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Electronic Structure

First-principles calculation of the parameters used by atomistic magnetic simulations

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Abstract

While the ground state of magnetic materials is in general well described on the basis of spin density functional theory (SDFT), the theoretical description of finite-temperature and non-equilibrium properties require an extension beyond the standard SDFT. Time-dependent SDFT (TD-SDFT), which give for example access to dynamical properties are computationally very demanding and can currently be hardly applied to complex solids. Here we focus on the alternative approach based on the combination of a parameterized phenomenological spin Hamiltonian and SDFT-based electronic structure calculations, giving access to the dynamical and finite-temperature properties for example via spin-dynamics simulations using the Landau–Lifshitz–Gilbert (LLG) equation or Monte Carlo simulations. We present an overview on the various methods to calculate the parameters of the various phenomenological Hamiltonians with an emphasis on the KKR Green function method as one of the most flexible band structure methods giving access to practically all relevant parameters. Concerning these, it is crucial to account for the spin–orbit coupling (SOC) by performing relativistic SDFT-based calculations as it plays a key role for magnetic anisotropy and chiral exchange interactions represented by the DMI parameters in the spin Hamiltonian. This concerns also the Gilbert damping parameters characterizing magnetization dissipation in the LLG equation, chiral multispin interaction parameters of the extended Heisenberg Hamiltonian, as well as spin–lattice interaction parameters describing the interplay of spin and lattice dynamics processes, for which an efficient computational scheme has been developed recently by the present authors.

1. Introduction

Density functional theory (DFT) is a 'formally exact approach to the static electronic many-body problem' for the electron gas in the equilibrium, which was adopted for a huge number of investigations during the last decades to describe the ground state of solids, both magnetic and non-magnetic, as well as various ground state properties [1].

However, dealing with real systems, the properties in an out-of-equilibrium situation are of great interest. An example for this is the presence of external perturbation varying in time, which could be accounted for by performing time-dependent first-principles electronic structure calculations. The time-dependent extension of density functional theory (TD-DFT) [2] is used successfully to study various dynamical processes in atoms and molecules, in particular, giving access to the time evolution of the electronic structure in a system affected by a femtosecond laser pulse. However, TD-DFT can be hardly applied to complex solids because of the lack of universal parameter-free approximations for the exchange-correlation kernel. Because of this, an approach based on the combination of simulation methods for spin- and lattice dynamics, using model spin and lattice Hamiltonians is more popular for the moment. A great progress with this approach has been achieved during last decade due to the availability of parameters for the model Hamiltonians calculated on a first principles level, that is a central issue of the present contribution. As it was pointed out in reference [1], this approach has the advantage, that the spin-related many-body effects in this case are much simpler to be taken into account

when compared to the *ab initio* approach. Thus, the isotropic exchange coupling parameters J_{ij} for the classical Heisenberg Hamiltonian worked out Liechtenstein *et al* [3, 4] have been successfully used by many authors to predict the ground state magnetic structure of material and to investigate its finite-temperature properties. Depending on the materials, the isotropic J_{ij} can exhibit only spatial anisotropy.

1.1. Models and Hamiltonians

Extension of the Heisenberg Hamiltonian accounting for anisotropy in spin subspace is often done by adding the so-called Dzyaloshinskii–Moriya interactions (DMIs) and the magnetic anisotropy term,

$$H_{H,\mathrm{rel}} = -\sum_{i,j} J_{ij}(\hat{e}_i \cdot \hat{e}_j) - \sum_{i,j} \vec{D}_{ij}(\hat{e}_i \times \hat{e}_j) + \sum_{i} \hat{e}_i \underline{K}_{ii} \hat{e}_i \tag{1}$$

with $\hat{e}_{i(j)}$ the orientation of the spin magnetic moment at site $i(j)$. Alternatively, one may describe exchange interactions in the more general tensorial form, $\underline{J}_{ij}$, leading to:

$$H_{H,\mathrm{rel}} = -\sum_{i,j} \hat{e}_i \underline{J}_{ij} \hat{e}_j + \sum_{i} \hat{e}_i \underline{K}_{ii} \hat{e}_i. \tag{2}$$

In the second case the DMI is represented as the antisymmetric part of the exchange tensor, i.e. $D_{ij}^{\alpha} = \frac{1}{2}(J_{ij}^{\beta\gamma} - J_{ij}^{\gamma\beta})\epsilon_{\alpha\beta\gamma}$. It should be stressed, that calculations of the spin-anisotropic exchange interaction parameters as well as of the magnetic anisotropy parameters require a relativistic treatment of the electronic structure in contrast to the case of the isotropic exchange parameters which can be calculated on a non-relativistic level. Various schemes to map the dependence of the electronic energy on the magnetic configuration were suggested in the literature to calculate the parameters of the spin Hamiltonians [5–8], depending of its form given in equations (1) or (2).

Despite of its simplicity, the spin Hamiltonian gives access to a reasonable description of the temperature dependence of magnetic properties of materials when combined with Monte Carlo (MC) simulations [9], or non-equilibrium spin dynamics simulations based on the phenomenological Landau–Lifshitz–Gilbert (LLG) equations [10, 11]

$$\frac{1}{\gamma} \frac{\mathrm{d}\vec{M}}{\mathrm{d}\tau} = -\vec{M} \times \vec{H}_{\mathrm{eff}} + \vec{M} \times \left[\frac{\tilde{G}(\vec{M})}{\gamma^2 M_s^2} \frac{\mathrm{d}\vec{M}}{\mathrm{d}\tau} \right]. \tag{3}$$

Here $\vec{H}_{\mathrm{eff}}$ is the effective magnetic field defined as $\vec{H}_{\mathrm{eff}} = -\frac{1}{M} \frac{\partial F}{\partial \hat{m}}$, where F is the free energy of the system and $\hat{m} = \frac{\vec{M}}{M_s}$ with M_s the saturation magnetization treated at first-principles level, and γ is the gyromagnetic ratio and $\tilde{G}$ is the Gilbert damping parameter. Alternatively, the effective magnetic field can be represented in terms of the spin Hamiltonian in equation (2), i.e. $\vec{H}_{\mathrm{eff}} = -\frac{1}{M} \frac{\partial \langle \mathcal{H}_{H,\mathrm{rel}} \rangle_T}{\partial \hat{m}}$, with $\langle \ldots \rangle_T$ denoting the thermal average for the extended Heisenberg Hamiltonian $\mathcal{H}_{H,\mathrm{rel}}$.

The first-principles calculation of the parameters for the Heisenberg Hamiltonian as well as for the LLG equation for spin dynamics have been reported in the literature by various groups who applied different approaches based on *ab initio* methods [3–5, 7, 12–27]. Here we will focus on calculations based on the Green function multiple-scattering formalism being a rather powerful tool to supply all parameters for the extended Heisenberg Hamiltonian as well as for the LLG equation.

1.2. Variables and units

In this section we list in table 1 the variables and parameters which show up in the review. Conventionally, the calculated parameters of the spin Hamiltonian are given in electronvolts (eV), while the unit Bohr magneton (μ_B) is used for atomic magnetic moments, and the Bohr radius (a_0) is used for interatomic distances. The dimensionless Gilbert damping parameter is defined as $\alpha^G = \tilde{G}/(\gamma M_s)$. The values for the basic constants listed in table 1 are given in the SI units using in particular Tesla (T) for the magnetic field, Kelvin (K) for the temperature, and Joule (J) for the energy.

2. Magnetic anisotropy

Let us first consider the magnetic anisotropy term in spin Hamiltonian, characterized by parameters (written in tensorial form in equations (1) and (2)) deduced from the total energy dependent on the orientation of the magnetization $\hat{m}$. The latter is traditionally split into the magneto-crystalline anisotropy (MCA) energy, $E_{\mathrm{MCA}}(\hat{m})$, induced by spin–orbit coupling (SOC) and the shape anisotropy energy, $\Delta E_{\mathrm{shape}}(\hat{m})$, caused by magnetic dipole interactions,

$$E_A(\hat{m}) = E_{\mathrm{MCA}}(\hat{m}) + \Delta E_{\mathrm{shape}}(\hat{m}). \tag{4}$$

Table 1. Key variables and parameters.

Variable	Symbol	Unit
Lattice parameter	a	a_0
Wave vector	q	$2\pi a_0^{-1}$
Interatomic distance	R_{ij}	a_0
Magnetic anisotropy constants	κ_l^m	eV
Exchange coupling parameters	J_{ij}	eV
DMI	$\vec{D}_{ij}$	eV
Three-spin chiral interaction (TCI)	J_{ijk}	eV
Four-spin isotropic		
Exchange interaction	J_{ijkl}^s	eV
Six-spin exchange parameters	$\kappa_{ijklmn}^{\text{six-spin}}$	eV
DMI-like four-spin		
Exchange interactions	$\vec{D}_{ijkl}$	eV
Spin–lattice coupling parameters	$\mathcal{J}_{ij,k}$	eV/a_0
Magnetic moment per atom	$|\vec{M}_i|$	μ_B
Magnetization	$|\vec{M}|$	μ_B/f.u.
Temperature	T	K
Gilbert damping parameter	α^G	Dimensionless

Parameter	Symbol	Value
Bohr magneton	μ_B	9.27401×10^{-24} J T^{-1}
Vacuum permeability	μ_0	$4\pi \times 10^{-7}$ T^2 m^3 J^{-1}
Gyromagnetic ratio	γ	1.76086×10^{11} T^{-1} s^{-1}
Bohr radius	a_0	5.29177×10^{-11} m
electronvolt	eV	1.60219×10^{-19} J

Although a quantum-mechanical description of the magnetic shape anisotropy deserves separate discussion [28] this contribution can be reasonably well estimated based on classical magnetic dipole–dipole interactions. Therefore, we will focus on the MCA contribution which is fully determined by the electronic structure of the considered system. In the literature the focus is in general on the MCA energy of the ground state, which can be estimated straightforwardly from the total energy calculated for different orientations of the magnetization followed by a mapping onto a model spin Hamiltonian, given e.g. by an expansion in terms of spherical harmonics $Y_{lm}(\hat{m})$ [29]

$$E_{\text{MCA}}(\hat{m}) = \sum_{l \text{ even}} \sum_{m=-l}^{m=l} \kappa_l^m \, Y_{lm}(\hat{m}). \tag{5}$$

Alternative approach to calculate the MCA parameters is based on magnetic torque calculations, using the definition

$$T^{\hat{m}}(\theta^{\hat{u}}) = -\frac{\partial E(\hat{m})}{\partial \theta^{\hat{u}}}, \tag{6}$$

avoiding the time-consuming total energy calculations. This scheme is based on the so-called magnetic force theorem (MFT) that allows to represent the MCA energy in terms of a corresponding electronic single-particle energies change under rotation of magnetization, as follows [30]:

$$\Delta\mathcal{E}_{\text{SOC}}(\hat{m}, \hat{m}') = -\int^{E_F^{\hat{m}}} dE \left[N^{\hat{m}}(E) - N^{\hat{m}'}(E) \right] - \frac{1}{2} n^{\hat{m}'}(E_F^{\hat{m}'}) (E_F^{\hat{m}} - E_F^{\hat{m}'})^2 + \mathcal{O}(E_F^{\hat{m}} - E_F^{\hat{m}'})^3 \tag{7}$$

with $N^{\hat{m}}(E) = \int^E dE' \, n^{\hat{m}}(E')$ the integrated DOS for the magnetization along the direction $\hat{m}$, and $n^{\hat{m}}(E)$ the density of states (DOS) represented in terms of the Green function as follows

$$n^{\hat{m}}(E) = -\frac{1}{\pi} \text{Im} \, \text{Tr} \, G^{\hat{m}}(E). \tag{8}$$

This expression can be used in a very efficient way within the framework of the multiple-scattering formalism. In this case the Green function is given in terms of the scattering path operator $\underline{\tau}(E)^{nn'}$ connecting the sites n and n' as follows

$$G_0(\vec{r}, \vec{r}', E) = \sum_{\Lambda\Lambda'} Z_\Lambda^n(\vec{r}, E) \, \tau_{\Lambda\Lambda'}^{nn'}(E) \, Z_{\Lambda'}^{n'\times}(\vec{r}', E)$$

$$- \sum_\Lambda \left[Z_\Lambda^n(\vec{r}, E) J_\Lambda^{n\times}(\vec{r}', E) \, \Theta(r' - r) + J_\Lambda^n(\vec{r}, E) Z_\Lambda^{n\times}(\vec{r}', E) \, \Theta(r - r') \right] \delta_{nn'}, \tag{9}$$

where the combined index $\Lambda = (\kappa, \mu)$ represents the relativistic spin–orbit and magnetic quantum numbers κ and μ, respectively [31]; $Z_\Lambda^n(\vec{r}, E)$ and $J_\Lambda^n(\vec{r}, E)$ are the regular and irregular solutions of the single-site Dirac equation (28) [32–34]. The scattering path operator is given by the expression

$$\underline{\underline{\tau}}(E) = [\underline{\underline{m}}(E) - \underline{\underline{G}}_0(E)]^{-1} \tag{10}$$

with $\underline{\underline{m}}(E) = \underline{\underline{t}}^{-1}(E)$ and $\underline{\underline{G}}_0(E)$ the inverse single-site scattering and structure constant matrices, respectively. The double underline used here indicates matrices with respect to site and angular momentum indices [33].

Using the Lloyd's formula that gives the integrated DOS in terms of the scattering path operator, equation (7) can be transformed to the form

$$\Delta\mathcal{E}_{\text{SOC}}(\hat{m}, \hat{m}') = -\frac{1}{\pi} \operatorname{Im} \operatorname{Tr} \int^{E_{\text{F}}} dE \left(\ln \underline{\underline{\tau}}(\hat{m}, E) - \ln \underline{\underline{\tau}}(\hat{m}', E) \right) \tag{11}$$

with the scattering path operator evaluated for the magnetization along $\hat{m}$ and $\hat{m}'$, respectively.

With this, the magnetic torque $T(\theta)$ can be expressed by means of multiple scattering theory leading for the torque component with respect to a rotation of the magnetization around an axis $\hat{u}$, to the expression [35]

$$T^{\hat{m}}(\theta^{\hat{u}}) = -\frac{1}{\pi} \Im \int^{E_{\text{F}}} dE \frac{\partial}{\partial\theta^{\hat{u}}} \left[\ln \det\left(\underline{t}(\hat{m})^{-1} - \underline{G}^0\right) \right]. \tag{12}$$

Mapping the resulting torque onto a corresponding parameterized expression as for example equation (5), one obtains the corresponding parameters of the spin Hamiltonian.

However, one should note that the magnetic anisotropy of materials changes when the temperature increases. This occurs first of all due to the increasing amplitude of thermally induced spin fluctuations responsible for a modification of the electronic structure. A corresponding expression for magnetic torque st finite temperature was worked out by Staunton *et al* [35], on the basis of the relativistic generalization of the disordered local moment (RDLM) theory [36]. To perform the necessary thermal averaging over different orientational configurations of the local magnetic moments it uses a technique similar to the one used to calculate the configurational average in the case of random metallic alloys, so-called coherent potential approximation (CPA) alloy theory [37, 38]. Accordingly, the free energy difference for two different orientations of the magnetization is given by

$$\Delta\mathcal{F}(\hat{m}, \hat{m}') = -\int dE f_{\text{FD}}(E, \hat{m}) \tag{13}$$

$$\left[\langle N^{\hat{m}} \rangle(E) - \langle N^{\hat{m}'} \rangle(E) \right]. \tag{14}$$

By using in this expression the configurational averaged integrated density of states [36, 39] given by Lloyd's formula, the corresponding expression for the magnetic torque at temperature T

$$T^{\hat{m}, T}(\theta^{\hat{u}}) = -\frac{\partial}{\partial\theta^{\hat{u}}} \left(\sum_i \int P_i^{\hat{m}}(\hat{e}_i) \langle \Omega^{\hat{m}} \rangle_{\hat{e}_i} d\hat{e}_i \right) \tag{15}$$

can be written explicitly as:

$$T^{\hat{m}, T}(\theta^{\hat{u}}) = -\frac{1}{\pi} \operatorname{Im} \int^{E_{\text{F}}} dE f_{\text{FD}}(E, \hat{m}) \left(\sum_i \int \frac{\partial P_i^{\hat{m}}(\hat{e}_i)}{\partial\theta^{\hat{u}}} \ln \det M_i^{\hat{m}}(\hat{e}_i, E) d\hat{e}_i \right). \tag{16}$$

where

$$\underline{M}_i^{\hat{m}}(\hat{e}_i, E) = 1 + ([\underline{t}_i(\hat{e}_i)]^{-1} - \underline{t}_{i,c}^{\hat{m}}(\hat{e}_i)]^{-1}) \underline{\tau}_{ii,c}^{\hat{m}}, \tag{17}$$

and

$$\underline{\tau}_{ii,c}^{\hat{m}} = ([\underline{t}_{i,c}^{\hat{m}}(\hat{e}_i)]^{-1} - \underline{G}_0)^{-1} \tag{18}$$

where the index c indicates quantities related to the CPA medium.

Figure 1 (top) shows as an example the results for the temperature-dependent magnetization ($M(T)$) calculated within the RDLM calculations for $L1_0$-ordered FePt [40]. Figure 1 (bottom) gives the corresponding parameter $K(T)$ for a uni-axial magneto-crystalline anisotropy, which is obviously in good agreement with experiment.

To complete the discussion on the magnetic anisotropy, one has to mention the shape anisotropy energy ΔE_{shape}, which may be a leading contribution in the case of thin magnetic films, favoring an in-plane orientation of the magnetization. Its origin is usually discussed in terms of the magnetic dipole–dipole interaction

Figure 1. RDLM calculations on FePt. Top: the magnetization $M(T)$ versus T for the magnetization along the easy [001] axis (filled squares). The full line shows the mean field approximation to a classical Heisenberg model for comparison. Bottom: the magnetic anisotropy energy ΔE_{SOC} as a function of the square of the magnetization $M(T)$. The filled circles show the RDLM-based results, the full line give $K(T) \sim [M(T)/M(0)]^2$, and the dashed line is based on the single-ion model function. Reprinted (figure) with permission from [40], Copyright (2004) by the American Physical Society.
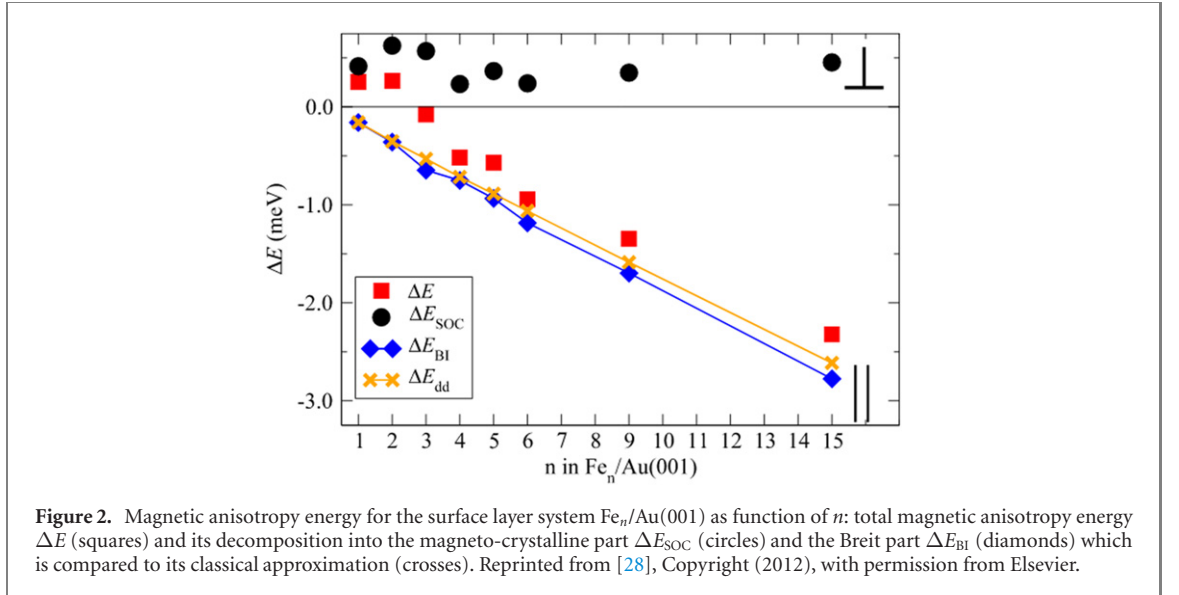
of individual magnetic moments on the lattice sites, $\Delta E_{\text{shape}} = E_{\text{dip}}(\vec{m}_{\parallel}) - E_{\text{dip}}(\vec{m}_{\perp})$, and can be calculated straightforwardly by performing the lattice summation [41]:

$$E_{\text{dip}} = \frac{1}{c^2} \sum_{i \neq j} \left[\frac{\vec{M}_i \cdot \vec{M}_j}{|\vec{R}_{ij}|^3} - 3 \frac{(\vec{M}_i \cdot \vec{R}_{ij})(\vec{M}_j \cdot \vec{R}_{ij})}{|\vec{R}_{ij}|^5} \right] \tag{19}$$

where $\vec{M}_{i(j)}$ is the magnetic moment on site $i(j)$. On the quantum mechanical level the origin of the shape anisotropy energy is the Breit interaction (BI) [42], i.e. one has $\Delta E_{\text{shape}} = \Delta E_{\text{BI}}$. Corresponding results for the magnetic anisotropy energy ΔE_{BI} of deposited Fe films, Fe$_n$/Au(001), calculated via the KKR Green function method [28] are compared in figure 2 with the shape anisotropy based on the classical dipole–dipole interaction, demonstrating good agreement.

3. Inter-atomic bilinear exchange interaction parameters

Most first-principles calculations on the bilinear exchange coupling parameters reported in the literature, are based on the MFT by evaluating the energy change due to a perturbation on the spin subsystem with respect to a suitable reference configuration [43]. Many results are based on calculations of the spin-spiral energy $\epsilon(\vec{q})$, giving access to the exchange parameters in the momentum space, $J_{\vec{q}}$[7, 14–16], followed by a Fourier transformation to the real space representation J_{ij}. Alternatively, the real space exchange parameters are calculated directly by evaluating the energy change due to the tilting of spin moments of interacting atoms. The corresponding non-relativistic expression (so-called Liechtenstein or LKAG formula) has been implemented based on the KKR as well as LMTO Green function (GF) [3, 4, 17, 43] band structure methods. It should be noted that the MFT provides a reasonable accuracy for the exchange coupling parameters in the case of infinitesimal rotations of the spins close to some equilibrium state, that can be justified only in the long wavelength and strong-coupling limits [44]. Accordingly, calculations of the exchange coupling parameters beyond the MFT, represented in terms of the inverse transverse susceptibility, were discussed in the literature by various authors [43–47]. Grotheer *et al*, for example, have demonstrated [45] a deviation of the spin-wave dispersion curves away from Γ point in the BZ, calculated for fcc Ni using the exchange parameters $\underline{J}_{\vec{q}} \sim \underline{\chi}_{\vec{q}}^{-1}$, from the MFT-based results for $\underline{J}_{\vec{q}}$. On the other hand, the results are close to each other in the long-wavelength limit

Figure 2. Magnetic anisotropy energy for the surface layer system Fe$_n$/Au(001) as function of n: total magnetic anisotropy energy ΔE (squares) and its decomposition into the magneto-crystalline part ΔE_{SOC} (circles) and the Breit part ΔE_{BI} (diamonds) which is compared to its classical approximation (crosses). Reprinted from [28], Copyright (2012), with permission from Elsevier.

(see figure 3). The calculations beyond the standard DFT are done by making use of the so-called constrained-field DFT. The latter theory was also used by Bruno [47] who suggested the 'renormalization' of the exchange coupling parameters expressed in terms of non-relativistic transverse magnetic susceptibility, according to $\underline{J} = \frac{1}{2}\underline{M}\underline{\chi}^{-1}\underline{M} = \frac{1}{2}\underline{M}(\underline{\tilde{\chi}}^{-1} - \underline{I}^{xc})\underline{M}$, with the various quantities defined as follows

$$\tilde{\chi}_{ij}^{-1} = \frac{2}{\pi}\int^{E_F} dE \int_{\Omega_i} d^3r \int_{\Omega_j} d^3r' \tag{20}$$

$$\times \text{Im}[G^\uparrow(\vec{r}, \vec{r}', E)G^\downarrow(\vec{r}', \vec{r}, E)], \tag{21}$$

$$M_i = \int_{\Omega_i} d^3r\, m(\vec{r}), \tag{22}$$

and

$$\tilde{I}_{ij}^{xc} = \delta_{ij}\frac{\Delta_i}{2M_i}, \tag{23}$$

with $\Delta_i = \frac{4}{M_i}\sum_j \tilde{J}_{ij}$, where

$$\tilde{J}_{ij} = \frac{1}{\pi}\text{Im}\int^{E_F} dE \int_{\Omega_i} d^3r \int_{\Omega_j} d^3r' \tag{24}$$

$$\times [B_{xc}(\vec{r})G^\uparrow(\vec{r}, \vec{r}', E)B_{xc}(\vec{r}')G^\downarrow(\vec{r}', \vec{r}, E)]. \tag{25}$$

This approach results in a Curie temperature of 634 K for fcc Ni (vs 350 K based on the MFT) which is in good agreement with the experimental value of (621–631 K). As was pointed out by Solovyev [44], such a corrections can be significant only for a certain class of materials, while, for instance, the calculations of spin-wave energies [45] and T_C [47] for bcc Fe demonstrate that these corrections are quite small. As most results in the literature were obtained using the exchange parameters based on the MFT, we restrict below to this approximation.

Similar to the case of the MCA discussed above, application of the MFT gives the energy change due to tilting of two spin moments represented in terms of the integrated DOS [4]. Within the multiple scattering formalism, this energy can be transformed using the Lloyd's formula leading to the expression

$$\Delta\mathcal{E} = -\frac{1}{\pi}\text{Im Tr}\int^{E_F} dE\left(\ln \underline{\tau}(E) - \ln \underline{\tau}^0(E)\right) \tag{26}$$

with $\underline{\tau}^{(0)}(E)$ and $\underline{\tau}(E)$ the scattering path operators for non-distorted and distorted systems, respectively.

As reported in reference [4], the expression for J_{ij} representing the exchange interaction between the spin moments on sites i and j, is given by the expression

$$J_{ij} = -\frac{1}{4\pi}\text{Im Tr}_L\int^{E_F} dE\underline{\Delta}_i\underline{\tau}_{ij}^\uparrow\underline{\Delta}_j\underline{\tau}_{ji}^\downarrow, \tag{27}$$
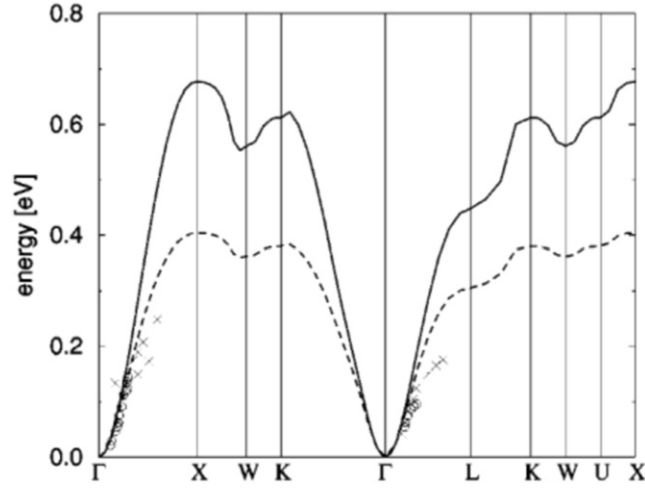
Figure 3. Adiabatic spin-wave dispersion relations along high-symmetry lines of the Brillouin zone for Ni. Broken line: frozen-magnon-torque method, full line: transverse susceptibility method. Reprinted (figure) with permission from [45], Copyright (2001) by the American Physical Society.

with $\underline{\Delta}_{i(j)} = ([\underline{t}^{\uparrow}]_{i(j)}^{-1} - [\underline{t}^{\downarrow}]_{i(j)}^{-1})$, where $\underline{t}_{i(j)}^{\uparrow}$ and $\underline{t}_{i(j)}^{\downarrow}$ are the spin-up and spin-down single-site scattering matrices, respectively, while $\underline{\tau}_{ij}^{\uparrow}$ and $\underline{\tau}_{ji}^{\downarrow}$ are the spin-up and spin-down, respectively, scattering path operators. As relativistic effects are not taken into account, the exchange interactions are isotropic with respect to the orientation of the magnetization as well as with respect to the direction of the spin tilting. On the other hand, spin–orbit coupling gives rise to an anisotropy for exchange interactions requiring a representation in the form of the exchange tensor $\underline{J}_{ij}$ with its antisymmetric part giving access to the DMI $\vec{D}_{ij}$.

Udvardi *et al* [5] and later Ebert and Mankovsky [6] suggested an extension of the classical Heisenberg Hamiltonian by accounting for relativistic effects for the exchange coupling (see also reference [43]). These calculations are based on a fully relativistic treatment of the electronic structure obtained by use of of the Dirac Hamiltonian

$$\mathcal{H}_D = -ic\vec{\alpha}\cdot\vec{\nabla} + \frac{1}{2}c^2(\beta - 1) + \bar{V}(\vec{r}) + \beta\,\vec{\sigma}\cdot\vec{B}(\vec{r}) + e\vec{\alpha}\cdot\vec{A}(\vec{r}). \tag{28}$$

Here, α_i and β are the standard Dirac matrices [31] while $\bar{V}(\vec{r})$ and $\vec{B}(\vec{r})$ are the spin independent and spin dependent parts of the electronic potential.

Considering a ferromagnetic (FM) state as a reference state with the magnetization along the z direction, a tilting of the magnetic moments on sites i and j leads to a modification of the scattering path operator implying the relation

$$\ln\underline{\tau} - \ln\underline{\tau}^0 = -\ln\left(1 + \underline{\tau}\left[\Delta\underline{m}_i + \Delta\underline{m}_j + \cdots\right]\right), \tag{29}$$

with $\underline{m}_i = \underline{t}_i^{-1}$. This allows to write down the expression for the energy change due to a spin tilting on sites i and j as follows

$$\mathcal{E}_{ij} = -\frac{1}{\pi}\text{ImTr}\int^{E_F}dE\,\Delta m_i\,\tau_{ij}\,\Delta m_j\,\tau_{ji}. \tag{30}$$

Within the approach of Udvardi *et al* [5], the dependence of the single-site inverse scattering matrix $\underline{m}_i$ on the orientation of magnetic moment $\hat{e}_i$ is accounted for by performing a corresponding rotation operation using the rotation matrix $\underline{R}(\theta,\phi)$, i.e., one has $\underline{m}_i(\theta,\phi) = \underline{R}(\theta,\phi)\underline{m}_i^0\underline{R}^+(\theta,\phi)$. The change of the scattering matrix $\underline{m}_i$ under spin rotation, $\Delta\underline{m}_i$, linearized with respect to the rotation angles, is given by the expression

$$\Delta\underline{m}_i = \underline{R}(\theta_i,\phi_i)\underline{m}_i^0\underline{R}^+(\theta_i,\phi_i) - \underline{m}_i^0$$

$$= \underline{m}_i^\theta\delta\theta_i + \underline{m}_i^\phi\delta\phi_i \tag{31}$$

with

$$\underline{m}_i^\theta = \frac{\partial}{\partial\theta}\underline{m}_i = \frac{\partial R}{\partial\theta}\underline{m}_iR^+ + R\underline{m}_i\frac{\partial R^+}{\partial\theta},$$

$$\underline{m}_i^\phi = \frac{\partial}{\partial\phi}\underline{m}_i = \frac{\partial R}{\partial\phi}\underline{m}_iR^+ + R\underline{m}_i\frac{\partial R^+}{\partial\phi}. \tag{32}$$

To calculate the derivatives of the rotation matrix, the definition

$$\hat{R}(\alpha_{\hat{n}}, \hat{n}) = e^{i\alpha_{\hat{n}}(\hat{n}\cdot\hat{\vec{J}})} \tag{33}$$

for the corresponding operator is used, with $\hat{\vec{J}}$ the total angular momentum operator. $\hat{R}(\alpha_{\hat{n}}, \hat{n})$ describes a rotation of the magnetic moment $\hat{m}$ by the angle $\alpha_{\hat{n}}$ about the direction $\hat{n}\perp\hat{m}$, that gives in particular $\underline{R}(\theta, \hat{n})$ for $\hat{n} = \hat{y}$ and $\underline{R}(\phi, \hat{n})$ for $\hat{n} = \hat{z}$.

This leads to the second derivatives of the total energy with respect to the titling angles $\alpha_i = \{\theta_i, \phi_i\}$ and $\beta_j = \{\theta_j, \phi_j\}$

$$\frac{\partial^2 \mathcal{E}}{\partial\alpha_i \partial\beta_j} = -\frac{1}{\pi}\text{Im}\,\text{Tr}\int^{E_\text{F}} dE \underline{m}_i^\alpha \underline{\tau}_{ij} \underline{m}_j^\beta \underline{\tau}_{ji}. \tag{34}$$

As is discussed by Udvardi *et al* [5], these derivatives give access to all elements $J_{ij}^{\mu\nu}$ of the exchange tensor, where $\mu(\nu) = \{x, y, z\}$. Note, however, that only the tensor elements with $\mu(\nu) = \{x, y\}$ can be calculated using the magnetization direction along the $\hat{z}$ axis, giving access to the z component D_{ij}^z of the DMI. In order to obtain all other tensor elements, an auxiliary rotation of the magnetization towards the $\hat{x}$ and $\hat{y}$ directions of the global frame of reference is required. For example, the component D_{ij}^x if the DMI vector can be evaluated via the tensor elements

$$J_{ij}^{zy} = \frac{\partial^2 E}{\partial\theta_i \partial\phi_j} \quad \text{and} \quad J_{ij}^{yz} = \frac{\partial^2 E}{\partial\phi_i \partial\theta_j} \tag{35}$$

for $\theta = \frac{\pi}{2}$ and $\phi = 0$.

An alternative expression within the KKR multiple scattering formalism has been worked out by Ebert and Mankovsky [6], by using the alternative convention for the electronic Green function (GF) as suggested by Dederichs and co-workers [48]. According to this convention, the off-site part of the GF is given by the expression:

$$G(\vec{r}_i, \vec{r}_j, E) = \sum_{\Lambda\Lambda'} R_\Lambda^i(\vec{r}_i, E) G_{\Lambda\Lambda'}^{ij}(E) R_{\Lambda'}^{j\times}(\vec{r}_j, E), \tag{36}$$

where $G_{\Lambda\Lambda'}^{ij}(E)$ is the so-called structural Green's function, R_Λ^i is a regular solution to the single-site Dirac equation labeled by the combined quantum numbers Λ [31]. The energy change $\Delta\mathcal{E}_{ij}$ due to a spin tilting on sites i and j, given by equation (30), transformed to the above mentioned convention is expressed as follows

$$\Delta\mathcal{E}_{ij} = -\frac{1}{\pi}\text{Im}\,\text{Tr}\int dE \Delta\underline{t}^i \underline{G}^{ij} \Delta\underline{t}^j \underline{G}^{ji}, \tag{37}$$

where the change of the single-site t-matrix $\Delta\underline{t}^i$ can be represented in terms of the perturbation $\Delta V^i(\vec{r})$ at site i using the expression

$$\Delta t_{\Lambda'\Lambda}^i = \int d^3 r R_{\Lambda'}^{i\times}(r)\Delta V(r) R_\Lambda^i(r) = \Delta V_{\Lambda'\Lambda}^{(R)i}, \tag{38}$$

where the perturbation caused by the rotation of the spin magnetic moment $\hat{e}_i$ is represented by a change of the spin-dependent potential in equation (28) (in contrast to the approach used in reference [5])

$$\Delta V(r) = V_{\hat{n}}(r) - V_{\hat{n}_0}(r) = \beta\vec{\sigma}(\hat{n} - \hat{n}_0)B(r). \tag{39}$$

Using again the frozen potential approximation implies that the spatial part of the potential $V_{\hat{n}}(r)$ does not change upon rotation of spin orientation.

Coming back to the convention for the GF used by Györffy and co-workers [49] according to equation (9) the expression for the elements of the exchange tensor represented in terms of the scattering path operator $\tau_{\Lambda'\Lambda}^{ij}(E)$ has the form

$$J_{ij}^{\alpha_i\alpha_j} = -\frac{1}{\pi}\text{Im}\,\text{Tr}\int dE\,\underline{T}^{\alpha_i}\underline{\tau}^{ij}\underline{T}^{\alpha_j}\underline{\tau}^{ji}, \tag{40}$$

where

$$T_{\Lambda\Lambda'}^{\alpha_i} = \int d^3 r Z_\Lambda^\times(\vec{r})\beta\sigma_\alpha B(r) Z_{\Lambda'}(\vec{r}). \tag{41}$$

As an example, figure 4 shows the exchange coupling parameters J_{ij} calculated for NbS$_2$ dichalcogenide intercalated by 3d elements, i.e. $M_{1/3}$NbS$_2$ (M = Cr, Mn, Fe) [50]. The strongest M–M interactions occur for the first two neighbor shells. The parameters are predominantly positive in the case of Cr$_{1/3}$NbS$_2$ and Mn$_{1/3}$NbS$_2$, leading to a FM ordering in these systems when the DMI is not taken into account. The critical temperatures calculated by means of Monte Carlo simulations are in good agreement with experiment (see table 2). Note in addition that the impact of the DMI discussed in reference [50] leads in the case of Cr$_{1/3}$NbS$_2$ and Mn$_{1/3}$NbS$_2$ compounds to the formation of a helimagnetic structure.

Figure 4. Isotropic exchange coupling parameters for $M_{1/3}NbS_2$ (M = Cr, Mn, Fe). Open symbols represent results obtained for the experimental structure parameters, full symbols correspond to the systems with the DFT-optimized structure parameters. Reprinted (figure) with permission from [50], Copyright (2016) by the American Physical Society.
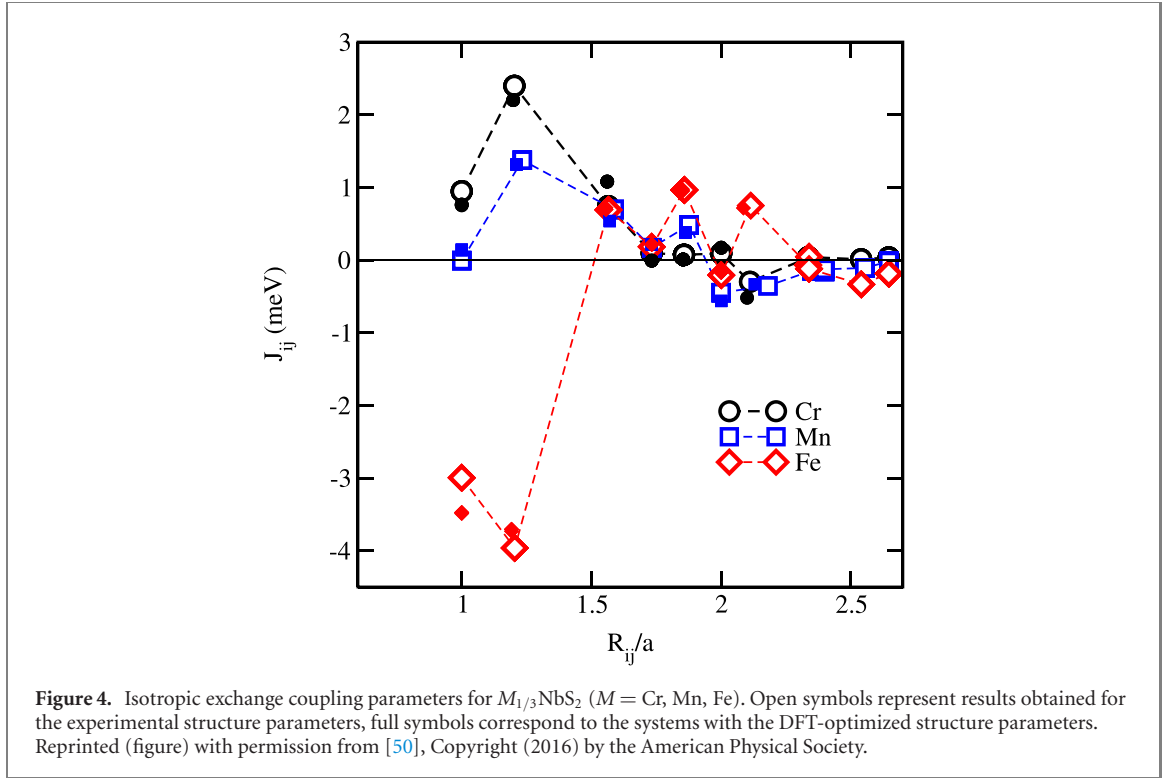
Table 2. The Curie and Néel temperatures (in K units) calculated for $M_{1/3}NbS_2$ compounds via MC simulations in comparison with experimental results. Reprinted table with permission from [51], Copyright (2016) by the American Physical Society.

	MC	EXPT
$Cr_{1/3}NbS_2$	115(FM)	127(FM) [51]
$Mn_{1/3}NbS_2$	80(FM)	65(FM)
$Fe_{1/3}NbS_2$	63(AFM)	47(AFM) [52], 44(AFM) [53]

When compared to the approach of Udvardi *et al* [5], the expression in equation (40) is given explicitly in Cartesian coordinates. However, auxiliary rotations of the magnetization are still required to calculate all tensor elements, and as a consequence, all components of the DMI vector. This can be avoided using the approach reported recently [54, 81] for DMI calculations.

In this case, using the grand-canonical potential in the operator form

$$\mathcal{K} = \mathcal{H} - \mu N, \tag{42}$$

with μ the chemical potential, the variation of single-particle energy density $\Delta\mathcal{E}(\vec{r})$ caused by a perturbation is written in terms of the electronic Green function for $T = 0$ K as follows

$$\Delta\mathcal{E}(\vec{r}) = -\frac{1}{\pi} \text{Im Tr} \int^{\mu} dE (E - \mu) \Delta G(\vec{r}, \vec{r}, E). \tag{43}$$

Assuming the perturbation ΔV responsible for the change of the Green function $\Delta G = G - G_0$ (the index 0 indicates here the collinear ferromagnetic reference state) to be small, ΔG can be expanded up to any order w.r.t. the perturbation

$$\Delta G(E) = G_0 \Delta V G_0 + G_0 \Delta V G_0 \Delta V G_0 + G_0 \Delta V G_0 \Delta V G_0 \Delta V G_0$$
$$+ G_0 \Delta V G_0 \Delta V G_0 \Delta V G_0 \Delta V G_0 + \cdots, \tag{44}$$

leading to a corresponding expansion for the energy change with respect to the perturbation as follows

$$\Delta\mathcal{E} = \Delta\mathcal{E}^{(1)} + \Delta\mathcal{E}^{(2)} + \Delta\mathcal{E}^{(3)} + \Delta\mathcal{E}^{(4)} + \cdots \tag{45}$$

Here and below we drop the energy argument for the Green function $G(E)$ for the sake of convenience. This expression is completely general as it gives the energy change as a response to any type of perturbation. When

ΔV is associated with tiltings of the spin magnetic moments, it can be expressed within the frozen potential approximation and in line with equation (39) as follows

$$\Delta V(\vec{r}) = \sum_i \beta(\vec{\sigma} \cdot \hat{s}_i - \sigma_z)B_{xc}(\vec{r}).\tag{46}$$

With this, the energy expansion in equation (45) gives access to the bilinear DMI as well as to higher order multispin interactions [55]. To demonstrate the use of this approach, we start with the x and y components of the DMI vector, which can be obtained by setting the perturbation ΔV in the form of a spin-spiral described by the configuration of the magnetic moments

$$\hat{s}_i = \Big(\sin(\vec{q} \cdot \vec{R}_i), 0, \cos(\vec{q} \cdot \vec{R}_i)\Big),\tag{47}$$

with the wave vector $\vec{q} = (0, q, 0)$. As it follows from the spin Hamiltonian, the slope of the spin wave energy dispersion at the Γ point is determined by the DMI as follows

$$\lim_{q \to 0}\frac{\partial E_{\text{DM}}^{(1)}}{\partial q_y} = \lim_{q \to 0}\frac{\partial}{\partial q_y}\sum_{ij}D_{ij}^y \sin(\vec{q} \cdot (\vec{R}_j - \vec{R}_i))$$

$$= \sum_{ij}D_{ij}^y (\vec{R}_j - \vec{R}_i)_y.\tag{48}$$

Identifying this with the corresponding derivative of the energy $\Delta\mathcal{E}^{(1)}$ in equation (45)

$$\frac{\partial \Delta\mathcal{E}^{(1)}}{\partial q_\alpha}\bigg|_{q \to 0} = \frac{\partial E_{\text{DM}}^{(1)}}{\partial q_\alpha}\bigg|_{q \to 0},\tag{49}$$

and equating the corresponding terms for each atomic pair (i, j), one obtains the following expression for the y component of the DMI vector:

$$D_{ij}^y = \Big(-\frac{1}{2\pi}\Big)\operatorname{Im} \operatorname{Tr} \int^\mu \mathrm{d}E\,(E - \mu)\big[\underline{O}^j(E)\,\underline{\tau}^{ji}(E)\,\underline{T}^{i,x}(E)\,\underline{\tau}^{ij}(E) - \underline{O}^i(E)\,\underline{\tau}^{ij}(E)\,\underline{T}^{j,x}(E)\,\underline{\tau}^{ji}(E)\big].\tag{50}$$

In a completely analogous way one can derive the x-component of the DMI vector, D_{ij}^x. The overlap integrals $O_{\Lambda\Lambda'}^j$ and matrix elements $T_{\Lambda\Lambda'}^{i,\alpha}$ of the operator $\mathcal{T}^{i,\alpha} = \beta\sigma_\alpha B_{xc}^i(\vec{r})$ (which are connected with the components of the torque operator $\beta[\vec{\sigma} \times \hat{m}]B_{xc}^i(\vec{r})$) are defined as follows [6]:

$$O_{\Lambda\Lambda'}^j = \int_{\Omega_j} \mathrm{d}^3r\, Z_\Lambda^{j\times}(\vec{r}, E)\, Z_{\Lambda'}^j(\vec{r}, E)\tag{51}$$

$$T_{\Lambda\Lambda'}^{i,\alpha} = \int_{\Omega_i} \mathrm{d}^3r\, Z_\Lambda^{i\times}(\vec{r}, E)\, \big[\beta\sigma_\alpha B_{xc}^i(\vec{r})\big]\, Z_{\Lambda'}^i(\vec{r}, E).\tag{52}$$

As is shown in reference [55], the D_{ij}^z component of the DMI, as well isotropic exchange parameter J_{ij} can also be obtained on the basis of equations (44) and (45) using the second order term w.r.t. the perturbation, for a spin spiral with the form

$$\hat{s}_i = (\sin\theta \, \cos(\vec{q} \cdot \vec{R}), \sin\theta \, \sin(\vec{q} \cdot \vec{R}), \cos\theta).\tag{53}$$

In this case case, the DMI component D_{ij}^z and the isotropic exchange interaction are obtained by taking the first- and second-order derivatives of the energy $\Delta\mathcal{E}^{(2)}(\vec{q})$ (see equation (45)), respectively, with respect to $\vec{q}$:

$$\frac{\partial}{\partial\vec{q}}\Delta E_H(\vec{q})\bigg|_{q \to 0} = -\sin^2\theta\sum_{i \neq j}^N D_{ij}^z \hat{q} \cdot (\vec{R}_i - \vec{R}_j)\tag{54}$$

and

$$\frac{\partial^2}{\partial\vec{q}^2}\Delta E_H(\vec{q})\bigg|_{q \to 0} = \sin^2\theta\sum_{i,j}J_{ij}(\hat{q} \cdot (\vec{R}_i - \vec{R}_j))^2\tag{55}$$

with $\hat{q} = \vec{q}/|\vec{q}|$ the unit vector giving the direction of the wave vector $\vec{q}$. Identifying these expressions again with the corresponding derivatives of $\Delta\mathcal{E}^{(2)}(\vec{q})$, one obtains the following relations for D_{ij}^z

$$D_{ij}^z = \frac{1}{2}(\mathcal{J}_{ij}^{xy} - \mathcal{J}_{ij}^{yx})\tag{56}$$

and for J_{ij}

$$J_{ij} = \frac{1}{2}(\mathcal{J}_{ij}^{xx} + \mathcal{J}_{ij}^{yy}), \tag{57}$$

where the tensor elements $\mathcal{J}^{\alpha\beta}$ are given by equations (40) and (41).

Similar to the magnetic anisotropy, the exchange coupling parameters depend on temperature, that should be taken into account within the finite temperature spin dynamic simulations. An approach that gives access to calculations of exchange coupling parameters for finite temperature has been reported in reference [55]. It accounts for the electronic structure modification due to temperature induced lattice vibrations by using the alloy analogy model in the adiabatic approximation. This implies calculations of the thermal average $\langle \ldots \rangle_T$ as the configurational average over a set of appropriately chosen set of atomic displacements, using the CPA alloy theory [56–58].

To make use of this scheme to account for lattice vibrations, a discrete set of N_v vectors $\Delta \vec{R}_v^q(T)$ is introduced for each atom, with the temperature dependent amplitude, which characterize a rigid displacement of the atomic potential in the spirit of the rigid muffin-tin approximation [59, 60]. The corresponding single-site t-matrix in the common global frame of the solid is given by the transformation:

$$\underline{t}_v^q = \underline{U}(\Delta \vec{R}_v) \, \underline{t}^{q,\mathrm{loc}} \, \underline{U}(\Delta \vec{R}_v)^{-1}, \tag{58}$$

with the so-called U-transformation matrix $\underline{U}(\vec{s})$ given in its non-relativistic form by [59, 60]:

$$U_{LL'}(\vec{s}) = 4\pi \sum_{L''} i^{l+l''-l'} \, C_{LL'L''} \, j_{l''}(|\vec{s}|k) \, Y_{L''}(\hat{s}). \tag{59}$$

Here $L = (l, m)$ represents the non-relativistic angular momentum quantum numbers, $j_l(x)$ is a spherical Bessel function, $Y_L(\hat{r})$ a real spherical harmonics, $C_{LL'L''}$ a corresponding Gaunt number and $k = \sqrt{E}$ is the electronic wave vector. The relativistic version of the U-matrix is obtained by a standard Clebsch–Gordan transformation [31].

Every displacement characterized by a displacement vectors $\Delta \vec{R}_v(T)$ can be treated as a pseudo-component of a pseudo alloy. Thus, the thermal averaging can be performed as the site diagonal configurational average for a substitutional alloy, by solving the multi-component CPA equations within the global frame of reference [58].

The same idea can be used also to take into account thermal spin fluctuations. A set of representative orientation vectors $\hat{e}_f$ (with $f = 1, \ldots, N_f$) for the local magnetic moment is introduced. Using the rigid spin approximation, the single-site t-matrix in the global frame, corresponding to a given orientation vector, is determined by:

$$\underline{t}_f^q = \underline{R}(\hat{e}_f) \, \underline{t}^{q,\mathrm{loc}} \, \underline{R}(\hat{e}_f)^{-1}, \tag{60}$$

where $\underline{t}^{q,\mathrm{loc}}$ is the single-site t-matrix in the local frame. Here the transformation from the local to the global frame of reference is expressed by the rotation matrices $\underline{R}(\hat{e}_f)$ that are determined by the vectors $\hat{e}_f$ or corresponding Euler angles [31]. Again, every orientation can be treated as a pseudo-component of a pseudo alloy, that allows to use the alloy analogy model to calculate the thermal average over all types of spin fluctuations [58].

The alloy analogy for thermal vibrations applied to the temperature dependent exchange coupling parameters leads to

$$\bar{J}_{ij}^{\alpha_i \alpha_j} = -\frac{1}{2\pi} \Im \int dE \, \mathrm{Trace} \langle \Delta \underline{V}^{\alpha_i} \underline{\tau}^{ij} \Delta \underline{V}^{\alpha_j} \underline{\tau}^{ji} \rangle_c, \tag{61}$$

where $\langle \ldots \rangle_c$ represents the configurational average with respect to the set of displacements. In case of the exchange coupling parameters one has to distinguish between the averaging over thermal lattice vibrations and spin fluctuations. In the first case the configurational average is approximated as follows $\langle \Delta \underline{V}^i \underline{\tau}^{ij} \Delta \underline{V}^j \underline{\tau}^{ji} \rangle_{\mathrm{vib}} \approx \langle \Delta \underline{V}^i \underline{\tau}^{ij} \rangle_{\mathrm{vib}} \langle \Delta \underline{V}^j \underline{\tau}^{ji} \rangle_{\mathrm{vib}}$, assuming a negligible impact of the so-called vertex corrections [61]. This averaging accounts for the impact of thermally induced phonons on the exchange coupling parameters for every temperature before their use in MC or spin dynamics simulations that deal subsequently with the thermal averaging in spin subspace. The impact of spin fluctuations can be incorporated as well within the electronic structure calculations. For a non-polarized paramagnetic reference state, this can be done, e.g., by using the so-called disorder local moment (DLM) scheme formulated in general within the non-relativistic (or scalar-relativistic) framework. Magnetic disorder in this case can be modeled by creating a pseudo alloy with an occupation of the atomic sites by two types of atoms with opposite spin moments oriented upwards, $M^\uparrow$ and downwards $M^\downarrow$, respectively, i.e. considering the alloy $M_{0.5}^\uparrow M_{0.5}^\downarrow$. In the relativistic case the corresponding RDLM scheme has to describe the magnetic disorder by a discrete set of N_f orientation vectors, and as a consequence, the average $\langle \underline{\tau}^{ij} \rangle_{\mathrm{spin}}$ has to be calculated taking into account all these orientations. A comparison of the results obtained for the isotropic exchange coupling constants J_{ij} for bcc Fe using the DLM and RDLM schemes is shown in
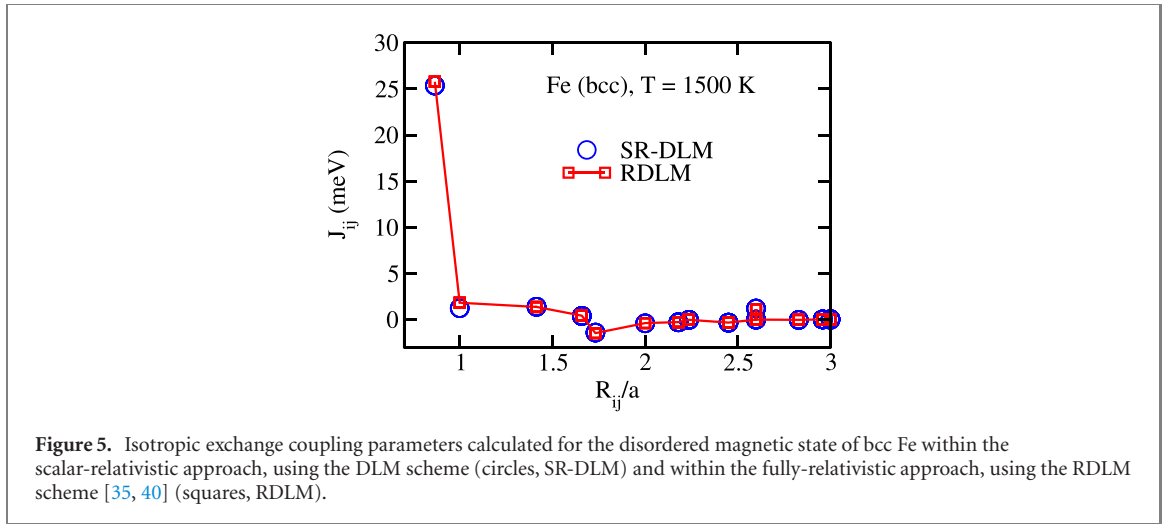
Figure 5. Isotropic exchange coupling parameters calculated for the disordered magnetic state of bcc Fe within the scalar-relativistic approach, using the DLM scheme (circles, SR-DLM) and within the fully-relativistic approach, using the RDLM scheme [35, 40] (squares, RDLM).

figure 5, demonstrating close agreement, with the small differences to be ascribed to the different account of relativistic effects, i.e. in particular the spin–orbit coupling.

Finally, it should be stressed once more that the methods for the calculation of the interatomic exchange coupling parameters, discussed in this section, are well applicable for the so-called Heisenberg systems, i.e. those having robust atomic magnetic moments weakly dependent on the magnetic configuration, ensuring that way an appropriate description of their magnetic properties. However, specific properties of various materials related to their electronic structure, may require for corresponding corrections to the exchange coupling parameters, e.g., by going beyond the use of the MFT in the case of fcc Ni. For some metallic materials composed of magnetic (M) and non-magnetic (N) elements, the M–M exchange interactions have to be re-normalized by taking into account the M–N–M interactions mediated by the non-magnetic atoms having induced magnetic moments. In practice, this was done using various schemes [62–64], as induced magnetic moments may depend strongly on the magnetic configuration of the surrounding atoms and in turn on the temperature. In the case of materials with strongly correlated electrons, the electronic structure calculations have to be done beyond the LSDA-DFT approach. This for instance implies a description of the exchange-correlation energy of electrons within the LSDA + U or dynamical mean field theory (DMFT), that has to be consistently taken into account also when calculating the interatomic exchange coupling parameters [65–67]. A rather detailed analysis of the various methods for calculating the exchange parameters has been done recently in the review by Szilva *et al* [43].

4. Multi-spin expansion of spin Hamiltonian: general remarks

Despite the obvious success of the classical Heisenberg model for many applications, higher-order multi-spin expansion H_{ms} of the spin Hamiltonian H, given by the expression
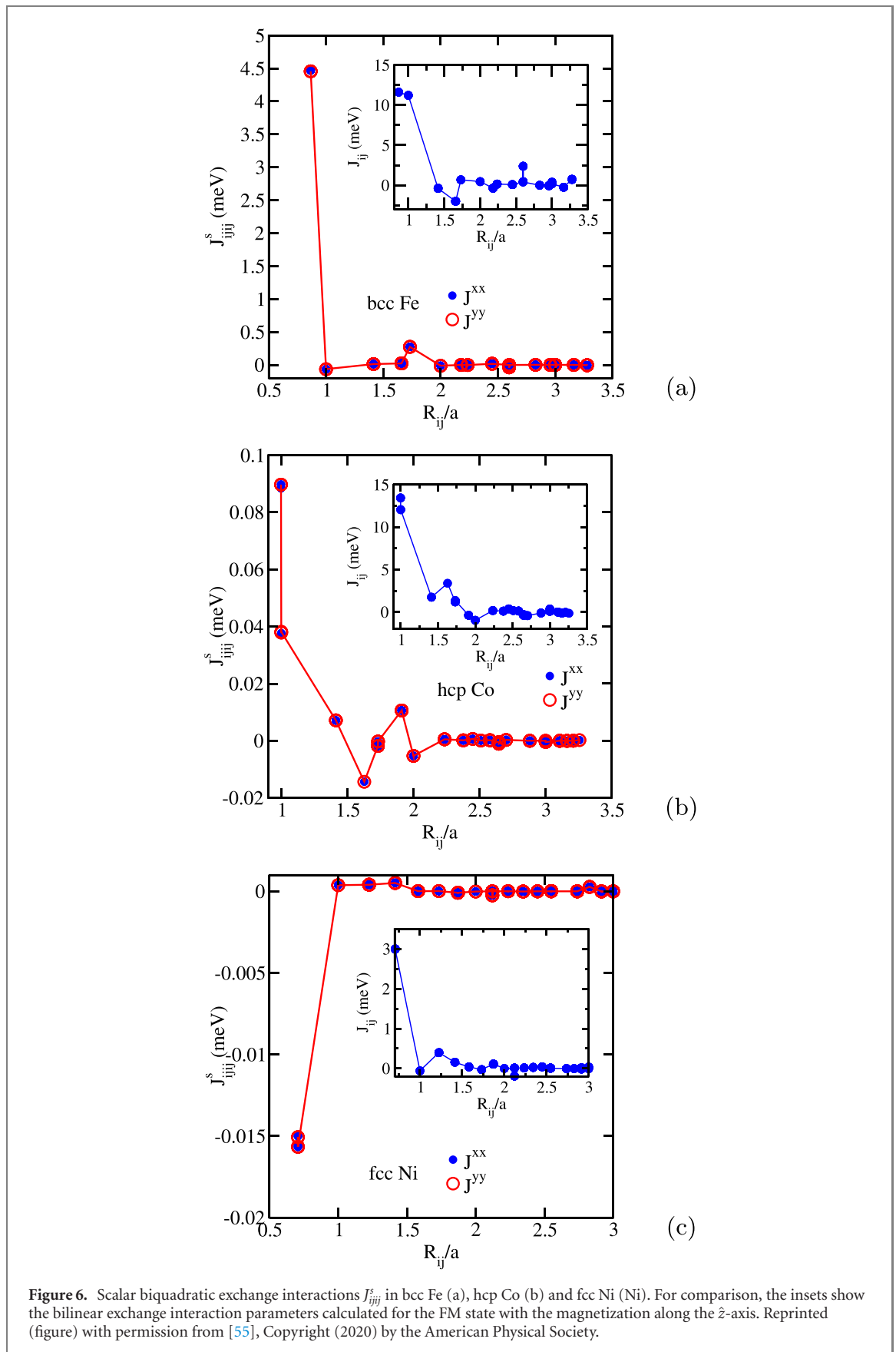
$$H_{\mathrm{ms}} = -\frac{1}{3!}\sum_{i,j,k} J_{ijk}\hat{s}_i \cdot (\hat{s}_j \times \hat{s}_k), -\frac{2}{p!}\sum_{i,j,k,l} J^s_{ijkl}(\hat{s}_i \cdot \hat{s}_j)(\hat{s}_k \cdot \hat{s}_l) - \frac{2}{p!}\sum_{i,j,k,l} \vec{\mathcal{D}}_{ijkl} \cdot (\hat{s}_i \times \hat{s}_j)(\hat{s}_k \cdot \hat{s}_l) + \cdots ,$$

$$= H^3 + H^{4,s} + H^{4,a} + \cdots \tag{62}$$

can be of great importance to describe more subtle properties of magnetic materials [13, 19, 21, 22, 24, 68–75].

This concerns first of all systems with a non-collinear ground state characterized by finite spin tilting angles, that makes multispin contributions to the energy non-negligible. In particular, many reports published recently discuss the impact of the multispin interactions on the stabilization of exotic topologically non-trivial magnetic textures, e.g. skyrmions, hopfions, etc [27, 76, 77].

Corresponding calculations of the multi-spin exchange parameters have been reported by different groups. The approach based on the Connolly–Williams scheme has been used to calculate the four-spin non-chiral (two-site and three-site) and chiral interactions for Cr trimers [13] and for a deposited Fe atomic chain [78], respectively, for the biquadratic, three-site four spin and four-site four spin interaction parameters [26, 27]. The authors discuss the role of these type of interactions for the stabilization of different types of non-collinear magnetic structures as skyrmions and antiskyrmions.

A more flexible mapping scheme using perturbation theory within the KKR Green function formalism was only reported recently by Brinker *et al* [79, 80], and by the present authors [55]. Here we discuss the latter

Figure 6. Scalar biquadratic exchange interactions J^s_{ijij} in bcc Fe (a), hcp Co (b) and fcc Ni (Ni). For comparison, the insets show the bilinear exchange interaction parameters calculated for the FM state with the magnetization along the $\hat{z}$-axis. Reprinted (figure) with permission from [55], Copyright (2020) by the American Physical Society.

approach, i.e. the energy expansion w.r.t. ΔV in equation (45). One has to point out that a spin tilting in a real system has a finite amplitude and therefore the higher order terms in this expansion might become non-negligible and in general should be taken into account. Their role obviously depends on the specific material and should increase with temperature that leads to an increasing amplitude of the spin fluctuations. As these

higher-order terms are directly connected to the multispin terms in the extended Heisenberg Hamiltonian, one has to expect also a non-negligible role of the multispin interactions for some magnetic properties.

Extending the spin Hamiltonian to go beyond the classical Heisenberg model, we discuss first the four-spin exchange interaction terms J_{ijkl} and $\vec{\mathcal{D}}_{ijkl}$. They can be calculated using the fourth-order term of the Green function expansion $\Delta\mathcal{E}^{(4)}$ given by:

$$\Delta\mathcal{E}^{(4)} = -\frac{1}{\pi}\text{Im Tr}\int^{E_F} dE\,(E - E_F)\Delta VG\Delta VG\Delta VG\Delta VG$$

$$= -\frac{1}{\pi}\text{Im Tr}\int^{E_F} dE\,\Delta VG\Delta VG\Delta VG\Delta VG \tag{63}$$

where the sum rule for the Green function $\frac{dG}{dE} = -GG$ followed by integration by parts was used to get a more compact expression. Using the multiple-scattering representation for the Green function, this leads to:

$$\Delta\mathcal{E}^{(4)} = \sum_{i,j,k,l} -\frac{1}{\pi}\text{Im Tr}\int^{E_F} dE\,\Delta\underline{V}_{ii}\underline{\tau}_{ij}\Delta\underline{V}_{jj}\underline{\tau}_{jk}\Delta\underline{V}_{kk}\underline{\tau}_{kl}\Delta\underline{V}_{ll}\underline{\tau}_{li} \tag{64}$$

with the matrix elements $\Delta\underline{V}_{ii} = \langle Z_i|\Delta V|Z_i\rangle$. Using the ferromagnetic state with $\vec{M}\|\hat{z}$ as a reference state, and creating the perturbation ΔV in the form of a spin-spiral according to equation (53), one obtains the corresponding $\vec{q}$-dependent energy change $\Delta\mathcal{E}^{(4)}(\vec{q})$, written here explicitly as an example

$$\Delta\mathcal{E}^{(4)} = -\frac{1}{\pi}\sum_{i,j,k,l}\text{Im Tr}\int^{E_F} dE\,\sin^4\theta\Big[I_{ijkl}^{xxxx}\,\cos(\vec{q}\cdot\vec{R}_i)\,\cos(\vec{q}\cdot\vec{R}_j)\,\cos(\vec{q}\cdot\vec{R}_k)\,\cos(\vec{q}\cdot\vec{R}_l)$$

$$+ I_{ijkl}^{xxyy}\,\cos(\vec{q}\cdot\vec{R}_i)\,\cos(\vec{q}\cdot\vec{R}_j)\,\sin(\vec{q}\cdot\vec{R}_k)\,\sin\vec{q}\cdot\vec{R}_l)$$

$$+ I_{ijkl}^{yyxx}\,\sin(\vec{q}\cdot\vec{R}_i)\,\sin(\vec{q}\cdot\vec{R}_j)\,\cos(\vec{q}\cdot\vec{R}_k)\,\cos(\vec{q}\cdot\vec{R}_l)$$

$$+ I_{ijkl}^{yyyy}\,\sin(\vec{q}\cdot\vec{R}_i)\,\sin(\vec{q}\cdot\vec{R}_j)\,\sin(\vec{q}\cdot\vec{R}_k)\,\sin(\vec{q}\cdot\vec{R}_l)$$

$$+ I_{ijkl}^{xyxx}\,\cos(\vec{q}\cdot\vec{R}_i)\,\sin(\vec{q}\cdot\vec{R}_j)\,\cos(\vec{q}\cdot\vec{R}_k)\,\cos(\vec{q}\cdot\vec{R}_l)$$

$$+ I_{ijkl}^{yxyy}\,\sin(\vec{q}\cdot\vec{R}_i)\,\cos(\vec{q}\cdot\vec{R}_j)\,\sin(\vec{q}\cdot\vec{R}_k)\,\sin\vec{q}\cdot\vec{R}_l)$$

$$+ I_{ijkl}^{yxxx}\,\sin(\vec{q}\cdot\vec{R}_i)\,\cos(\vec{q}\cdot\vec{R}_j)\,\cos(\vec{q}\cdot\vec{R}_k)\,\cos(\vec{q}\cdot\vec{R}_l)$$

$$+ I_{ijkl}^{xyyy}\,\cos(\vec{q}\cdot\vec{R}_i)\,\sin(\vec{q}\cdot\vec{R}_j)\,\sin(\vec{q}\cdot\vec{R}_k)\,\sin(\vec{q}\cdot\vec{R}_l) + \cdots\Big] \tag{65}$$

where

$$I_{ijkl}^{\alpha\beta\gamma\delta} = \underline{T}^{i,\alpha}(E)\,\underline{\tau}^{ij}(E)\underline{T}^{j,\beta}(E)\,\underline{\tau}^{jk}(E)\underline{T}^{k,\gamma}(E)\,\underline{\tau}^{kl}(E)\underline{T}^{l,\delta}(E)\,\underline{\tau}^{li}(E). \tag{66}$$

As is shown in reference [55], the four-spin isotropic exchange interaction J_{ijkl} and z-component of the DMI-like interaction $\mathcal{D}_{ijkl}^z$ can be obtained calculating the energy derivatives $\frac{\partial^4}{\partial q^4}\Delta\mathcal{E}^{(4)}$ and $\frac{\partial^3}{\partial q^3}\Delta\mathcal{E}^{(4)}$ in the limit of $q = 0$, and then identified with the corresponding derivatives of the terms $H^{4,s}$ and $H^{4,a}$ in equation (62). These interaction terms are given by the expressions

$$J_{ijkl}^s = \frac{1}{4}\Big[\mathcal{J}_{ijkl}^{xxxx} + \mathcal{J}_{ijkl}^{xxyy} + \mathcal{J}_{ijkl}^{yyxx} + \mathcal{J}_{ijkl}^{yyyy}\Big] \tag{67}$$

and

$$D_{ijkl}^z = \frac{1}{4}\Big[\mathcal{J}_{ijkl}^{xyxx} + \mathcal{J}_{ijkl}^{xyyy} - \mathcal{J}_{ijkl}^{yxxx} - \mathcal{J}_{ijkl}^{yxyy})\Big], \tag{68}$$

where the following definition is used:

$$\mathcal{J}_{ijkl}^{\alpha\beta\gamma\delta} = \frac{1}{2\pi}\text{Im Tr}\int^{E_F} dE\,T_i^\alpha\tau_{ij}T_j^\beta\tau_{jk}T_k^\gamma\tau_{kl}T_l^\delta\tau_{li}. \tag{69}$$

These expression obviously give also access to a special cases, i.e. the four-spin three-site interactions with $l = j$, and the four spin two-site, so called biquadratic exchange interactions with $k = i$ and $l = j$.

The scalar biquadratic exchange interaction parameters J_{ijij}^s calculated on the basis of equation (67) for the three 3d bulk ferromagnetic systems bcc Fe, hcp Co and fcc Ni have been reported in reference [55]. The results are plotted in figure 6 as a function of the distance $R_{ij} + R_{jk} + R_{kl} + R_{li}$. For comparison, the insets give the corresponding bilinear isotropic exchange interactions for these materials. One can see rather strong first-neighbor interactions for bcc Fe, demonstrating the non-negligible character of the biquadratic interactions.

This is of course a material-specific property, and one notes as decrease for the biquadratic exchange parameters when going to Co and Ni as shown in figures 6(b) and (c), respectively.

In order to calculate the x and y components of the four-spin and as a special case the three-site-DMI (TDMI) and biquadratic-DMI (BDMI) type interactions, the scheme suggested in reference [55] for the calculation of the DMI parameters [54, 81] can be used, which exploited the DMI-governed behavior of the spin-wave dispersion having a finite slope at the Γ point of the Brillouin zone. Note, however, that a more general form of perturbation is required in this case described by a 2D spin modulation field according to the expression

$$\hat{s}_i = \left(\sin(\vec{q}_1 \cdot \vec{R}_i) \, \cos(\vec{q}_2 \cdot \vec{R}_i), \sin(\vec{q}_2 \cdot \vec{R}_i), \cos(\vec{q}_1 \cdot \vec{R}_i) \cos(\vec{q}_2 \cdot \vec{R}_i) \right), \tag{70}$$

where the wave vectors $\vec{q}_1$ and $\vec{q}_2$ are orthogonal to each other, as for example $\vec{q}_1 = q_1 \hat{y}$ and $\vec{q}_2 = q_2 \hat{x}$.

Taking the second-order derivative with respect to the wave-vector $\vec{q}_2$ and the first-order derivative with respect to the wave-vectors $\vec{q}_1$ and $\vec{q}_2$, and considering the limit $q_{1(2)} \to 0$, one obtains

$$\left. \frac{\partial^3}{\partial q_2^3} \right|_{q_2=0} H^{4,a} = \sum_{i,j,k,l} \mathcal{D}_{ijkl}^x \, (\hat{q}_2 \cdot \vec{R}_{ij}) \, (\hat{q}_2 \cdot \vec{R}_{lk})^2,$$

and

$$\left. \frac{\partial}{\partial q_1} \right|_{q_1=0} \left. \frac{\partial^2}{\partial q_2^2} \right|_{q_2=0} H^{4,a} = \sum_{i,j,k,l} \mathcal{D}_{ijkl}^y \, (\hat{q}_1 \cdot \vec{R}_{ij}) \, (\hat{q}_2 \cdot \vec{R}_{lk})^2,$$

where $\vec{R}_{ij} = \vec{R}_j - \vec{R}_i$ and $\vec{R}_{lk} = \vec{R}_k - \vec{R}_l$.

The microscopic expressions for the x and y components of $\vec{\mathcal{D}}_{ijkl}$ describing the four-spin interactions is derived on the basis of the third-order term in equation (44)

$$\Delta \mathcal{E}^{(3)} = -\frac{1}{\pi} \mathrm{Im} \, \mathrm{Tr} \int^{E_F} dE(E - E_F) \, G_0 \Delta V G_0 \Delta V G_0 \Delta V G_0. \tag{71}$$

The final expression for $\mathcal{D}_{ijkl}^\alpha$ is achieved by taking the second-order derivative with respect to the wave-vector $\vec{q}_2$ and the first-order derivative with respect to the wave-vectors $\vec{q}_{1(2)}$, considering the limit $q_{1(2)} \to 0$, i.e. equating within the *ab initio* and model expressions the corresponding terms proportional to $(\vec{R}_i - \vec{R}_j)_y (\vec{R}_k - \vec{R}_l)_x^2$ and $(\vec{R}_i - \vec{R}_j)_x (\vec{R}_k - \vec{R}_l)_x^2$ (we keep a similar form in both cases for the sake of convenience) gives the elements $\mathcal{D}_{ijkl}^{y,x}$ and $\mathcal{D}_{ijkl}^{y,y}$, as well as $\mathcal{D}_{ijkl}^{x,x}$ and $\mathcal{D}_{ijkl}^{x,y}$, respectively, of the four-spin chiral interaction as follows

$$\mathcal{D}_{ijkj}^{\alpha,\beta} = \epsilon_{\alpha\gamma} \frac{1}{8\pi} \mathrm{Im} \, \mathrm{Tr} \int^{E_F} dE(E - E_F) \left[\underline{O}^i \, \underline{\tau}^{ij} \underline{T}^{j,\gamma} \, \underline{\tau}^{jk} \underline{T}^{k,\beta} \, \underline{\tau}^{kl} \underline{T}^{l,\beta} \, \underline{\tau}^{li} - \underline{T}^{i,\gamma} \, \underline{\tau}^{ij} \underline{O}^j \underline{\tau}^{jk} \underline{T}^{k,\beta} \, \underline{\tau}^{kl} \underline{T}^{l,\beta} \, \underline{\tau}^{li} \right]$$

$$+ \left[\underline{O}^i \, \underline{\tau}^{ij} \underline{T}^{j,\beta} \, \underline{\tau}^{jk} \underline{T}^{k,\beta} \, \underline{\tau}^{kl} \underline{T}^{l,\gamma} \, \underline{\tau}^{li} - \underline{T}^{i,\gamma} \, \underline{\tau}^{ij} \underline{T}^{j,\beta} \underline{\tau}^{jk} \underline{T}^{k,\beta} \, \underline{\tau}^{kl} \underline{O}^l \, \underline{\tau}^{li} \right] \tag{72}$$

with $\alpha, \beta = x, y$, and $\epsilon_{\alpha\gamma}$ the elements of the transverse Levi-Civita tensor $\underline{\epsilon} = \begin{bmatrix} 0 & 1 \\ -1 & 0 \end{bmatrix}$. The TDMI and BDMI parameters can be obtained as the special cases $l = j$ and $l = j, k = i$, respectively, from equation (72).
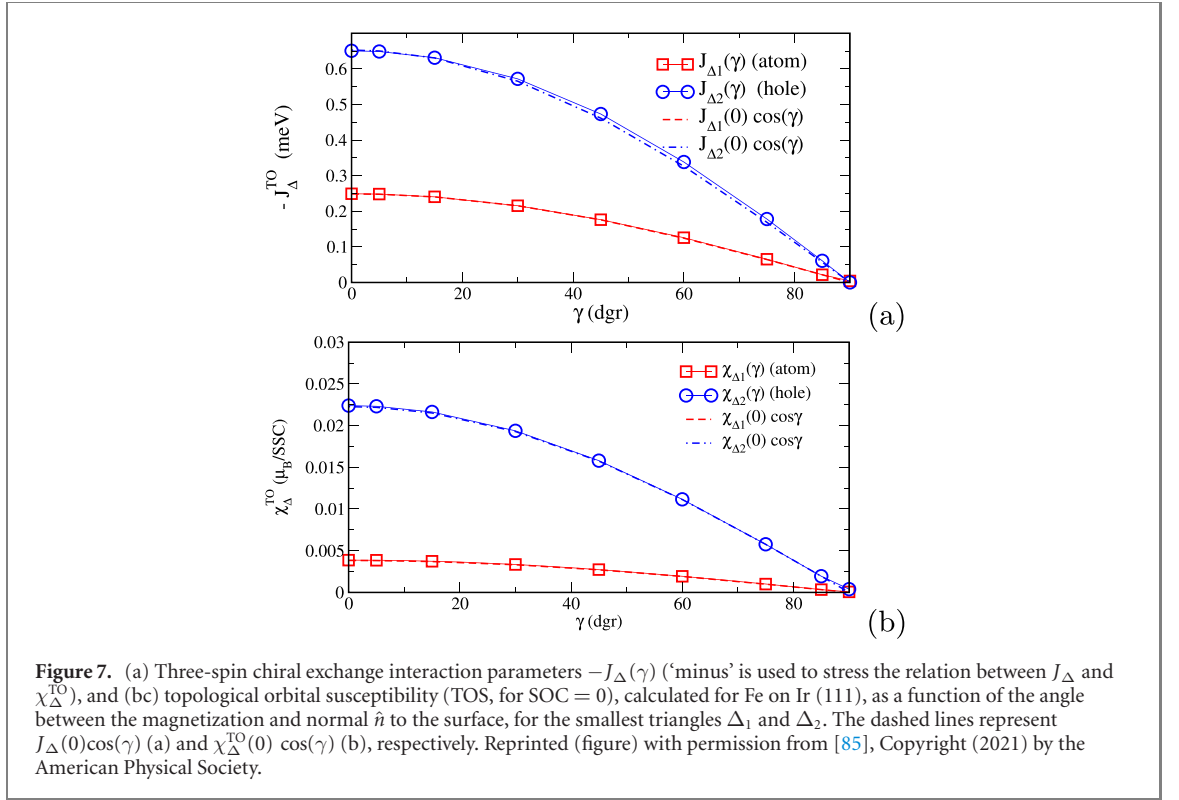
The expression in equation (72) gives access to the x and y components of the DMI-like three-spin interactions

$$\mathcal{D}_{ijkj}^\alpha = \mathcal{D}_{ijkj}^{\alpha,x} + \mathcal{D}_{ijkj}^{\alpha,y}. \tag{73}$$

Finally, three-spin chiral exchange interaction (TCI) represented by first term in the extended spin Hamiltonian has been discussed in reference [55]. As it follows from this expression, the contribution due to this type of interaction is non-zero only in case of a non-co-planar and non-collinear magnetic structure characterized by the scalar spatial type product $\hat{s}_i \cdot (\hat{s}_j \times \hat{s}_k)$ involving the spin moments on three different lattice sites.

In order to work out the expression for the J_{ijk} interaction, one has to use a multi-Q spin modulation [82–84] which ensure a non-zero scalar spin chirality for every three atoms. The energy contribution due to the TCI, is non-zero only if $J_{ijk} \neq J_{ikj}$, etc. Otherwise, the terms ijk and ikj cancel each other due to the relation $\hat{s}_i \cdot (\hat{s}_j \times \hat{s}_k) = -\hat{s}_i \cdot (\hat{s}_k \times \hat{s}_j)$.

Accordingly, the expression for the TCI is derived using the 2Q non-collinear spin texture described by equation (70), which is characterized by two wave vectors oriented along two mutually perpendicular directions, as for example $\vec{q}_1 = (0, q_y, 0)$ and $\vec{q}_2 = (q_x, 0, 0)$. Applying such a spin modulation in equation (70) for the term H^3 associated with the three-spin interaction in the spin Hamiltonian in equation (62), the second-order derivative of the energy $E^{(3)}(\vec{q}_1, \vec{q}_2)$ with respect to the wave vectors q_1 and q_2 is given in the limit $q_1 \to 0$,

Figure 7. (a) Three-spin chiral exchange interaction parameters $-J_\Delta(\gamma)$ ('minus' is used to stress the relation between J_Δ and χ_Δ^{TO}), and (bc) topological orbital susceptibility (TOS, for SOC = 0), calculated for Fe on Ir (111), as a function of the angle between the magnetization and normal $\hat{n}$ to the surface, for the smallest triangles Δ_1 and Δ_2. The dashed lines represent $J_\Delta(0)\cos(\gamma)$ (a) and $\chi_\Delta^{TO}(0)\cos(\gamma)$ (b), respectively. Reprinted (figure) with permission from [85], Copyright (2021) by the American Physical Society.

$q_2 \to 0$ by the expression

$$\frac{\partial^2}{\partial \vec{q}_1 \partial \vec{q}_2} H^{(3)} = -\sum_{i \neq j \neq k} J_{ijk}\left(\hat{z} \cdot [(\vec{R}_i - \vec{R}_j) \times (\vec{R}_k - \vec{R}_j)]\right). \tag{74}$$

The microscopic energy term of the electron system, giving access to the chiral three-spin interaction in the spin Hamiltonian is described by the second-order term

$$\Delta \mathcal{E}^{(2)} = -\frac{1}{\pi} \text{Im Tr} \int^{E_F} dE(E - E_F)\, G_0 \Delta V G_0 \Delta V G_0 \tag{75}$$

of the free energy expansion. Taking the first-order derivative with respect to q_1 and q_2 in the limit $q_1 \to 0$, $q_2 \to 0$, and equating the terms proportional to $\left(\hat{z} \cdot [(\vec{R}_i - \vec{R}_j) \times (\vec{R}_k - \vec{R}_j)]\right)$ with the corresponding terms in the spin Hamiltonian, one obtains the following expression for the three-spin interaction parameter

$$J_{ijk} = \frac{1}{8\pi} \text{Im Tr} \int^{E_F} dE(E - E_F) \left[\underline{T}^{i,x}\, \underline{\tau}^{ij} \underline{T}^{j,y}\, \underline{\tau}^{jk} \underline{Q}^k\, \underline{\tau}^{ki} - \underline{T}^{i,y}\, \underline{\tau}^{ij} \underline{T}^{j,x}\, \underline{\tau}^{jk} \underline{Q}^k\, \underline{\tau}^{ki} \right.$$

$$- \underline{T}^{i,x}\, \underline{\tau}^{ij} \underline{Q}^j\, \underline{\tau}^{jk} \underline{T}^{k,y}\, \underline{\tau}^{ki} + \underline{T}^{i,y}\, \underline{\tau}^{ij} \underline{Q}^j\, \underline{\tau}^{jk} \underline{T}^{k,x}\, \underline{\tau}^{ki}$$

$$\left. + \underline{Q}^i\, \underline{\tau}^{ij} \underline{T}^{i,x}\, \underline{\tau}^{jk} \underline{T}^{k,y}\, \underline{\tau}^{ki} - \underline{Q}^j\, \underline{\tau}^{ij} \underline{T}^{i,y}\, \underline{\tau}^{jk} \underline{T}^{k,x}\, \underline{\tau}^{ki} \right], \tag{76}$$

giving access to the three-spin chiral interaction determined as $J_\Delta = J_{ijk} - J_{ikj}$. Its interpretation was discussed in reference [85], where its dependence on the SOC as well as on the topological orbital susceptibility $\chi_\Delta^{TO} = \chi_{ijk}^{TO} - \chi_{ikj}^{TO}$ was demonstrated. In fact that the expression for χ_{ijk}^{TO} worked out in reference [85] has a rather similar form as J_{ijk}, as that can be seen from the expression

$$\chi_{ijk}^{TO} = -\frac{1}{4\pi} \text{Im Tr} \int^{E_F} dE \left[\underline{T}^{i,x}\, \underline{\tau}^{ij} \underline{T}^{j,y}\, \underline{\tau}^{jk} \underline{l}_z^k\, \underline{\tau}^{ki} - \underline{T}^{i,y}\, \underline{\tau}^{ij} \underline{T}^{j,x}\, \underline{\tau}^{jk} \underline{l}_z^k\, \underline{\tau}^{ki} - \underline{T}^{i,x}\, \underline{\tau}^{ij} \underline{l}_z^j\, \underline{\tau}^{jk} \underline{T}^{k,y}\, \underline{\tau}^{ki} \right.$$

$$\left. + \underline{T}^{i,y}\, \underline{\tau}^{ij} \underline{l}_z^j\, \underline{\tau}^{jk} \underline{T}^{k,x}\, \underline{\tau}^{ki} + \underline{l}_z^i\, \underline{\tau}^{ij} \underline{T}^{j,x}\, \underline{\tau}^{jk} \underline{T}^{k,y}\, \underline{\tau}^{ki} - \underline{l}_z^i\, \underline{\tau}^{ij} \underline{T}^{j,y}\, \underline{\tau}^{jk} \underline{T}^{k,x}\, \underline{\tau}^{ki} \right]. \tag{77}$$

For every trimer of atoms, both quantities, χ_{ijk}^{TO} and J_{ijk}, are non-zero only in the case of non-zero scalar spin chirality $\hat{s}_i \cdot (\hat{s}_j \times \hat{s}_k)$ and depend on the orientation of the trimer magnetic moment with respect to the trimer plain. This is shown in figure 7 [85] representing ΔJ and $\Delta \chi^{TO}$ as a function of the angle between the magnetization and normal $\hat{n}$ to the surface, which are calculated for the two smallest trimers, Δ_1 and Δ_2, centered at the Ir atom and the hole site in the Ir surface layer for 1ML Fe/Ir(111), respectively (figure 8).
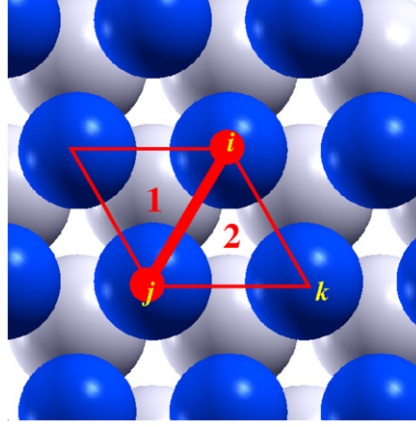
Figure 8. Geometry of the smallest three-atom clusters in the monolayer of $3d$-atoms on $M(111)$ surface ($M =$ Au, Ir): M-centered triangle Δ_1 and hole-centered triangle Δ_2. Reprinted (figure) with permission from [85], Copyright (2021) by the American Physical Society.
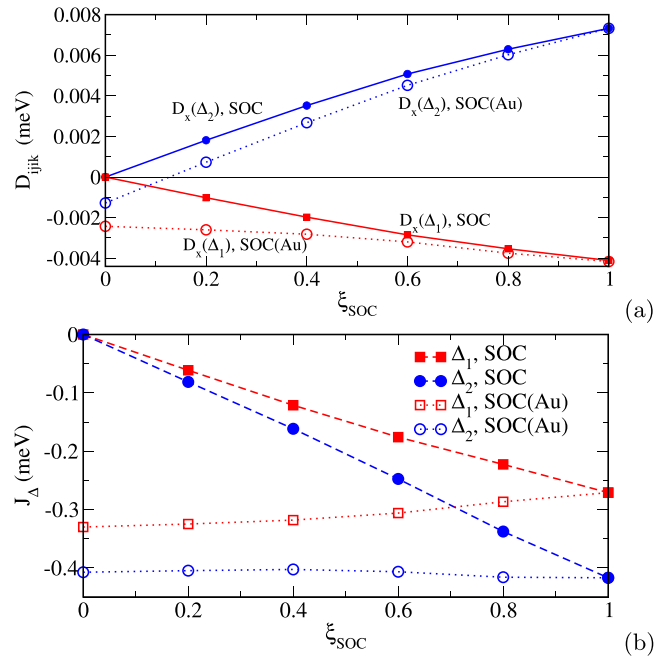

Figure 9. (a) Three-site four-spin DMI-like interaction, $\mathcal{D}^z_{ijkj}$ and (c) three-spin chiral exchange interaction (TCI) parameters J_Δ calculated for Fe on Au (111) on the basis of equation (76) as a function of SOC scaling parameter ξ_{SOC} for the smallest triangles Δ_1 and Δ_2. In figure (b), full symbols represent the results obtained when scaling the SOC for all elements in the system, while open symbols show the results when scaling only the SOC for Au. Reprinted (figure) with permission from [85], Copyright (2021) by the American Physical Society.

The role of the SOC for the three-site four-spin DMI-like interaction, $\mathcal{D}^z_{ijik}$, and the three-spin chiral inter-action, J_Δ is shown in figure 9. These quantities are calculated for 1ML Fe on Au (111), for the two smallest triangles Δ_1 and Δ_2 centered at an Au atom or a hole site, respectively (see figure 8). Here, setting the SOC scaling factor $\xi_{SOC} = 0$ implies a suppression of the SOC, while $\xi_{SOC} = 1$ corresponds to the fully relativis-tic case. Figure 9(a) shows the three-site four-spin DMI-like interaction parameter, $\mathcal{D}^z_{ijik}(\xi_{SOC})$ when the SOC scaling parameter ξ_{SOC} applied to all components in the system, shown by full symbols, and with the SOC scaling applied only to the Au substrate. One can see a dominating role of the SOC of substrate atoms for $\mathcal{D}^z_{ijik}$. Also in figure 9(b), a nearly linear variation can be seen for $J_\Delta(\xi_{SOC})$ when the SOC scaling parameter ξ_{SOC} is applied to all components in the system (full symbols). Similar to $\mathcal{D}^z_{ijik}$, this shows that the SOC is an ulti-mate prerequisite for a non-vanishing TCI J_Δ. When scaling the SOC only for Au (open symbols), figure 9(b) show only weak changes for the TCI parameters $J_\Delta(\xi_{SOC})$, demonstrating a minor impact of the SOC of the substrate on these interactions, in contrast to the DMI-like interaction shown in figure 9(a). One can see also that $\mathcal{D}^z_{ijik}$ is about two orders of magnitude smaller than J_Δ for this particular system.

The origin of the TCI parameters have been discussed in the literature suggesting a different interpretation of the corresponding terms derived also within the multiple-scattering theory Green function formalism [79, 86, 87]. However, the expression worked out in reference [86] has obviously not been applied for calculations so far. As pointed out in reference [85], the different interpretation of this type of interactions can be explained by their different origin. In particular, one has to stress that the parameters in references [85, 86] were derived in a different order of perturbation theory. On the other hand, the approach used for calculations of the multispin exchange parameters reported in references [79, 88] is very similar to the one used in references [55, 85]. The corresponding expressions have been worked out within the framework of multiple-scattering Green function formalism using the MFT. In particular, the Lloyd formula has been used to express the energy change due to the perturbation ΔV leading to the expression

$$\Delta \mathcal{E} = -\frac{1}{\pi} \mathrm{Im\, Tr} \int^{E_F} \mathrm{d}E \sum_p \frac{1}{p} \, \mathrm{Tr} \left[\underline{G}(E) \, \Delta \underline{V} \right]^p. \tag{78}$$

Using the off-site part of the GF in equation (36), as defined by Dederichs *et al* [48], equation (78) is transformed to the form

$$\Delta \mathcal{E} = -\frac{1}{\pi} \mathrm{Im\, Tr} \int^{E_F} \mathrm{d}E \sum_p \frac{1}{p} \, \mathrm{Tr} \left[\underline{G}^{\mathrm{str}}(E) \, \underline{\Delta t}(E) \right]^p. \tag{79}$$

By splitting the structural Green function $\underline{G}^{\mathrm{str}}_{ij}$ into a spin-dependent ($\vec{\underline{B}}^{\mathrm{str}}_{ij}$) and a spin-independent ($\underline{A}^{\mathrm{str}}_{ij}$) parts according to

$$\underline{G}^{\mathrm{str}}_{ij} = \underline{A}^{\mathrm{str}}_{ij} \sigma_0 + \vec{\underline{B}}^{\mathrm{str}}_{ij} \cdot \vec{\sigma} \tag{80}$$

and expressing the change of the single-site scattering matrix

$$\underline{\Delta t}_i(E) = (\underline{t}^{\uparrow}_i(E) - \underline{t}^{\downarrow}) \delta \hat{s}_i \times \vec{\sigma}, \tag{81}$$

by means of the rigid spin approximation, the different terms in equation (79) corresponding to different numbers p give access to corresponding multispin terms, chiral and non-chiral, in the extended spin Hamiltonian. In particular, the isotropic six-spin interactions, that are responsible for the non-collinear magnetic structure of B20-MnGe according to Grytsiuk *et al* [86], is given by the expression

$$\kappa^{\mathrm{six-spin}}_{ijklmn} = \frac{1}{3\pi} \mathrm{Im Tr} \int^{E_F} \mathrm{d}E A_{ij} t^{\sigma}_j A_{jk} t^{\sigma}_k A_{kl} t^{\sigma}_l A_{lm} t^{\sigma}_m A_{mn} t^{\sigma}_n A_{ni} t^{\sigma}_i. \tag{82}$$

A rather different point of view concerning the multispin extension of the spin Hamiltonian was adopted by Streib *et al* [89, 90], who suggested to distinguish so-called local and global Hamiltonians. According to that classification, a global Hamiltonian implies to include in principle all possible spin configurations for the energy mapping in order to calculate exchange parameters that characterize in turn the energy of any spin configuration. On the other hand, a local Hamiltonian is '*designed to describe the energetics of spin configurations in the vicinity of the ground state or, more generally, in the vicinity of a predefined spin configuration*' [89]. This implies that taking the ground state as a reference state, it has to be determined first before the calculating the exchange parameters which are in principle applicable only for small spin tiltings around the reference state and can be used e.g. to investigate spin fluctuations around the ground state spin configuration. In reference [89], the authors used a constraining field to stabilize the non-collinear magnetic configuration. This leads to the effective two-spin exchange interactions corresponding to a non-collinear magnetic spin configuration [89, 90]. According to the authors, '*local spin Hamiltonians do not require any spin interactions beyond the bilinear order (for Heisenberg exchange as well as DMIs)*'. On the other hand, they point out the limitations for these exchange interactions in the case of non-collinear system in the regime when the standard Heisenberg model is not valid [90], and multi-spin interactions get more important.

Thus, to summarize, one can say that the extended description of the spin–spin interaction beyond the bilinear approximation may be crucial for some materials. The perturbative approach, we focus on here, is especially attractive in this context as it allows a systematic extension of the interactions and in turn a corresponding improvement of the results of simulations, by adding more and more higher order multispin terms to the spin Hamiltonian without changing the contributions due to the low-order terms in the Hamiltonian. This obviously is not the case, when the energy fitting scheme is used to estimate the parameters of the spin Hamiltonian, although, both approaches should be equivalent in the limit when accounting for all multispin contributions. Note also, that a restricted number of multispin terms in the extended Hamiltonian leads to a dependence of the results on the reference state. Therefore, the reference state is often chosen according to the problem under consideration, e.g. related to the thermodynamic properties, spin-wave excitations, etc.

Finally, for systems with strongly correlated electrons requiring a description of their electronic structure beyond the LSDA-DFT, the calculations of the exchange coupling parameters may need a revised scheme taking into account many-body effects, e.g. within the DMFT approach (see, e.g. [65, 91, 92]).

5. Gilbert damping

Another parameter entering the LLG equation in equation (3) is the Gilbert damping parameter $\tilde{G}$ characterizing energy dissipation associated with the magnetization dynamics.

Theoretical investigations on the Gilbert damping parameter have been performed by various groups and accordingly the properties of GD is discussed in detail in the literature. Many of these investigations are performed assuming a certain dissipation mechanism, like Kambersky's breathing Fermi surface (BFS) [93, 94], or more general torque-correlation models (TCM) [95, 96] to be evaluated on the basis of electronic structure calculations. The earlier works in the field relied on the relaxation time parameter that represents scattering processes responsible for the energy dissipation. Only few computational schemes for Gilbert damping parameter account explicitly for disorder in the systems, which is responsible for the spin-flip scattering process. This issue was addressed in particular by Brataas *et al* [97] who described the Gilbert damping mechanism by means of scattering theory. This development supplied the formal basis for the first parameter-free investigations on disordered alloys [56, 57, 98].

A formalism for the calculation of the Gilbert damping parameter based on linear response theory has been reported in reference [57] and implemented using fully relativistic multiple scattering or Korringa–Kohn–Rostoker (KKR) formalism. Considering the FM state as a reference state of the system, the energy dissipation can be expressed in terms of the GD parameter by:

$$\dot{E}_{\text{mag}} = \vec{H}_{\text{eff}} \cdot \frac{\mathrm{d}\vec{M}}{\mathrm{d}\tau} = \frac{1}{\gamma^2}\dot{\vec{m}}[\tilde{G}(\vec{m})\dot{\vec{m}}]. \tag{83}$$

On the other hand, the energy dissipation in the electronic system is determined by the underlying Hamiltonian $\hat{H}(\tau)$ as follows $\dot{E}_{\text{dis}} = \left\langle \frac{\mathrm{d}\hat{H}}{\mathrm{d}\tau} \right\rangle$. Assuming a small deviation of the magnetic moment from the equilibrium $\vec{u}(\tau)$, the normalized magnetization $\vec{m}(\tau)$ can be written in a linearized form $\vec{m}(\tau) = \vec{m}_0 + \vec{u}(\tau)$, that in turn leads to the linearized time dependent electronic Hamiltonian $\hat{H}(\tau)$

$$\hat{H} = \hat{H}_0(\vec{m}_0) + \sum_{\mu} \vec{u}_{\mu} \frac{\partial}{\partial \vec{u}_{\mu}} \hat{H}(\vec{m}_0). \tag{84}$$

As shown in reference [56], the energy dissipation within the linear response formalism is given by:

$$\dot{\mathcal{E}}_{\text{dis}} = -\pi\hbar \sum_{ij} \sum_{\mu\nu} \dot{u}_{\mu}\dot{u}_{\nu} \left\langle \psi_i \left| \frac{\partial \hat{H}}{\partial u_{\mu}} \right| \psi_j \right\rangle \left\langle \psi_j \left| \frac{\partial \hat{H}}{\partial u_{\nu}} \right| \psi_i \right\rangle \delta(E_{\text{F}} - E_i)\delta(E_{\text{F}} - E_j). \tag{85}$$

Identifying it with the corresponding phenomenological quantity in equation (83), $\dot{E}_{\text{mag}} = \dot{\mathcal{E}}_{\text{dis}}$ one obtains for the GD parameter α^G a Kubo–Greenwood-like expression:

$$\alpha_{\mu\nu}^G = -\frac{\hbar\gamma}{\pi M_s} \text{Trace} \left\langle \frac{\partial \hat{H}}{\partial u_{\mu}} \operatorname{Im} G^+(E_{\text{F}}) \frac{\partial \hat{H}}{\partial u_{\nu}} \operatorname{Im} G^+(E_{\text{F}}) \right\rangle_{\text{c}}, \tag{86}$$
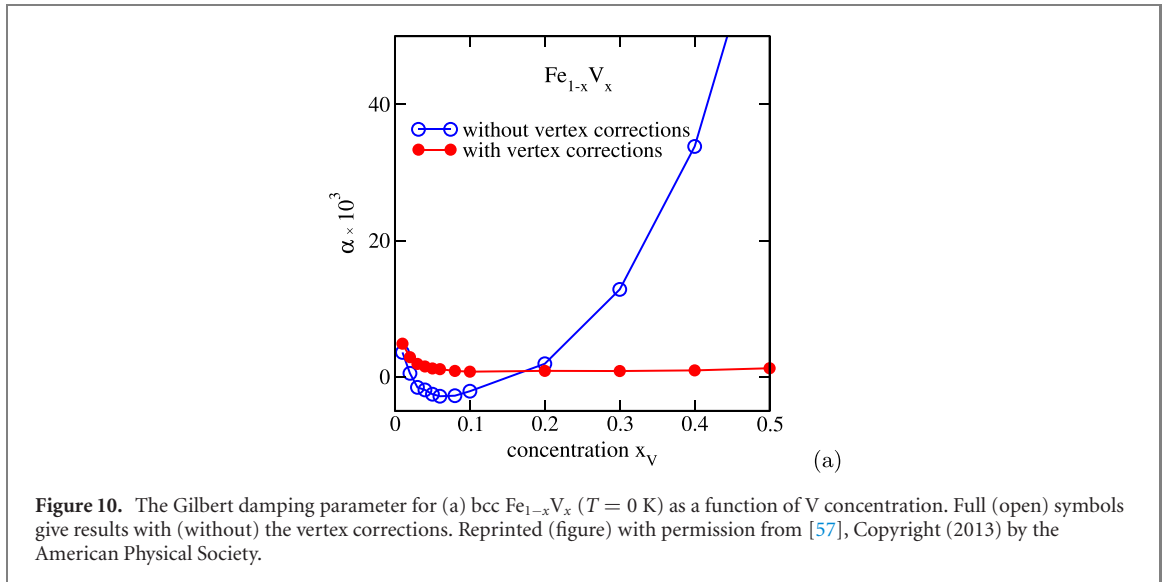
where $\alpha^G = \tilde{G}/(\gamma M_s)$, and $\langle \ldots \rangle_{\text{c}}$ indicates a configurational average required in the presence of chemical or thermally induced disorder responsible for the dissipation processes. Within the multiple scattering formalism with the representation of the Green function given by equation (9), equation (86) leads to

$$\alpha_{\mu\mu}^G = \frac{g}{\pi\mu_{\text{tot}}} \sum_{n} \text{Trace} \left\langle \underline{T}^{0\mu} \tilde{\underline{\tau}}^{0n} \underline{T}^{n\mu} \tilde{\underline{\tau}}^{n0} \right\rangle_{\text{c}} \tag{87}$$

with the g-factor $2(1 + \mu_{\text{orb}}/\mu_{\text{spin}})$ in terms of the spin and orbital moments, μ_{spin} and μ_{orb}, respectively, the total magnetic moment $\mu_{\text{tot}} = \mu_{\text{spin}} + \mu_{\text{orb}}$, and $\tilde{\tau}_{\Lambda\Lambda'}^{0n} = \frac{1}{2i}(\tau_{\Lambda\Lambda'}^{0n} - \tau_{\Lambda'\Lambda}^{0n})$ and with the energy argument E_F omitted. The matrix elements $\underline{T}^{n\mu}$ are identical to those occurring in the context of exchange coupling [6] and can be expressed in terms of the spin-dependent part B of the electronic potential with matrix elements:

$$T_{\Lambda'\Lambda}^{n\mu} = \int \mathrm{d}^3 r \, Z_{\Lambda'}^{n\times}(\vec{r}) \left[\beta\sigma_{\mu}B_{xc}(\vec{r}) \right] Z_{\Lambda}^n(\vec{r}). \tag{88}$$

As is discussed in reference [57], for a system having chemical disorder, the configurational average is performed using the scattering path operators evaluated on the basis of the coherent potential approximation

Figure 10. The Gilbert damping parameter for (a) bcc $Fe_{1-x}V_x$ ($T = 0$ K) as a function of V concentration. Full (open) symbols give results with (without) the vertex corrections. Reprinted (figure) with permission from [57], Copyright (2013) by the American Physical Society.

(CPA) alloy theory. In the case of thermally induced disorder, the alloy analogy model is used, which was discussed already above. When evaluating equation (87), the so-called vertex corrections have to be included [61] that accounts for the difference between the averages $\langle T_\mu \operatorname{Im} G^+ T_\nu \operatorname{Im} G^+ \rangle_c$ and $\langle T_\mu \operatorname{Im} G^+ \rangle_c \langle T_\nu \operatorname{Im} G^+ \rangle_c$. Within the Boltzmann formalism these corrections account for scattering-in processes.

The crucial role of these corrections is demonstrated [57] in figure 10 representing the Gilbert damping parameter for an $Fe_{1-x}V_x$ disordered alloy as a function of the concentration *x*, calculated with and without vertex corrections. As one can see, neglect of the vertex corrections may lead to the nonphysical result $\alpha^G < 0$. This wrong behavior does not occur when the vertex corrections are included, that obviously account for energy transfer processes connected with scattering-in processes.
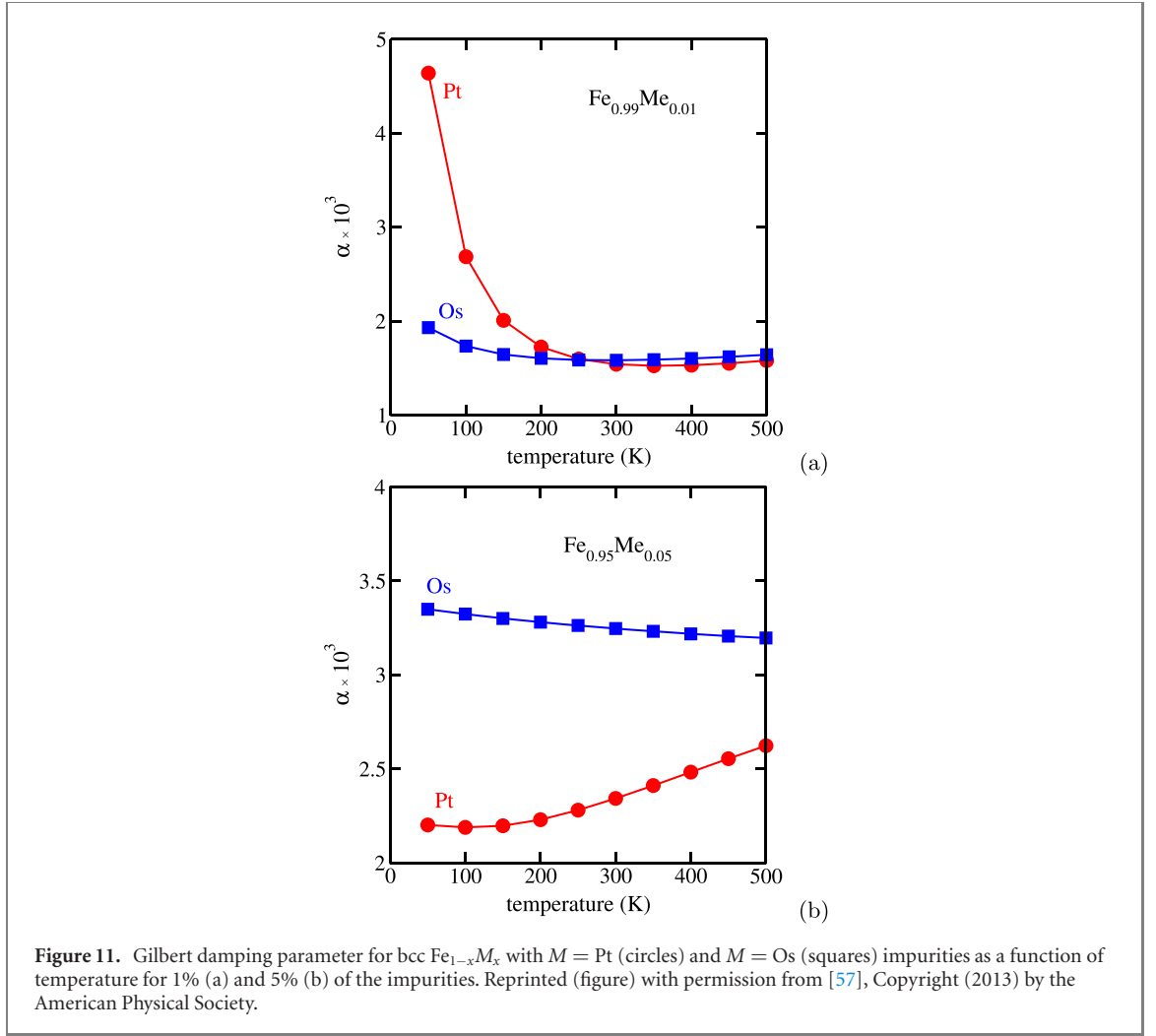
The impact of thermal vibrations onto the Gilbert damping can be taken into account within the alloy-analogy model (see above) by averaging over a discrete set of thermal atom displacements for a given temperature *T*. Figure 11 represents the temperature dependent behavior of the Gilbert damping parameter α^G for bcc Fe with 1% and 5% of impurities of Os and Pt [56, 57]. One can see a strong impact of impurities on GD. In the case of 1% of Pt in figure 11(a), α^G decreases in the low-temperature regime much steeper upon increasing the temperature, indicating that the breathing Fermi surface mechanism dominates. When the concentration of the impurities increases up to 5% (figure 11(a)), the spin-flip scattering mechanism takes the leading role for the magnetization dissipation practically for the whole region of temperatures under consideration. The different behavior of GD for Fe with Os and Pt is a result of the different density of states (DOS) of the impurities at the Fermi energy (see reference [57] for a discussion).

An alternative approach to elaborate the expression for the Gilbert damping tensor based on a generalized Kubo–Středa formula was reported by Pervishko *et al* [99]. The authors focus on the Gilbert damping at the interface of ferromagnetic (FM) and non-magnetic (NM) metallic subsystems. In the case of semiconducting or insulating substrates the Gilbert damping enhancement may be associated with the enhanced Rashba spin–orbit interaction, as was demonstrated by Tu *et al* [100] for the CoFeB/GaAs(001) interface. However, in the case of a metallic interface, the delocalized conduction electrons of the non-magnetic subsystem are responsible for the enhancement of the magnetization dissipation when compared to bulk materials as was demonstrated both experimentally [101, 102] and theoretically [98, 103–105]. Pervishko *et al* [99] consider a model FM/NM bilayer system with the conduction electrons of the NM coupled via *sd* exchange interaction, Δ, with the *d* electrons of the FM subsystem. Describing the non-equilibrium spin density of the conduction electrons of the NM film $s_\mu(\vec{r}, t)$ (driven by the time-dependent magnetization $\vec{m}(\vec{r}, t)$ of the FM film) in terms of the lesser Green function, $G^<(\vec{r}t; \vec{r}t)$, i.e.,

$$s_\mu(\vec{r}, t) = -\frac{i}{2} \operatorname{Tr}[\sigma_\mu G^<(\vec{r}t; \vec{r}t)] = \sum_\nu Q_{\mu\nu} \partial_t m_\nu + \cdots, \tag{89}$$

the authors work out an expression for the response function $Q_{\mu\nu} = Q_{\mu\nu}^1 + Q_{\mu\nu}^2$. The first term $Q_{\mu\nu}^1$ is the so-called Středa contribution

$$Q_{\mu\nu}^1 = \frac{1}{4} \operatorname{Tr}\left[\sigma_\mu \int_{-\infty}^\infty \frac{dE}{2\pi} f(E) \int \frac{d^2p}{(2\pi)^2}\left(\frac{\partial g_{\vec{p}}^R}{\partial E} \sigma_\nu g_{\vec{p}}^R - g_{\vec{p}}^R \sigma_\nu \frac{\partial g_{\vec{p}}^R}{\partial E} + g_{\vec{p}}^A \sigma_\nu \frac{\partial g_{\vec{p}}^A}{\partial E} - \frac{\partial g_{\vec{p}}^A}{\partial E} \sigma_\nu g_{\vec{p}}^A\right)\right], \tag{90}$$

Figure 11. Gilbert damping parameter for bcc $Fe_{1-x}M_x$ with $M = Pt$ (circles) and $M = Os$ (squares) impurities as a function of temperature for 1% (a) and 5% (b) of the impurities. Reprinted (figure) with permission from [57], Copyright (2013) by the American Physical Society.

which involves an integration over the whole Fermi sea. The additional term $Q^2_{\mu\nu}$ represents the conventional contribution due to the electrons at the Fermi level

$$Q^2_{\mu\nu} = \frac{1}{4} \text{Tr} \left[\sigma_\mu \int_{-\infty}^{\infty} \frac{dE}{2\pi} \partial f(E) \partial E \int \frac{d^2 p}{(2\pi)^2} \left(g_{\vec{p}}^R \sigma_\nu g_{\vec{p}}^R + g_{\vec{p}}^A \sigma_\nu g_{\vec{p}}^A - 2 g_{\vec{p}}^R \sigma_\nu g_{\vec{p}}^A \right) \right], \quad (91)$$

with $g_{\vec{p}}^R$, $g_{\vec{p}}^A$ and $g_{\vec{p}}^<$ the bare retarded, advanced, and lesser Green functions respectively. The diagonal and off-diagonal elements of the response function tensor lead to a renormalization of the gyromagnetic ratio γ and the damping parameter α^G, according to

$$\bar{\gamma} = \frac{\gamma}{1 + \chi \Delta Q_{xy}} \quad \bar{\alpha}^G = -\frac{\chi \Delta Q_{xx}}{1 + \chi \Delta Q_{xy}}, \quad (92)$$

where $\chi = (g \mu_B / \hbar)^2 \mu_0 / d$, $g = 2$ the electron g-factor and d the thickness of the nonmagnetic layer. The authors point out that their expression for the Gilbert damping worked out on the basis of Kubo–Středa formula is more accurate when compared to the Kubo–Greenwood approach discussed above, as it takes into account the Fermi sea contribution $Q^1_{\mu\nu}$ missing in that theory.

Note that the expression for the Gilbert damping in terms of the response function in equations (90) and (91) is similar to that derived previously by Freimuth *et al* [104] based on a one-dimensional Rashba model Hamiltonian. According to reference [104], the Gilbert damping is given by the expression

$$\bar{\alpha}^G = \frac{S_{xx}}{|A_{xy}|}, \quad (93)$$

in terms of even $(S(\hat{m}) = S(-\hat{m}))$ and odd $(S(\hat{m}) = -S(-\hat{m}))$, under magnetization reversal, parts of the linear response tensor $\Lambda_{\mu\nu}$, where

$$S_{\mu\nu}(\hat{m}) = \frac{1}{2} \left[\Lambda_{\mu\nu}(\hat{m}) + \Lambda_{\mu\nu}(-\hat{m}) \right], \quad (94)$$

and

$$A_{\mu\nu}(\hat{m}) = \frac{1}{2}\left[\Lambda_{\mu\nu}(\hat{m}) - \Lambda_{\mu\nu}(-\hat{m})\right]. \tag{95}$$

Similar to equations (90) and (91), the linear response tensor $\Lambda_{\mu\nu} = \Lambda_{\mu\nu}^{Ia} + \Lambda_{\mu\nu}^{Ib} + \Lambda_{\mu\nu}^{II}$ is composed of the Fermi surface $\Lambda_{\mu\nu}^{Ia}$, $\Lambda_{\mu\nu}^{Ib}$ and Fermi sea $\Lambda_{\mu\nu}^{II}$ terms, where

$$\Lambda_{\mu\nu}^{Ia} = \frac{1}{h}\int\frac{\mathrm{d}^d k}{(2\pi)^d}\,\mathrm{Tr}\,\langle\mathcal{T}_\alpha G_k^R(E_\mathrm{F})\mathcal{T}_\beta G_k^A(E_\mathrm{F})\rangle, \tag{96}$$

$$\Lambda_{\mu\nu}^{Ia} = -\frac{1}{h}\int\frac{\mathrm{d}^d k}{(2\pi)^d}\mathrm{ReTr}\,\langle\mathcal{T}_\alpha G_k^R(E_\mathrm{F})\mathcal{T}_\beta G_k^R(E_\mathrm{F})\rangle, \tag{97}$$

$$\Lambda_{\mu\nu}^{II} = \frac{1}{h}\int\frac{\mathrm{d}^d k}{(2\pi)^d}\int_{-\infty}^{E_\mathrm{F}}\mathrm{d}E\,\mathrm{Re}\,\mathrm{Tr}\left\langle\mathcal{T}_\alpha G_k^R(E_\mathrm{F})\mathcal{T}_\beta\frac{G_k^A(E_\mathrm{F})}{\mathrm{d}E} - \mathcal{T}_\alpha\frac{G_k^A(E_\mathrm{F})}{\mathrm{d}E}\mathcal{T}_\beta G_k^R(E_\mathrm{F})\right\rangle. \tag{98}$$

Here d stands for the dimensionality ($d = 1$, 2 or 3), $G_k^R(E_\mathrm{F})$ and $G_k^A(E_\mathrm{F})$ are the retarded and advanced Green functions, and $\mathcal{T}_\alpha$ the α component of the torque operator. The authors point out that the tensor $\Lambda_{\mu\nu}^{II}$ is antisymmetric under the interchange of the indices μ and ν, therefore the Gilbert damping tensor, which is symmetric, is contributed by only $\Lambda_{\mu\nu}^{Ia}$ and $\Lambda_{\mu\nu}^{Ib}$ terms, although it depends also on $|\Lambda_{\mu\nu}^{II}|$.

The role of the electron–phonon scattering for the ultrafast laser-induced demagnetization was investigated by Carva *et al* [106] based on the Elliott–Yafet theory of spin relaxation in metals, that puts the focus on spin-flip (SF) transitions upon the electron–phonon scattering. As the evaluation of the spin-dependent electron–phonon matrix elements entering the expression for the rate of the spin-flip transition is a demanding problem, various approximations are used for this. In particular, Carva *et al* [106, 107] use the so-called Elliott approximation to evaluate a SF probability $P_S^b = \frac{\tau}{\tau_{sf}}$ with the spin lifetime τ_{sf} and a spin-diagonal lifetime τ:

$$P_S^b = \frac{\tau}{\tau_{\mathrm{sf}}} = 4\langle b^2\rangle \tag{99}$$

with the Fermi-surface averaged spin mixing of Bloch wave eigenstates

$$\langle b^2\rangle = \sum_{\sigma,n}\int \mathrm{d}^3k\,|b_{kn}^\sigma|\delta(E_{kn}^\sigma - E_\mathrm{F}). \tag{100}$$
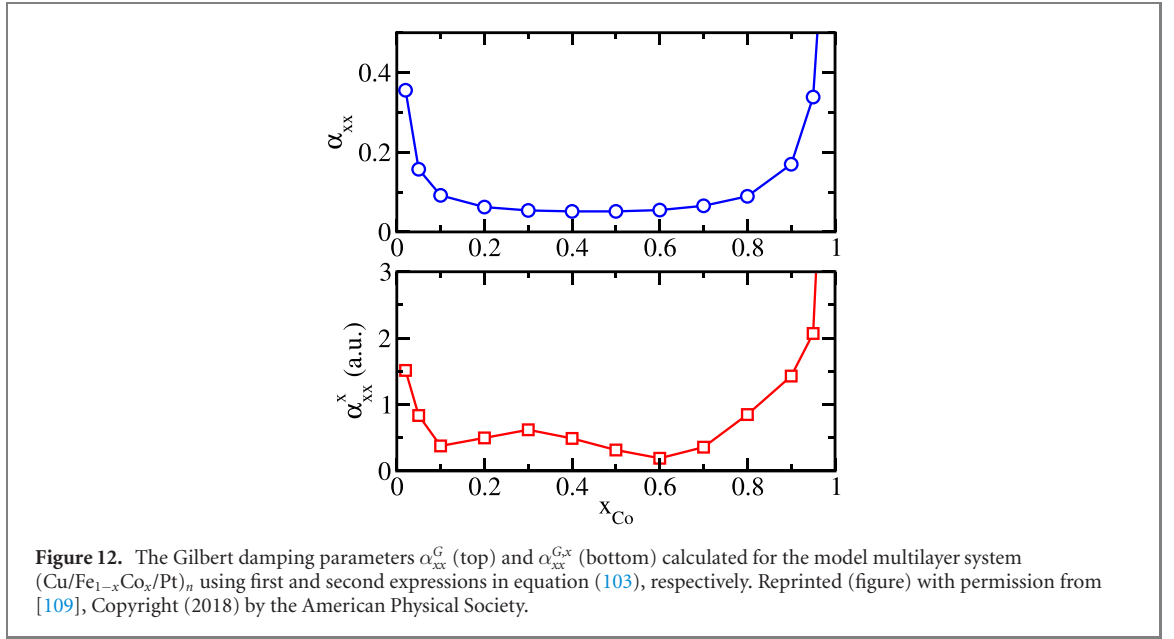
In the case of a non-collinear magnetic structure, the description of the Gilbert damping can be extended by adding higher-order non-local contributions. The role of non-local damping contributions has been investigated by calculating the precession damping $\alpha^G(\vec{q})$ for magnons in FM metals, characterized by a wave vector $\vec{q}$. Following the same idea, Thonig *et al* [108] used a torque–torque correlation model based on a tight binding approach, and calculated the Gilbert damping for the itinerant-electron ferromagnets Fe, Co and Ni, both in the reciprocal, $\alpha^G(\vec{q})$, and real α_{ij}^G space representations. The important role of non-local contributions to the GD for spin dynamics has been demonstrated using atomistic magnetization dynamics simulations [109]. Using linear response theory for weakly-noncollinear magnetic systems it gives access to the GD parameters represented as a function of a wave vector $\vec{q}$. Using the definition for the spin susceptibility tensor $\chi_{\alpha\beta}(\vec{q},\omega)$, the Fourier transformation of the real-space Gilbert damping can be represented by the expression [110, 111]

$$\alpha_{\alpha\beta}^G(\vec{q}) = \frac{\gamma}{M_0 V}\lim_{\omega\to 0}\frac{\partial\Im[\chi^{-1}]_{\alpha\beta}(\vec{q},\omega)}{\partial\omega}. \tag{101}$$

Here $\gamma = g\mu_\mathrm{B}$ is the gyromagnetic ratio, $M_0 = \mu_\mathrm{tot}\mu_\mathrm{B}/V$ is the equilibrium magnetization and V is the volume of the system. As is shown in reference [109], this expression can be transformed to the form which allows an expansion of GD in powers of wave vector $\vec{q}$:

$$\underline{\alpha}^G(\vec{q}) = \underline{\alpha}^G + \sum_\mu\underline{\alpha}^{G,\mu}q_\mu + \frac{1}{2}\sum_{\mu\nu}\underline{\alpha}^{G,\mu\nu}q_\mu q_\nu + \cdots, \tag{102}$$

where $\underline{\alpha}^G = \frac{1}{4}\left(\underline{\alpha}^{G,++} + \underline{\alpha}^{G,--} - \underline{\alpha}^{G,+-} - \underline{\alpha}^{G,-+}\right)$ with the following expansion coefficients:

Figure 12. The Gilbert damping parameters α_{xx}^G (top) and $\alpha_{xx}^{G,x}$ (bottom) calculated for the model multilayer system $(Cu/Fe_{1-x}Co_x/Pt)_n$ using first and second expressions in equation (103), respectively. Reprinted (figure) with permission from [109], Copyright (2018) by the American Physical Society.

$$\alpha_{\alpha\alpha}^{G,0\pm\pm} = \frac{g}{\pi\mu_{\text{tot}}}\frac{1}{\Omega_{BZ}}\,\text{Tr}\int d^3k \left\langle \underline{T}_\beta\underline{\mathcal{T}}(\vec{k},E_F^\pm)\underline{T}_\beta\underline{\mathcal{T}}(\vec{k},E_F^\pm)\right\rangle_c$$

$$\alpha_{\alpha\alpha}^{G,\mu\pm\pm} = \frac{g}{\pi\mu_{\text{tot}}}\frac{1}{\Omega_{BZ}}\,\text{Tr}\int d^3k \left\langle \underline{T}^\beta\frac{\partial\underline{\mathcal{T}}(\vec{k},E_F^\pm)}{\partial k_\mu}\underline{T}^\beta\underline{\mathcal{T}}(\vec{k},E_F^\pm)\right\rangle_c$$

$$\alpha_{\alpha\alpha}^{G,\mu\nu\pm\pm} = -\frac{g}{2\pi\mu_{\text{tot}}}\frac{1}{\Omega_{BZ}}\,\text{Tr}\int d^3k \left\langle \underline{T}^\beta\frac{\partial\underline{\mathcal{T}}(\vec{k},E_F^\pm)}{\partial k_\mu}\underline{T}^\beta\frac{\partial\underline{\mathcal{T}}(\vec{k},E_F^\pm)}{\partial k_\nu}\right\rangle_c. \quad (103)$$

For the prototype multilayer system $(Cu/Fe_{1-x}Co_x/Pt)_n$ the calculated zero-order (uniform) GD parameter α_{xx}^G and the corresponding first-order (chiral) $\alpha_{xx}^{G,x}$ correction term for $\vec{q}\,\|\hat{x}$ are plotted in figure 12 top and bottom, respectively, as a function of the Co concentration x. Both terms, α_{xx}^G and $\alpha_{xx}^{G,x}$, increase approaching the pure limits w.r.t. the $Fe_{1-x}Co_x$ alloy subsystem. As is pointed out in reference [109], this increase is associated with the dominating so-called breathing Fermi-surface damping mechanism due to the modification of the Fermi surface (FS) induced by the SOC, which follows the magnetization direction that slowly varies with time. As $\underline{\alpha}^G$ is caused for a ferromagnet exclusively by the SOC one can expect that it vanishes for vanishing SOC. This was indeed demonstrated before [57]. The same holds also for $\underline{\alpha}^{G,x}$ that is caused by SOC as well.

Alternatively, a real-space extension for classical Gilbert damping tensor was proposed recently by Brinker *et al* [112], by introducing two-site Gilbert damping tensor $\underline{\mathcal{G}}_{ij}$ entering the site-resolved LLG equation

$$\frac{1}{\gamma}\frac{d\vec{M}_i}{d\tau} = -\gamma\vec{M}_i\times\left(\vec{H}_{i,\text{eff}} + \sum_j\left[\underline{\mathcal{G}}_{ij}(\vec{M})\cdot\frac{d\vec{M}_i}{d\tau}\right]\right), \quad (104)$$

which is related to the inverse dynamical susceptibility $\underline{\chi}_{ij}$ via the expression

$$\frac{d}{d\omega}\text{Im}[\underline{\chi}]_{ij}^{\alpha\beta} = \delta_{ij}\left(\frac{1}{\gamma M_i}\epsilon_{\alpha\beta\gamma}\right) + \left(\mathcal{R}_i\underline{\mathcal{G}}_{ij}\mathcal{R}_j^T\right)_{\alpha\beta}, \quad (105)$$

where $\mathcal{R}_i$ and $\mathcal{R}_j$ are the rotation matrices to go from the global to the local frames of reference for atoms i and j, respectively, assuming a non-collinear magnetic ground state in the system. Thus, an expression for the GD can be directly obtained using the adiabatic approximation for the slow spin-dynamics processes. This justifies the approximation $([\chi]^{-1}(\omega))'_\omega \approx ([\underline{\chi}_0]^{-1}(\omega))'_\omega$, with the un-enhanced dynamical susceptibility given in terms of electronic Green function G_{ij}

$$\chi_{ij}^{\alpha\beta}(\omega+i\eta) = -\frac{1}{\pi}\text{Tr}\int^{E_F}dE\big[\sigma^\alpha G_{ij}(E+\omega+i\eta)\sigma^\beta\text{Im}\,G_{ij}(E) + \sigma^\alpha G_{ij}(E)\sigma^\beta\text{Im}\,G_{ij}(E-\omega-i\eta)\big], \quad (106)$$

with the Green function $G(E\pm i\eta) = (E-\mathcal{H}\pm i\eta)^{-1}$ corresponding to the Hamiltonian $\mathcal{H}$.

Moreover, this approach allows a multisite expansion of the GD accounting for higher-order non-local contributions for non-collinear structures [112]. For this purpose, the Hamiltonian $\mathcal{H}$ is split into the on-site

contribution $\mathcal{H}_0$ and the intersite hopping term t_{ij}, which is spin dependent in the general case. The GF can then be expanded in a perturbative way using the Dyson equation

$$G_{ij} = G_i^0 \delta_{ij} + G_i^0 t_{ij} G_j^0 + G_i^0 t_{ik} G_k^0 t_{kj} G_j^0 + \cdots .$$ (107)

As a result, the authors generalize the LLG equation by splitting the Gilbert damping tensor in terms proportional to scalar, anisotropic, vector-chiral and scalar-chiral products of the magnetic moments, i.e. terms like $\hat{e}_i \cdot \hat{e}_j$, $(\hat{n}_{ij} \cdot \hat{e}_i)(\hat{n}_{ij} \cdot \hat{e}_j)$, $\hat{n}_{ij} \cdot (\hat{e}_i \times \hat{e}_j)$, etc.

Note that chiral properties of the Gilbert damping in the presence of a noncollinear magnetic structure have been discussed also by Freimuth *et al* [104] for a one-dimensional Rashba model, for which the authors have demonstrated that the Gilbert damping differs between left-handed and right-handed Néel-type magnetic domain walls.

It should be stressed that the Gilbert damping parameter accounts for the energy transfer connected with the magnetization dynamics but gives no information on the angular momentum transfer that plays an important role e.g. for ultrafast demagnetization processes. The formal basis to account simultaneously for the spin and lattice degrees of freedom was considered recently by Aßmann and Nowak [113] and Hellsvik *et al* [114]. Hellsvik *et al* [114, 115] report on an approach solving simultaneously the equations for spin and lattice dynamics, accounting for spin–lattice interactions in the Hamiltonian, calculated on a first-principles level. These interactions appear as a correction to the exchange coupling parameters due to atomic displacements. As a result, this leads to the three-body spin–lattice coupling parameters $\Gamma_{ijk}^{\alpha\beta\mu} = \frac{\partial J_{ij}^{\alpha\beta}}{\partial u_k^\mu}$ and four-body parameters $\Lambda_{ijkl}^{\alpha\beta\mu\nu} = \frac{\partial J_{ij}^{\alpha\beta}}{\partial u_k^\mu \partial u_l^\nu}$ represented by rank 3 and rank 4 tensors, respectively, entering the spin–lattice Hamiltonian

$$\mathcal{H}_{sl} = -\frac{1}{2} \sum_{i,j,k,\alpha\beta,\mu} \Gamma_{ijk}^{\alpha\beta\mu} e_i^\alpha e_j^\beta u_k^\mu - \frac{1}{4} \sum_{i,j,k,l,\alpha\beta,\mu,\nu} \Lambda_{ijkl}^{\alpha\beta\mu\nu} e_i^\alpha e_j^\beta u_k^\mu u_l^\nu .$$ (108)

The parameters $\Gamma_{ijk}^{\alpha\beta\mu}$ in reference [114] are calculated using a finite difference method, using the exchange coupling parameters $\underline{J}_{ij}$ for the system without displacements ($\underline{J}_{ij}^0$) and with a displaced atom k ($\underline{J}_{ij}^\Delta(\vec{u}_k)$), used to estimate the coefficient $\Gamma_{ijk}^{\alpha\beta\mu} \approx \frac{(\underline{J}_{ij}^\Delta(\vec{u}_k) - \underline{J}_{ij}^0)}{u_\mu}$.

Alternatively, to describe the coupling of spin and spatial degrees of freedom the present authors (see reference [116]) adopt an atomistic approach and start with the expansion of a phenomenological spin–lattice Hamiltonian

$$\mathcal{H}_{sl} = -\sum_{i,j,\alpha,\beta} \sum_{k,\mu} J_{ij,k}^{\alpha\beta,\mu} e_i^\alpha e_j^\beta u_k^\mu - \sum_{i,j} \sum_{k,l} J_{ij,kl}^{\alpha\beta,\mu\nu} e_i^\alpha e_j^\beta u_k^\mu u_l^\nu ,$$ (109)

that can be seen as a lattice extension of a Heisenberg model. Accordingly, the spin and lattice degrees of freedom are represented by the orientation vectors $\hat{e}_i$ of the magnetic moments $\vec{m}_i$ and displacement vectors $\vec{u}_i$ for each atomic site i. The spin–lattice Hamiltonian in equation (109) is restricted to three and four-site terms. As relativistic effects are taken into account, the SLC is described in tensorial form with $J_{ij,k}^{\alpha\beta,\mu}$ and $J_{ij,kl}^{\alpha\beta,\mu\nu}$ represented by rank 3 and rank 4 tensors, similar to those discussed by Hellsvik *et al* [114].

The same strategy as for the exchange coupling parameters J_{ij} [4] or $J_{ij}^{\alpha\beta}$ [5, 6], is used to map the free energy landscape $\mathcal{F}(\{\hat{e}_i\}, \{\vec{u}_i\})$ accounting for its dependence on the spin configuration $\{\hat{e}_i\}$ as well as atomic displacements $\{\vec{u}_i\}$, making use of the MFT and the Lloyd formula to evaluate integrated DOS $\Delta N(E)$. With this, the free energy change due to any perturbation in the system is given by equation (26).
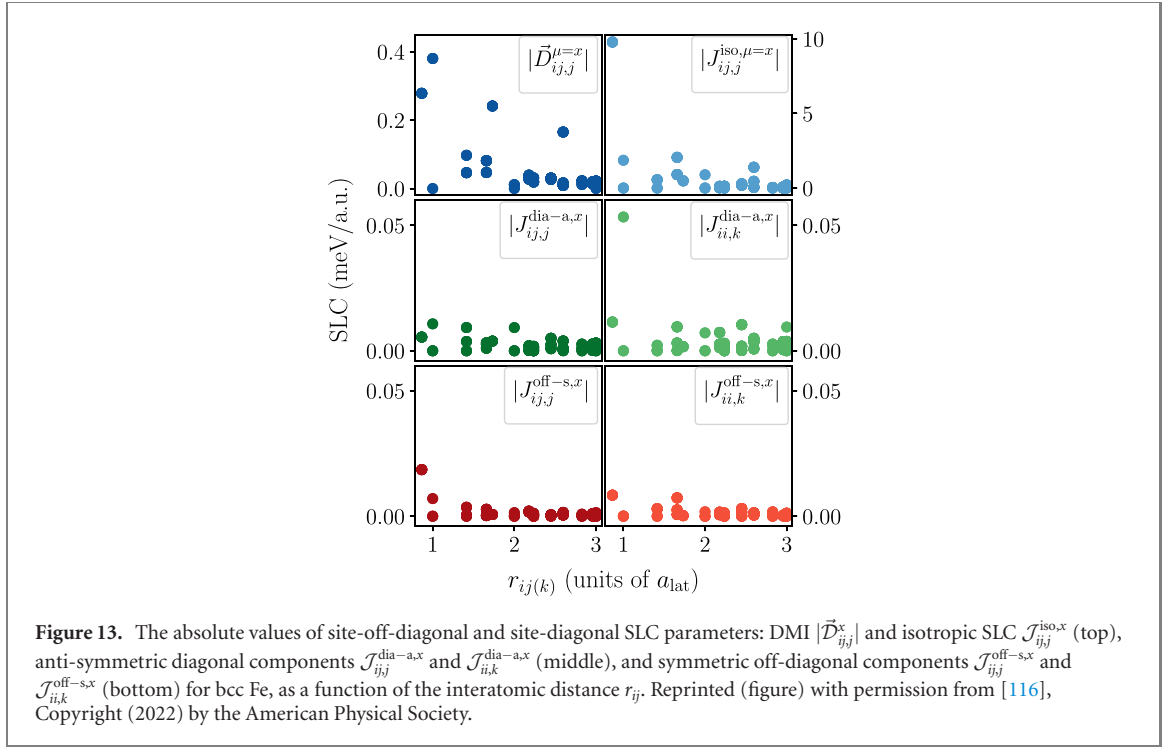
Using as a reference the ferromagnetically ordered state of the system with a non-distorted lattice, and the perturbed state characterized by finite spin tiltings $\delta \hat{e}_i$ and finite atomic displacements $\vec{u}_i$ at site i, one can write the corresponding changes of the inverse t-matrix as $\Delta_\mu^s \underline{m}_i = \underline{m}_i(\delta \hat{e}_i^\mu) - \underline{m}_i^0$ and $\Delta_\nu^u \underline{m}_i = \underline{m}_i(u_i^\nu) - \underline{m}_i^0$. This allows to replace the integrand in equation (11) by

$$\ln \underline{\tau} - \ln \underline{\tau}^0 = -\ln\left(1 + \underline{\tau} \left[\Delta_\mu^s \underline{m}_i + \Delta_\nu^u \underline{m}_j + \cdots\right]\right),$$ (110)

where all site-dependent changes in the spin configuration $\{\hat{e}_i\}$ and atomic positions $\{\vec{u}_i\}$ are accounted for in a one-to-one manner by the various terms on the right-hand side. Due to the use of the MFT these blocks may be written in terms of the spin tiltings $\delta \hat{e}_i^\mu$ and atomic displacements of the atoms u_i^ν together with the corresponding auxiliary matrices $\underline{T}_i^\mu$ and $\mathcal{U}_i^\nu$, respectively, as

$$\Delta_\mu^s \underline{m}_i = \delta \hat{e}_i^\mu \underline{T}_i^\mu ,$$ (111)

$$\Delta_\nu^u \underline{m}_i = u_i^\nu \underline{\mathcal{U}}_i^\nu .$$ (112)

Figure 13. The absolute values of site-off-diagonal and site-diagonal SLC parameters: DMI $|\vec{\mathcal{D}}_{ij,j}^{x}|$ and isotropic SLC $\mathcal{J}_{ij,j}^{\text{iso},x}$ (top), anti-symmetric diagonal components $\mathcal{J}_{ij,j}^{\text{dia}-\text{a},x}$ and $\mathcal{J}_{ii,k}^{\text{dia}-\text{a},x}$ (middle), and symmetric off-diagonal components $\mathcal{J}_{ij,j}^{\text{off}-\text{s},x}$ and $\mathcal{J}_{ii,k}^{\text{off}-\text{s},x}$ (bottom) for bcc Fe, as a function of the interatomic distance r_{ij}. Reprinted (figure) with permission from [116], Copyright (2022) by the American Physical Society.

Inserting these expressions into equation (110) and the result in turn into equation (26) allows us to calculate the parameters of the spin–lattice Hamiltonian as the derivatives of the free energy with respect to tilting angles and displacements. This way one gets for example for the three-site term:

$$\mathcal{J}_{ij,k}^{\alpha\beta,\mu} = \frac{\partial^3 \mathcal{F}}{\partial e_i^\alpha \, \partial e_j^\beta \, \partial u_k^\mu} = \frac{1}{2\pi} \text{ImTr} \int^{E_\text{F}} dE \Big[\underline{T}_i^\alpha \, \underline{\tau}_{ij} \underline{T}_j^\beta \, \underline{\tau}_{jk} \underline{\mathcal{U}}_k^\mu \underline{\tau}_{ki} + \underline{T}_i^\alpha \, \underline{\tau}_{ik} \underline{\mathcal{U}}_k^\mu \underline{\tau}_{kj} \underline{T}_j^\beta \, \underline{\tau}_{ji} \Big] \tag{113}$$

and for the four-site term:

$$\mathcal{J}_{ij,kl}^{\alpha\beta,\mu\nu} = \frac{\partial^4 \mathcal{F}}{\partial e_i^\alpha \, \partial e_j^\beta \, \partial u_k^\mu \partial u_l^\nu} = \frac{1}{4\pi} \text{ImTr} \int^{E_\text{F}} dE \Big[\underline{\mathcal{U}}_k^\mu \, \underline{\tau}_{ki} \underline{T}_i^\alpha \, \underline{\tau}_{ij} \underline{T}_j^\beta \, \underline{\tau}_{jl} \underline{\mathcal{U}}_l^\nu \, \underline{\tau}_{lk} + \underline{T}_i^\alpha \, \underline{\tau}_{ik} \underline{\mathcal{U}}_k^\mu \, \underline{\tau}_{kj} \underline{T}_j^\beta \, \underline{\tau}_{jl} \underline{\mathcal{U}}_l^\nu \, \underline{\tau}_{li}$$

$$+ \underline{\mathcal{U}}_k^\mu \, \underline{\tau}_{ki} \underline{T}_i^\alpha \, \underline{\tau}_{il} \underline{\mathcal{U}}_l^\nu \, \underline{\tau}_{lj} \underline{T}_j^\beta \, \underline{\tau}_{jk} + \underline{T}_i^\alpha \, \underline{\tau}_{ik} \underline{\mathcal{U}}_k^\mu \, \underline{\tau}_{kl} \underline{\mathcal{U}}_l^\nu \, \underline{\tau}_{lj} \underline{T}_j^\beta \, \underline{\tau}_{ji} \Big]. \tag{114}$$

Figure 13 shows corresponding results for the SLC parameters of bcc Fe, plotted as a function of the distance r_{ij} for $i = k$ which implies that a displacement along the *x* direction is applied for one of the interacting atoms. The absolute values of the DMI-like SLC parameters (DSLC) $|\vec{\mathcal{D}}|_{ij,k}^{\mu=x}$ (note that $\mathcal{D}_{ij,k}^{z,\mu} = \frac{1}{2}(\mathcal{J}_{ij,k}^{xy,\mu} - \mathcal{J}_{ij,k}^{yx,\mu})$) show a rather slow decay with the distance r_{ij}. The isotropic SLC parameters $\mathcal{J}_{ij,j}^{\text{iso},\mu=x}$, which have only a weak dependence on the SOC, are about one order of magnitude larger than the DSLC. All other SOC-driven parameters shown in figure 13, characterizing the displacement-induced contributions to MCA, are much smaller than the DSLC.

To summarize, linear response theory describes the Gilbert damping of ferromagnetic materials well, leading to results in rather good agreement, e.g., with the results from FMR experiments [57, 117]. Nowadays, however, many investigations are focused on the dynamical properties of non-collinear magnetic structures, e.g. magnetic domain walls, magnetic skyrmion textures, etc, which are of great interest for various spintronic applications. The development of appropriate theoretical methods is required, accounting for specific features of the magnetization dissipation in non-collinear magnetic systems, as was discussed in the present sections. In the case of ultrafast magnetization dynamics, it is of great importance to establish the demagnetization mechanism, which obviously may be different for different materials and depends on whether they have a metallic or an insulating character. Several competing mechanisms discussed in the literature as those responsible for ultrafast demagnetization, are mentioned in this section and are associated with the spin-flip electron–phonon or magnon–phonon scattering events [118, 119]. It is however still a challenge for further investigations, which mechanism is dominating in different materials.

6. Summary

To summarize, we have considered a multi-level atomistic approach commonly used to simulate finite temperature and dynamical magnetic properties of solids, avoiding in particular time-consuming TD-SDFT calculations. The approach is based on a phenomenological parameterized spin Hamiltonian which allows to separate the spin and orbital degrees of freedom and that way to avoid the demanding treatment of complex spin-dependent many-body effects. As these parameters are fully determined by the electronic structure of a system, they can be deduced from the information provided by relativistic band structure calculations based on SDFT. We gave a short overview of the various methods to calculate these parameters entering for example the LLG equation. It is shown that the KKR Green function formalism is one of the most powerful band structure methods as it gives straightforward access to practically all parameters of the phenomenological models. It allows in particular to add in a very simple way further extensions to the model Hamiltonians, accounting for example for multi-site interaction terms. Another important issue are spin–lattice interactions, that couple the degrees of freedom of the spin and lattice subsystems. The key role of the SOC for the interaction parameters is pointed out as it gives not only rise to the MCA but also to the Gilbert damping as well as the anisotropy of the exchange coupling and spin–lattice interaction with many important physical phenomena connected to these.

Data availability statement

No new data were created or analysed in this study.

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