments, keeping the intrinsic spins quenched. The terms labelled as "spin–orb" arise only when the total local magnetic moments are rotated; these terms should not be confused with spin–orbit coupling, which contributes in general to all terms.

It should be noted that the possibility of rotating the total local spins is not applicable within Density Functional Theory (DFT) formulations, where observables are expressed in terms of the charge density and the intrinsic-spin density. The possibility of rotating local total spins is related to the representation of the electronic Hamiltonian in terms of localized wave functions, which implies a higher number of degrees of freedom with respect to DFT (related to the fact that the set of localized states would be over-complete in theory, or not even complete in practice due to truncation).

The computation of the magnetic parameters via DMFT is greatly simplified if they are formulated in terms of single-particle Green's functions and self-energies Σ in magnetically ordered states, since this avoids the initial step of a tight-binding parameterization of the single-electron Hamiltonian T. To remove T and introduce Σ, we use the equations of motion for Matsubara Green's functions (Dyson equations), which we write in general matrix notation as

$$(\omega - i\mu)G(i\omega) + iT \cdot G(i\omega) = 1 - \Sigma(i\omega) \cdot G(i\omega),$$
$$(\omega - i\mu)G(i\omega) + iG(i\omega) \cdot T = 1 - G(i\omega) \cdot \Sigma(i\omega). \tag{5}$$

These equations hold for the Matsubara Green's functions defined according to the following convention:

$$G_2^1(\tau) \equiv -i \left\langle \mathcal{T} \hat{\psi}^1(\tau)\, \hat{\psi}_2^\dagger \right\rangle \equiv \frac{1}{\beta} \sum_\omega G_2^1(i\omega) e^{-i\omega\tau}, \tag{6}$$

where $\omega = (2n+1)\pi/\beta$ is a fermionic Matsubara frequency, β being the inverse temperature. As a particular case, the single-electron density matrix is given by

$$\rho \equiv -i\, G(\tau = 0^-) = -i \frac{1}{\beta} \sum_\omega e^{i\omega 0^+} G(i\omega). \tag{7}$$

We now have to distinguish between the magnetic parameters that can be computed from the second-order response in the rotation angles and those which are computed from the first-order response. From Ref. [12], we note that the former terms can all be written in terms of the following quantity:

$$F_{o\alpha,o'\alpha'}^{XY} \equiv -\delta_{oo'} \frac{1}{2} \text{Tr}_{m,\sigma} \left(\{S_{o\alpha}^X; S_{o\alpha'}^Y\} \cdot \{\rho; T\}_o^o \right)$$
$$+ \text{Tr}_{m,\sigma} \left(S_{o\alpha}^X \cdot T_o^{o'} \cdot S_{o'\alpha'}^Y \cdot \rho_o^{o'} + S_{o'\alpha'}^Y \cdot T_o^{o'} \cdot S_{o\alpha}^X \cdot \rho_{o'}^o \right)$$
$$+ \frac{1}{\beta} \sum_\omega e^{i\omega 0^+} \text{Tr}_{m,\sigma} \Big\{ S_{o\alpha}^X \cdot [G(i\omega) \cdot T]_{o'}^o \cdot S_{o'\alpha'}^Y \cdot [G(i\omega) \cdot T]_o^{o'}$$
$$- S_{o\alpha}^X \cdot G(i\omega)_o^{o'} \cdot S_{o'\alpha'}^Y \cdot [T \cdot G(i\omega) \cdot T]_o^{o'}$$
$$- S_{o\alpha}^X \cdot [T \cdot G(i\omega) \cdot T]_{o'}^o \cdot S_{o'\alpha'}^Y \cdot G(i\omega)_o^{o'}$$
$$+ S_{o\alpha}^X \cdot [T \cdot G(i\omega)]_{o'}^o \cdot S_{o'\alpha'}^Y \cdot [T \cdot G(i\omega)]_o^{o'} \Big\}, \tag{8}$$

where $X, Y \in \{\text{spin, orb}\}$ refer to either spin- or orbital-related terms, that is,

$$S_{o\alpha}^{\text{spin}} \equiv s_{o\alpha} \equiv \frac{1}{2}\sigma_{o\alpha}, \quad S_{o\alpha}^{\text{orb}} \equiv l_{o\alpha}, \tag{9}$$

where $s_{o\alpha}$ is an intrinsic spin matrix ($\sigma_{o\alpha}$ is a Pauli matrix), while $l_{o\alpha}$ is an orbital angular momentum matrix. In Eq. (8) we have used