Extension of the standard Heisenberg Hamiltonian to multispin exchange interactions

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An extension of the Heisenberg Hamiltonian is discussed, that allows to go beyond the standard bilinear spin Hamiltonian taking into account various contributions due to multispin interactions having both chiral and non-chiral character. The parameters of the extended Hamiltonian are calculated from first principles within the framework of the multiple scattering Green function formalism giving access to an explicit representation of these parameters in real space. The discussions are focused on the inter-atomic exchange interactions, i.e. biquadratic and three-spin Dzyaloshinskii-Moriya like vector interactions $\mathbf{D}_{ij}$ (BDMI) and $\mathbf{D}_{ijk}$ (TDMI), respectively, as well as three-spin chiral interaction (TCI) $J_{ijk}$. Some properties of these interactions are demonstrated by calculations for realistic materials. In particular, the non-relativistic character of the three-spin chiral interaction $J_{ijk}$ is demonstrated that is in contrast to the TDMI. As the magnitude of these chiral interactions can be quite sizable, they can lead to a stabilization of a noncollinear magnetic texture in some materials that is absent when these interactions are neglected. In particular, Monte Carlo simulations for some model systems demonstrate the stabilization of a skyrmion lattice due to DMI and CTI, without external magnetic field.

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I. INTRODUCTION

The Heisenberg spin Hamiltonian is nowadays a rather popular tool providing a bridge between the electronic structure of magnetic materials and their spin-dynamical and finite-temperature magnetic properties. However, restriction of the classical model to only isotropic bilinear exchange parameters is not always able to describe successfully the experimental findings. In this case an extension of Heisenberg model is used to take into account specific features of the system under consideration. This concerns in particular the impact of spin-orbit coupling (SOC) leading to magnetocrystalline anisotropy (MCA) and to the spin-space anisotropy of the exchange coupling described by an exchange tensor $L_{ij}$ instead of scalar parameters. The latter one can often be reduced to the chiral Dzyaloshinskii-Moriya (DM) vector $\mathbf{D}_{ij}$ representing the antisymmetric part of the exchange tensor $L_{ij}$.

Still, this form of the Hamiltonian implies for example neglecting the dependence of the exchange parameters on the relative orientation of the magnetic moments in the system. To go beyond this bilinear approximation for the inter-atomic exchange interactions, one can take into account higher-order contributions to the Heisenberg Hamiltonian, i.e., biquadratic, fourth-order three-spin, four-spin interactions, etc. terms [1–9].

The origin of higher-order interactions was discussed already many years ago by various authors [10–12], focusing on those being isotropic in spin space. Obviously, the dominating mechanism responsible for these terms can be different for different materials. Kittel [10] discussing the transition from the antiferromagnetic (AFM) to the ferromagnetic (FM) state in metamagnetic materials (including metals) suggested an important role of the biquadratic exchange interaction due to exchange magnetostriction caused by a dependence of the exchange interaction on the volume during an AFM/FM transition. MacDonald et al. [12] discussed the Hubbard model Hamiltonian, which can be transformed in the limit of large on-site Coulomb interaction $U$ and assuming half-filling of the electron energy bands implying electron localization around atomic sites to a form equivalent to the Heisenberg spin Hamiltonian. An expansion of the Hamiltonian in powers of the ratio $t/U$ gives access to high-order terms of the spin Hamiltonian with bilinear and four-spin exchange interactions $\sim t^2/U$ and $\sim t^4/U^3$, respectively [12–14]. The three-spin term should vanish as it is antisymmetric with respect to time reversal transformation. Tanaka and Uryu [11] have derived the four-spin interactions based on the Heitler-London theory by expanding the ground state energy in terms of the overlap integrals between the orbitals of electrons located at different lattice sites. Detailed calculations of bilinear and biquadratic exchange interactions within a real-space tight-binding framework have been performed for FM Fe by Spisak and Hafner [15] who demonstrate a significant contribution of the biquadratic exchange interactions to the Curie temperature.

During the last decade the interest in skyrmions grew rapidly because their specific magnetic texture stabilized by chiral spin interactions makes them attractive for various spintronic applications (see e.g. [16–18]). Most investigations in the field were restricted to the bilinear Dzyaloshinskii-Moriya interaction (DMI) and focused on materials for which a strong DMI can be expected [18–20].

The DMI is caused by spin-orbit coupling (SOC) and
is non-zero in non-centrosymmetric systems only. Competing with isotropic FM or AFM interactions it leads to a deviation from the collinear magnetic state by creating a helimagnetic structure in the absence of an external magnetic field, characterized by a non-zero vector spin chirality \( \vec{\chi}_{ij} = \vec{s}_i \times \vec{s}_j \).

Recently, first-principles investigations have been performed going beyond the bilinear approximation, taking into account higher-order chiral interactions [21, 22] in the extended Heisenberg model. The calculation of the chiral biquadratic DMI-like interaction (BDMI) for deposited dimers [21] has demonstrated that its magnitude can be comparable to that of the conventional bilinear Dzyaloshinskii-Moriya interaction (DMI), implying the non-negligible role of biquadratic contributions. In addition, the first-principles investigations on the magnetic properties of Fe monatomic chains on a Re(0001) substrate have shown [22] that chiral four-spin interactions can be responsible for the opposite chirality of the spin spirals when compared to that determined by DMI.

Another type of chiral interaction, the three-spin chiral interaction (TCI) term, was considered when discussing the formation of chiral magnetic phases in different frustrated magnetic systems either due to geometric frustration or created by the competition of FM and AFM interactions [23–25]. This three-spin interaction term in the Heisenberg Hamiltonian, \( \sum_{ijk} J_{ijk} \, \vec{s}_i \cdot (\vec{s}_j \times \vec{s}_k) \), gives a non-zero contribution only for a non-coplanar magnetic structure, i.e. in the case of non-zero scalar chirality \( \chi_{ijk} = \vec{s}_k \cdot (\vec{s}_i \times \vec{s}_j) \). This can lead to the transition to a chiral spin liquid state, for which the time-reversal symmetry is broken spontaneously by the appearance of long-range order of scalar chirality even in the absence of long-range magnetic order or an external magnetic field. Describing a transition in a frustrated quantum spin system from a spin liquid to a chiral spin liquid state within the framework of the Hubbard model, it was shown that expanding the Hubbard Hamiltonian in powers of \( t/U \) leads to a third-order term which is proportional to the flux \( \Phi_{ijk} \) enclosed by the three-spin loop [26, 27]. This term becomes non-zero in the case of broken time-reversal and parity symmetry, caused by the spontaneous development of spin chirality characterizing the chiral spin liquid state [28–31]. This approach results in an explicit expression for the three-spin interaction term [26, 27] with \( J_{ijk} = (24/U^2) |t_{ij}||t_{jk}| |t_{ki}| \sin(\Phi_{ijk}/\Phi_0) \) (with \( \Phi_0 = \hbar c/e \)) that enters the spin Hamiltonian [25, 29, 31, 32]. Despite many theoretical investigations on such a transition, only recently Machida et al. [33] have reported about the experimental observation of the time-reversal symmetry broken spontaneously on a macroscopic scale in the absence of long-range spin magnetic order, by measuring the spontaneous Hall effect associated with chiral spin textures.

Discussing skyrmion-hosting materials, the formation of skyrmion magnetic texture is usually ascribed to the DMI, implying the lack of the inversion symmetry in these systems. However, recently it was suggested that the magnetic frustration could stabilize skyrmions even in materials with centrosymmetric lattices. This idea was proposed and discussed by various authors within theoretical investigations [14, 34, 35]. In these works complex superstructures or the skyrmion-lattice state are characterized by multiple ordering wave vectors (multiple-Q), allowing to characterize a non-coplanar magnetic structure via a double-Q description. This approach applied to metallic systems allowed to demonstrate that the non-coplanar vortex state can be stabilized having lower energy than the helimagnetic structure expected due to RKKY interactions [36]. Solenov et al. [36] showed that such a non-coplanar state can be stabilized even in the absence of SOC, i.e. without the DMI. The authors attribute this feature of a double-Q state to the chirality-induced emergent magnetic field associated with a persistent electric current in such systems (see e.g. [37–39]), which is proportional to the scalar chirality in the system. Considering magnetic properties in terms of the extended spin Hamiltonian, the above mentioned property can be attributed to the three-spin interaction term also proportional to the scalar chirality in the system. The impact of higher order terms going beyond the bilinear level and their anisotropy is discussed on the basis of corresponding numerical results for various systems.

II. ELECTRONIC STRUCTURE

Following our previous work [41], we consider the change of the grand canonical potential caused by the formation of a modulated spin structure seen as a perturbation. This quantity is represented in terms of the Green function \( G_0(E) \) for the FM reference state and its modification due to the perturbation. Neglecting all temperature effects, and denoting the corresponding change in the Green function \( \Delta G(E) \) one can write for the change in energy:

\[
\Delta \mathcal{E} \approx -\frac{1}{\pi} \text{Im} \text{Tr} \int_{E_F}^{E_F} dE \left( E - E_F \right) \Delta G(E),
\]

with the expansion

\[
\Delta G(E) = G_0 \Delta V G_0 + G_0 \Delta V G_0 \Delta V G_0 + G_0 \Delta V G_0 \Delta V G_0 \Delta V G_0 + \ldots,
\]

for \( \Delta G(E) \), where \( \Delta V \) is the perturbation operator associated with the modulated spin structure. For the sake
of readability we dropped the energy argument for the unperturbed Green function \(G_0(E)\).

Using the FM state as a reference state, the perturbation connected with the tilting of rigid magnetic moments on lattice sites \(i\) has the real space representation [41, 42]

\[
\Delta V(\vec{r}) = \sum_i \beta (\vec{\sigma} \cdot \vec{m}_i - \sigma_z) B_{xc}(\vec{r}) ,
\]

where \(B_{xc}(\vec{r})\) is the spin-dependent part of the exchange-correlation potential and all other quantities have their usual meaning [43, 44]. It is assumed here that \(B_{xc}(\vec{r})\) on site \(i\) is aligned along the orientation of the spin moment \(\vec{m}_i\), i.e. \(B_{xc}(\vec{r}) = B_{xc}(\vec{r})\delta_{ii}\).

A very convenient and flexible way to represent the electronic Green function \(G_0(E)\) in Eqs. (1)-(2) is provided by the so-called KKR (Korringa-Kohn-Rostoker) or multiple-scattering formalism. Adopting this approach a real space expression for \(G_0(\vec{r},\vec{r}';E)\) can be written in a fully relativistic way as [44]:

\[
G_0(\vec{r},\vec{r}', E) = \sum_{\Lambda_1\Lambda_2} Z_{\Lambda_1}^\alpha(\vec{r}, E) Z_{\Lambda_2}^{\alpha\times}(\vec{r}', E) \delta_{\vec{r}, \vec{r}'} - \sum_{\Lambda_1} \int_{\Lambda_1} Z_{\Lambda_1}^\alpha(\vec{r}, E) J_{\Lambda_1}^{\alpha\times}(\vec{r}', E) \Theta(\vec{r}' - \vec{r}) J_{\Lambda_1}^{\alpha}(\vec{r}, E) Z_{\Lambda_1}^{\alpha\times}(\vec{r}', E) \Theta(\vec{r} - \vec{r}') \delta_{\vec{r}, \vec{r}'}
\]

Here \(Z_{\Lambda_1}^\alpha(\vec{r}, E)\) and \(J_{\Lambda_1}^{\alpha}(\vec{r}, E)\) are the regular and irregular solutions of the single site Dirac equation and \(\tau^{\alpha\times}\) is the so-called scattering path operator matrix [44]. Substituting the expression in Eq. (4) into Eq. (2) and using Eq. (1) one obtains in a straight and natural way a real space expression for the energy change \(\Delta E\), which will be used below to derive expressions for the exchange coupling parameters entering the extended Heisenberg Hamiltonian.

III. FOUR-SPIN EXCHANGE INTERACTIONS

Extending the spin Hamiltonian to go beyond the classical Heisenberg model, we discuss first the four-spin exchange interaction term, which can be written in general form as follows

\[
H^{(4)} = -\sum_{i,j,k,l} \sum_{\alpha,\beta,\gamma,\delta} J_{ijkl}^{\alpha\beta\gamma\delta} \hat{s}_i^\alpha \hat{s}_j^\beta \hat{s}_k^\gamma \hat{s}_l^\delta ,
\]

with the site indices \(i, j, k, l\) and cartesian coordinates \(\alpha, \beta, \gamma, \delta\). The four-spin exchange interactions given by a fourth-rank tensor \(J_{ijkl}^{\alpha\beta\gamma\delta}\) which accounts also for pair \((k = i, l = j, \text{so-called biquadratic})\) and three-spin \((l = j)\) interactions. The tensor elements \(J_{ijkl}^{\alpha\beta\gamma\delta}\) can be calculated using the fourth-order term of the Green function expansion in Eq. (2). Substituting this expression into Eq. (1) and using the sum rule \(\frac{dG}{dE} = -GG\) for the Green function, one obtains after integration by parts the forth-order term of the total energy change \(\Delta E^{(4)}\) given by:

\[
\Delta E^{(4)} = -\frac{1}{\pi} \text{Im} \text{Tr} \int_{E_F}^{E_F} dE \times \Delta V G_0 \Delta V G_0 \Delta V G_0 \Delta V G_0 .
\]

Using the ferromagnetic state with \(\vec{M}||\hat{z}\) as a reference state, and considering the spin-spiral \(\vec{m}_i = (\sin \theta \cos \varphi \cdot \vec{R}_i, \sin \theta \sin \varphi \cdot \vec{R}_i, \cos \theta)\) as the source for the perturbation \(\Delta V\), at small \(\varphi\) values, only the \(x\) and \(y\) components of the exchange tensor get involved (see also Ref. [41]). Following the scheme used to derive an expression for the bilinear exchange parameters [41], the fourth-order derivative with respect to different components of the \(\varphi\)-vector give the elements of the exchange tensor represented via the expression

\[
J_{ijkl}^{\alpha\beta\gamma\delta} = -\frac{1}{\pi} \text{Im} \text{Tr} \int_{E_F}^{E_F} dE \times \left[ T_{ij}^{\alpha\alpha}(E) \tau^{\beta\beta}(E) \tau^{\gamma\gamma}(E) \tau^{\delta\delta}(E) \right],
\]

where the matrix elements of the torque operator \(T_{ij}^{\alpha\alpha}\) are defined as follows:[45]

\[
T_{ij}^{\alpha\alpha} = \int_{\Omega_1} d^3r Z_{ij}^{\alpha\times}(\vec{r}, E) \left[ \beta \sigma_i \sigma_j B_{xc}(\vec{r}) \right] Z_{ij}^{\alpha\times}(\vec{r}, E) .
\]

A. Non-chiral exchange interactions

The four-spin scalar interaction, and as special cases, also the fourth-order three-spin term with \(l = j\) and the biquadratic exchange interaction term with \(k = i\) and \(l = j\), can also be written in a form often used in the literature, i.e. they can be represented in terms of scalar products of spin directions:

\[
H^{(4)}_s = -\sum_{i,j,k,l} J_{ijkl}^{s} \hat{s}_i \cdot \hat{s}_j \hat{s}_k \cdot \hat{s}_l .
\]

The exchange interaction parameters written in the form of a 4-rank tensor in Eq. (5) give access to non-chiral as well as to chiral four-spin interactions. The parameters \(J_{ijkl}^{s}\) (where \(s\) means ‘symmetric’) are represented by the symmetric part of the exchange tensor of 4-th rank in Eq. (7) and are given by the expression

\[
J_{ijkl}^{s} = \frac{1}{4} \left( J_{ijkl}^{xx} + J_{ijkl}^{yy} + J_{ijkl}^{xy} + J_{ijkl}^{yx} \right) .
\]

The biquadratic exchange interaction terms (with \(k = i\) and \(l = j\)) can be seen as a linear term of an expansion of the bilinear exchange parameters in powers of \(\hat{s}_i \cdot \hat{s}_j\), in order to take into account the dependence of these exchange parameters on the relative orientation of the
interacting spin magnetic moments on sites $i$ and $j$. Focusing first on the scalar-interaction terms, this leads to the expression
\[ H^s = -\sum_{ij} J_{ij}(\hat{s}_i \cdot \hat{s}_j) \]
\[ = -\sum_{ij} J_{ij}(\hat{s}_i \cdot \hat{s}_j) - \sum_{ij} J'_{ij}(\hat{s}_i \cdot \hat{s}_j)(\hat{s}_i \cdot \hat{s}_j). \] (11)

**B. Chiral multispin DMI-like exchange interactions**

Discussing chiral interactions, we start with the exchange interactions represented by the vector characterizing the DMI-like interaction between two spin moments, $i$ and $j$, but taking into account the magnetic configuration of surrounding atoms, leading to the extension of the Heisenberg Hamiltonian written in the following form
\[ H^{a(4)} = -\sum_{ijkl} \Delta D_{ijkl} \cdot (\hat{s}_i \times \hat{s}_j)(\hat{s}_k \cdot \hat{s}_l). \] (12)
Assuming the magnetization direction of the reference system along the $z$ axis, we distinguish between the $x$ and $y$ components of this chiral interaction on the one hand side, and its $z$ component on the other hand as they require different approaches for their calculation. This is in full analogy to the DMI discussed recently [41, 42].

1. DMI-like interactions: $z$-component

The $z$-component of the four-spin chiral interaction $\Delta D_{ijkl}$, when all site indices $i, j, k, l$ may be different, is represented by the antisymmetric part of the exchange tensor characterizing the interaction between sites $i, j, k$ and $j$. In full analogy to the DMI, $D^{z}_{ijkl}$, can be written as follows
\[ D^{z}_{ijkl} = \frac{1}{4}(J^{xyxy}_{ijkl} + J^{yyxy}_{ijkl} - J^{yxxy}_{ijkl} - J^{xyyx}_{ijkl}) \] (13)
with the tensor elements $J^{\alpha\beta\gamma\delta}_{ijkl}$ determined via Eq. (7).

In the following, we will focus on the three-spin DMI-like interactions TDMI (implying $l = j$) and biquadratic vector interactions (with $l = j, k = i$), which were calculated and discussed recently for some systems with special geometry [21, 22] in comparison with the DMI. Using Eq. (13) for the special case $l = j, k = i$ one has for the $z$ component of the biquadratic interaction the expression:
\[ D^{z}_{ijij} = \frac{1}{4}(J^{xyxy}_{ijij} + J^{yyxy}_{ijij} - J^{yxxy}_{ijij} - J^{xyyx}_{ijij}). \] (14)

2. DMI-like interactions: $x$- and $y$-component

To calculate the $x$ and $y$ components of the four-spin and as a special case the TDMI and BDMI terms in a system magnetized along the $z$ direction, we follow the scheme suggested by the authors for the calculation of the DMI parameters [41, 42], which exploited the DMI-governed behaviour of the spin-wave dispersion having a finite slope at the $\Gamma$ point of the Brillouin zone. However, in the present case a more general form of perturbation is required. For this purpose we assume a 2D spin modulation according to the expression
\[ \hat{m}_i = (\sin(\hat{q}_1 \cdot \hat{R}_i) \cos(\hat{q}_2 \cdot \hat{R}_i), \sin(\hat{q}_2 \cdot \hat{R}_i), \cos(\hat{q}_1 \cdot \hat{R}_i)\cos(\hat{q}_2 \cdot \hat{R}_i)), \] (15)
which is characterized by two wave vectors, $\hat{q}_1$ and $\hat{q}_2$, orthogonal to each other, as for example $\hat{q}_1 = q_1 \hat{y}$ and $\hat{q}_2 = q_2 \hat{x}$. The microscopic expression for the $x$ and $y$ components of $\Delta D_{ijkl}$ describing the most general, four-spin interaction, can be obtained on the basis of the third-order term of the Green function expansion in Eq. (2) leading to a corresponding third-order energy correction
\[ \Delta \mathcal{E}^{(3)} = -\frac{1}{\pi} \operatorname{Im} \operatorname{Tr} \int_{E_F} dE (E - E_F) \]
\[ \times G_0 \Delta V G_0 \Delta V G_0 \] (16)
by taking its derivatives in the limit $q_1, q_2 \to 0$; to first-order with respect to the wave-vector $\hat{q}_1$ and to second-order with respect to the wave-vectors $\hat{q}_1$ and $\hat{q}_2$. The non-zero elements of the first-order derivative in the limit $q_1 \to 0$ imply an antisymmetric character of the interactions between the magnetic moments on sites $i$ and $j$ in Eq. (12), similar to the case of the conventional DMI. At the same time, the non-zero second-order derivative with respect to $q_1$ and to $q_2$ correspond to a scalar interaction between the magnetic moments on sites $k$ and $l$, which is symmetric with respect to a sign change of the wave vector. The same properties should apply to the corresponding contribution to the model spin Hamiltonian in Eq. (12). Equating for the ab-initio and model approaches the corresponding terms proportional to $(\hat{R}_i - \hat{R}_j)(\hat{R}_k - \hat{R}_l)$ and $(\hat{R}_i - \hat{R}_j)(\hat{R}_k - \hat{R}_l)$, we keep a similar form in both cases for the sake of convenience gives access to the elements $D_{ijkl}^x$ and $D_{ijkl}^y$ of the four-spin chiral interaction. As we focus here on TDMI and BDMI, they can be obtained as the special cases $l = j$ and $l = j, k = i$, respectively. With this, the elements of the TDMI vector can be written as follows
\[ D^{x,\alpha}_{ijkl} = -\epsilon_{\alpha\gamma} \frac{1}{8\pi} \operatorname{Im} \operatorname{Tr} \int_{E_F} dE (E - E_F) \]
\[ \left[ Q^{ij} T^{\alpha\gamma} T^{jk} T^{\alpha\beta} T^{ij} - T^{i\alpha} T^{j\gamma} T^{jk} T^{i\alpha} T^{ij} \right] \] (17)
with $\alpha, \beta = x, y$, and $\epsilon_{\alpha\gamma}$ the elements of the transverse Levi-Civita tensor $\epsilon = \begin{bmatrix} 0 & 1 \\ -1 & 0 \end{bmatrix}$. The matrix elements of
the torque operator $T^{\text{DM}}_{ij}$ occurring in Eq. (17) are defined in an analogous way [45]:

$$O^{i}_{\Lambda\Lambda'} = \frac{1}{8\pi} \Im \text{Tr} \int \frac{d^{3}r}{V} Z^{r}_{\Lambda}(\vec{r},E) Z^{i}_{\Lambda'}(\vec{r},E).$$

The expression in Eq. (17) gives access to the $x$ and $y$ components of the DMI-like three-spin interactions in Eq. (12)

$$D^{\alpha}_{ijk} = D^{\alpha,x}_{ijk} + D^{\alpha,y}_{ijk}.$$  

An expression for the BDMI also follows directly from Eq. (17) using the restriction $k = i$. This leads to the elements $D^{\alpha,\beta}_{ijk}$ determining chiral biquadratic exchange interactions (similar to the case of four-spin interactions), which can be written in the following form

$$D^{\alpha,\beta}_{ijk} = -\epsilon_{\alpha\gamma} \frac{1}{8\pi} \Im \text{Tr} \int \frac{d^{3}r}{V} E(E - E_{F}) [\left(O^{i} z^{j}z^{k} \tau^{i}z^{j}z^{k} + T^{i}z^{j}z^{k} \tau^{i}z^{j}z^{k} \tau^{i} \tau^{j} \tau^{k} - T^{i}z^{j}z^{k} \tau^{i}z^{j}z^{k} \tau^{i} \tau^{j} \tau^{k} \tau^{i} \tau^{j} \tau^{k}ight) + \left(O^{i} z^{j}z^{k} \tau^{i}z^{j}z^{k} \tau^{i} \tau^{j} \tau^{k} - T^{i}z^{j}z^{k} \tau^{i}z^{j}z^{k} \tau^{i} \tau^{j} \tau^{k} \tau^{i} \tau^{j} \tau^{k}ight)] .$$

C. Chiral exchange: three-spin exchange interactions

Here we discuss the three-spin chiral exchange interaction entering a corresponding extension term to the Heisenberg Hamiltonian

$$H^{(3)} = -\sum_{i\neq j \neq k}^{N} J_{ijk} \hat{s}_{i} \cdot (\hat{s}_{j} \times \hat{s}_{k}).$$

As it follows from this expression, the contribution due to the three-spin interaction is non-zero only in case of a non-co-planar and non-co-linear magnetic structure characterized by the spin chirality $\hat{s}_{i} \cdot (\hat{s}_{j} \times \hat{s}_{k})$ involving the spin moment on three different lattice sites.

Considering the torque acting in a FM system on the magnetic moment of any atom $i$, which is associated with the three-spin interactions, one can evaluate its projection onto an arbitrary direction $\hat{u}$, $T^{(3)}_{ijk,\hat{u}} = -(\partial H^{(3)}/\partial \hat{s}_{i}) \cdot (\hat{u} \times \hat{s}_{i})$, which is equal to $J_{ijk}(\hat{e}_{i} \cdot \hat{u})$. This value is non-zero only in the case of a non-zero scalar product $\hat{s}_{i} \cdot \hat{u}$, implying that a nonvanishing torque on spin $\hat{s}_{i}$ created by the spin $\hat{s}_{j}$ coupled via the three-spin interaction, requires a tilting of the third spin moment $\hat{e}_{k}$ to have a non-zero projection on the torque direction. In contrast to that, the torque $T^{\text{DM}}_{ijk} = \hat{D}_{ijk} \cdot \hat{u}$ [46] acting due to the spin of atom $j$ on the spin moment of atom $i$ via the DMI, is non-vanishing even in the system with all spin moments being collinear. This makes clear that in order to work out the expression for the $J_{ijk}$ interaction term, a more complicated multi-Q modulation [14, 34, 36] of the magnetic structure is required when compared to a helimagnetic structure characterized by a wave vector $\vec{q}$, which was used to derive expressions for the $x$- and $y$-components of the DMI [41, 42].

Accordingly, we use here the 2D non-collinear spin texture described by Eq. (15), which is characterized by two different wave vectors in two perpendicular directions, $\vec{q}_{1} = (0, q_{y}, 0)$ and $\vec{q}_{2} = (q_{x}, 0, 0)$. In this case the spin chirality driven by the three-spin interaction should lead to the asymmetry of the energy $E(\vec{q}_{1}, \vec{q}_{2})$ with respect to a sign change of any of the vectors $\vec{q}_{1}$ and $\vec{q}_{2}$, as a consequence of full antisymmetry of the scalar spin chirality. As a result, the three-spin interactions can be derived assuming a non-zero slope of the energy dispersion $E(\vec{q}_{1}, \vec{q}_{2})$ as function of the two wave vectors, in the limit $\vec{q}_{1}(2) = 0$.

Substituting the spin modulation in Eq. (15) into the spin Hamiltonian in Eq. (21) associated with the three-spin interaction, the second-order derivative of the energy $E^{(3)}(\vec{q}_{1}, \vec{q}_{2})$ with respect to $q_{1}$ and $q_{2}$ wave vectors in the limit $q_{1} \to 0$, $q_{2} \to 0$ is given by the expression

$$\frac{\partial^{2} E^{(3)}}{\partial \vec{q}_{1} \cdot \partial \vec{q}_{2}} = -\sum_{i \neq j \neq k} J_{ijk} \left(\hat{z} \cdot ((\vec{R}_{i} - \vec{R}_{j}) \times (\vec{R}_{k} - \vec{R}_{j})) \right).$$

The microscopic energy term of the electron system, giving access to the chiral three-spin interaction in the spin Hamiltonian is determined by the second-order term of the free energy expansion given by the expression

$$\Delta E^{(2)} = -\sum_{i \neq j \neq k} J_{ijk} \left(\hat{z} \cdot ((\vec{R}_{i} - \vec{R}_{j}) \times (\vec{R}_{k} - \vec{R}_{j})) \right) \frac{1}{8\pi} \Im \text{Tr} \int \frac{d^{3}r}{V} E(E - E_{F}) G_{0} \Delta VG_{0} \Delta VG_{0} .$$

To make a connection between the two approaches associated with the ab-initio and model spin Hamiltonians, we consider a second-order term with respect to the perturbation $\Delta V$ induced by the spin modulation in Eq. (15). 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

$$J_{ijk} = -\frac{1}{8\pi} \Im \text{Tr} \int \frac{d^{3}r}{V} \left[ T^{i,x} z^{j,y} z^{k} + T^{i,y} z^{j,x} z^{k} + T^{i,z} z^{j,x} z^{k} + T^{i,z} z^{j,y} z^{k} + T^{i,z} z^{j,k} z^{x} + T^{i,z} z^{j,k} z^{y} + T^{i,z} z^{j,k} z^{z} + T^{i,z} z^{j,k} z^{z} + T^{i,z} z^{j,k} z^{z} + T^{i,z} z^{j,k} z^{z} \right].$$
FIG. 1. The four-spin interaction exchange parameters $J_{ijkl}$ according to Eq. (10) calculated for the FM hcp Co (a), bcc Fe (b) and fcc Ni (c) with the magnetization along the $\hat{z}$-axis.

IV. NUMERICAL RESULTS

In order to illustrate the expressions developed above by their application to realistic systems, corresponding calculations on various representative systems have been performed.

A. Four-spin and biquadratic exchange interactions

Figure 1 represents an example for the four-spin exchange parameters $J_{ijkl}^s$ calculated on the basis of Eq. (10) for the three 3d bulk ferromagnetic systems bcc Fe, hcp Co and fcc Ni. The results are plotted as a function of the distance $R_{ij} + R_{jk} + R_{kl} + R_{il}$, including only the interactions corresponding to $i \neq j \neq k \neq l$, i.e., all sites are different. For these systems the exchange parameters are about two orders of magnitude smaller than the first-neighbor bilinear exchange interactions. However, in general their contribution can be non-negligible due to the large number of such four-spin loops. Therefore, in some particular cases they should be taken into account.

Examples for the scalar biquadratic exchange interaction parameters $J_{ijij}^s$ are shown in Fig. 2 for bcc Fe, hcp Co and fcc Ni, and in Fig. 3 for the compounds FePt and FePd having CuAu crystal structure. For comparison, the insets give the corresponding bilinear isotropic exchange interactions. One can see rather strong first-neighbor interactions in bcc Fe and in the compounds FePt and FePd. This confirms the previous theoretical results for bcc Fe [15], and demonstrates the non-negligible character of biquadratic interactions. This is of course a material-specific property.

B. DMI-like multispin exchange interactions

The properties of the chiral multispin exchange interaction parameters in Eq. (12) can be compared with the DM interactions as both are vector quantities. Similarly to the DMI, these parameters are caused by SOC, i.e., they vanish in the case of SOC = 0. This feature is indeed demonstrated by our test calculations. The calculations have been performed for bulk bcc Fe, for (Pt/X/Cu)$_n$ multilayers with $X = \text{Mn, Fe and Co}$, and for an Fe overlayer deposited on TMDC (transition metal dichalcogenide) monolayers, e.g., 1H-TaTe$_2$, 1H-WTe$_2$, and 1H-WS$_2$. The model multilayer system is composed of Pt, $X$ and Cu on subsequent (111) layers of the fcc lattice, without structural relaxation. In the case of the Fe/TMDC systems the structural relaxation has been performed both within the layers as well as in the $z$ direction perpendicular to the layer plane.

The calculations demonstrate similar symmetry properties of the BDMI when compared with the conventional DMI, as was already pointed out recently [21]. In bcc Fe having inversion symmetry, the BDMI is equal to zero, while it is finite in the multilayer and the Fe/TMDC systems, following the properties of the DMI interactions. Figure 4 gives results for the $z$-component of the chiral biquadratic exchange interactions, $D_{ijij}^z$, calculated for a Fe overlayer deposited on a TaTe$_2$ and WTe$_2$ single layers, respectively, on the basis of Eq. (19). As one can see, $D_{ijij}^z$ has a significant magnitude when compared to the bilinear DMI parameters. Interestingly, the $x$ and $y$ components in these two materials are much smaller than the corresponding components of the bilinear DMI.

In the case of the multilayer systems (Pt/Fe/Cu)$_n$, (Pt/Mn/Cu)$_n$ and (Pt/Co/Cu)$_n$, all three components, $x, y, z$, have the same order of magnitude as it is seen in Fig. 5. The orientation of these interactions between first nearest neighbour sites is shown in Fig. 6. As can be seen from Table I, in contrast to the Fe/TMDC system, all components are more than one order of magnitude smaller than the corresponding DMI components.

Figure 6 shows schematically the in-plane components of the DMI and BDMI, which have the same orientation for (Pt/Fe/Cu)$_n$ and (Pt/Mn/Cu)$_n$, but not for (Pt/Co/Cu)$_n$. The $y$-component of $D_{ijij}^z$ representing the interaction between atoms with $\vec{R}_{ij} = a(0.707, 0, 0)$ are given in Table I. These values give the total in-plane interaction as for the taken pair of atoms $D_{ij}^x = 0$ and $D_{ij}^y = 0$. Note also that in (Pt/Mn/Cu)$_n$ the $D_{ijij}^z$ com-
FIG. 2. Scalar biquadratic exchange interactions in bcc Fe (a), hcp Co (b) and fcc Ni (Ni). The insets show the bilinear exchange interaction parameters calculated for the FM state with the magnetization along the $\hat{z}$-axis.

ponent has an opposite sign when compared to $D_{ij}^z$.

Similar to the DMI and BDMI, the TDMI $D_{ijkj}$ is a SOC-induced interaction between atoms $i$ and $j$ which depends on the relative orientation of the spin moment of the atoms $j$ and $k$. In contrast to the biquadratic interaction, it does not vanish for centrosymmetric systems, as it is demonstrated by the calculations for bcc Fe represented in Fig. 7. Let us consider the TDMI as the DMI-like interaction between atoms $i$ and $j$ which depends on the relative orientation of the spin moment of the atoms $j$ and $k$. One finds obviously a different sign for the various interactions for the same value of $|\vec{R}_{ij}|$ in bcc Fe as a function of the position of the third atom $k$. For this geometry $D_{ijkj}^x$ and $D_{ijkj}^y$ represent the magnitude of the in-plane projection of corresponding interactions with the first nearest neighbors, as in this case $D_{1212}^x = 0$ and $D_{12}^x = 0$.

![Graph](image)

**TABLE I.** The $y$- and $z$-components of the DMI and chiral biquadratic exchange interaction (in meV) between 3d-metals in $(Pt/X/Cu)_n$ multilayers ($X = Mn, Fe, Co$). The $y$-component corresponds to the interactions between atoms 1 and 2 (see Fig. 6) with $R_{12} = a(0.707, 0, 0)$. For this geometry $D_{1212}^r$ and $D_{12}^r$ represent the magnitude of the in-plane projection of corresponding interactions with the first nearest neighbors, as in this case $D_{1212}^r = 0$ and $D_{12}^r = 0$.

| Material  | $D_{ij}^y$ | $D_{ij}^x$ | $D_{ij1}^y$ | $D_{ij1}^x$ |
|-----------|------------|------------|-------------|------------|
| (Pt/Mn/Cu)$_n$ | -1.14 | -1.22 | -0.039 | 0.031 |
| (Pt/Fe/Cu)$_n$ | 0.17 | 0.35 | 0.024 | 0.034 |
| (Pt/Co/Cu)$_n$ | 0.63 | 0.40 | -0.003 | 0.008 |

![Graph](image)

**FIG. 3.** Scalar biquadratic Fe-Fe exchange interactions in the FM-ordered FePt and FePd with the magnetization along the $\hat{z}$-axis.
the direction of this interaction depends on the position of atom 3.

However, in the case of systems without inversion symmetry, the TDM interactions do not cancel each other and can play a certain role in the formation of the magnetic ground state configuration. This is demonstrated by calculations for (Pt/X/Cu)$_n$ multilayer systems. Fig. 8 shows corresponding results for the (Pt/Mn/Cu)$_n$ multilayer system where, using a similar representation as before, the arrows represent the 'vector' interactions (i.e. $\sim (\hat{s}_1 \times \hat{s}_2)$) between atoms 1 and 2, controlled by the third atom 3. Obviously, the direction of this interaction depends on the position of the third atom as one can see in Fig. 8 (a) and (b). Moreover, the magnitude of this interaction follow the 3-fold in-plane symmetry of the system, and is comparable to that of the biquadratic interactions and is smaller by more than one order of magnitude when compared to the DMI interactions.

FIG. 4. $z$-component of the BDMI (squares, top) and biquadratic scalar interactions (squares, bottom) as a function of the site distance $R_{ij}$ (in multiples of the lattice parameter $a$). Results are shown for a Fe overlayer deposited on a single layer of TaTe$_2$ (a) and WTe$_2$ (b), in comparison with DMI and bilinear interaction parameters shown by circles.

FIG. 5. The $x$-, $y$- and $z$-components of chiral biquadratic exchange interaction, $\vec{D}_{ijij}$, between the magnetic 3d-metals $X$ in (Pt/$X$/Cu)$_n$ multilayers with $X = $ Mn (a), Fe (b), and Co (c), plotted as a function of the interatomic distance $R_{ij}$. The orientation of the interaction vectors between first nearest neighbours is shown in Fig. 6

FIG. 6. Orientation of the DMI vector $\vec{D}_{ij}$ (a) and the vector of the BDMI $\vec{D}_{ijij}$ (b) corresponding to the interaction of the first neighbor 3d-atoms $X$ in (Pt/$X$/Cu)$_n$ multilayers: for $X = $ Mn (left), Fe (middle), and Co (right). The magnitude of the interactions are given in Table I.
C. Chiral exchange: Three-spin exchange interactions

Eq. (24) was used to calculate the three-spin interaction parameters for a couple of representative 3D and 2D systems. The results of such calculations for bulk bcc Fe are shown in Fig. 9. First of all, one can see that the three-spin interactions are finite despite they have been obtained for SOC=0, while taking the SOC into account leads to a very tiny modification of these interactions. This behaviour is in strong contrast to that of the SOC-driven DMI that requires the lack of inversion symmetry in the system. As soon as in the absence of SOC the system is invariant with respect to the rotation of the magnetization, no anisotropy of the three-spin interaction should be observed. This can clearly be seen in Fig. 9 representing the results for three different orientations of the magnetic moment: [001] (circles), [111] (squares) and [110] (diamonds).

Fig. 10 (a) shows the three-spin chiral interaction between 3$d$ atoms in the (Pt/Mn/Cu)$_n$ multilayer system, calculated for SOC = 0. One can see that the three-spin interactions are non-zero and undergo only weak modification when the SOC is taken into account; similar to the case of bcc Fe, corroborating that the SOC does not play a critical role for the three-spin interactions. The three-spin interactions between first-neighbors in (Pt/Mn/Cu)$_n$ (squares) and (Pt/Fe/Cu)$_n$ (circles) have opposite sign with respect to each other. In the case of (Pt/Co/Cu)$_n$ (diamonds) the sign of the first-neighbor coupling parameters $J_{ijk}$ is different for two different triangles depending whether it is Pt- or Cu centered (see Fig. 11).

Comparing these interactions with the conventional DMI shown in Table I, one can see that both values have the same order of magnitude, and both are non-negligible when compared to the isotropic interactions shown in Fig. 10 (b). Note once more that the DMI is caused by conventional SOC (i.e., of relativistic origin) in systems with chiral crystal structure. In contrast to the DMI, the three-spin interactions do not vanish in the case of vanishing SOC, as is shown above. This feature
acting on each spin moment in the atomic triangle with one can talk for such a system about an emergent torque operator) having topological origin [47, 48]. As a result, one can see in Fig. 12(a) the formation of the anticrossing gaps caused by spin modulation, which are modified due to the changing helicity of the spin modulation along (−0.5, 1/√2) direction (Fig. 12(b)), that can be an indication of their chiral behaviour. In full analogy to the DMI this can be connected to a chiral interaction between two spin moments, associated with the SOC having topological origin. Fig. 13 shows the energy dependent TCI demonstrating an increase of its magnitude above and below the Fermi energy where the anticrossing points are located.

Figs. 14 and 15 represent the CTI (a) in comparison with BDMI (b) DMI (c) and isotropic exchange interactions, plotted as a function of energy characterizing the occupation of the valence band (i.e. an artificial Fermi energy position). One can see an oscillating behaviour for all parameters when the occupation increases, with spin-orbit interaction.

Obviously, the chirality properties associated with the TCI should be seen also in the electronic band structure, although they cannot be easily separated from other effects. Nevertheless these properties can also be compared with those responsible for DMI. In the latter case, the chiral properties of the exchange interactions are connected to the asymmetric changes of the SOC-induced anticrossing gap when spin-spirals with different helicity are created (see, e.g. [49]). A similar feature can also be expected in the absence of conventional SOC. When the anticrossing splittings occur due to a non-collinear spin structure, e.g., spin spirals in different directions, they can also exhibit the asymmetry with respect to helicity of one of spin spirals. This can be demonstrated for example for the model 1ML Fe(110) system. Creating a 3 × 3 supercell, and tilting the magnetic moments by 5° away from z-direction one can construct a spin structure with a modulation in two different directions (−0.5, 1/√2) and (0.5, 1/√2) in the (x, y)-plane. The Bloch spectral function in Fig. 12(a) and (b), plotted for k varying along the line π/√3(−1/3, 0, 0) − Γ − π/√3(1/3, 0, 0), represent the electronic states for the systems with opposite helicity along the (−0.5, 1/√2) direction. First of all one can see in Fig. 12(a) the formation of the anticrossing gaps caused by spin modulation, which are modified due to the changing helicity of the spin modulation along (−0.5, 1/√2) direction (Fig. 12(b)), that can be an indication of their chiral behaviour. In full analogy to the DMI this can be connected to a chiral interaction between two spin moments, associated with the SOC having topological origin. Fig. 13 shows the energy dependent TCI demonstrating an increase of its magnitude above and below the Fermi energy where the anticrossing points are located.

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TABLE II. Three-spin interaction parameters (in meV) between the Fe atoms forming a smallest triangle, calculated for Fe(1ML)/TMDC(1ML) and (Pt/X/Cu)n multilayer systems (X = Mn, Fe, Co). The columns A and B give results for S-centered (A) and hollow-centered (B) triangles in the former case and to Pt-centered (A) and Cu-centered (B) triangles in the latter case.
FIG. 12. The majority-spin Bloch spectral function (BSF) along the line $\frac{2\pi}{a}(-\frac{1}{3}, 0, 0) - \Gamma - \frac{2\pi}{a}(\frac{1}{3}, 0, 0)$ for 1 ML of Fe(110) with a $3 \times 3$ supercell for the spin modulation along two directions $(-0.5, 1/\sqrt{2})$ and $(0.5, 1/\sqrt{2})$. Top and bottom figures show the BSF for the opposite spin helicity along direction $(-0.5, 1/\sqrt{2})$.

FIG. 13. The three-spin interactions $J_{ijk}(E)$ in 1 ML of Fe(110) as a function of occupation. Their sign changing at different energies because of different origin of these interactions. Note, however, that all quantities shown in Figs. 14 and 15 have a maximum at approximately half occupation of the Mn(Co) $d$-band (see Figs. 14(e) and 15(e)), that correlates also with the maximum of the spin magnetic moment (Figs. 14(a) and 15(a)) and maximum of antiferromagnetic exchange interactions (Figs. 14(d) and 15(d)).

Comparing the $y$- and $z$-components of the BDMI and DMI shown in Figs. 14 (a) and (b), respectively, one can see a more narrow energy region, in which the former quantity has a significant magnitude. Note, however, that the biquadratic interaction is a higher order term in the energy expansion and should represent simultaneously the features of vector and scalar interactions of two spin moments. Thus, plotting in Fig. 14 (a) (thin lines) the function $D_{ij}^y(E)/\max(J_{ij}(E))$ for the nearest-neighbor interactions, one can see a localization in energy of this function similar to the one seen for the BDMI.

FIG. 14. Three-spin interaction parameters $J_{ijk}(E)$ in (Pt/Mn/Cu)$_n$ multilayer for the Mn-triangles centered at Cu ($J_{Cuijk}$) and Pt ($J_{Ptijk}$); the dotted line represents the Mn spin magnetic moment $m(E)$ (in $\mu_B$) as a function of the occupation (a); $y$, $z$-components of the chiral biquadratic exchange interaction, $D_{1212}$ (b) and DM interactions, $D_{ij}$ (c) between Mn atoms as a function of the occupation; (d) the isotropic exchange $J_{ij}(E)$, and (e) the element-projected DOS(E).
were aimed only to demonstrate the possible impact of the three-spin interaction on magnetic texture, therefore they are performed for a rather small system size, which implies that the characteristic size of the stabilized magnetic texture should also be small. Therefore the DMI and three-spin interaction parameters used in the MC simulations are chosen unrealistically large.

The simulations were performed for a monolayer system having a triangular crystal structure. The atoms couple ferromagnetically with the nearest neighbor parameters \( J_1 (J_1 = 3 \, \text{meV}) \). When no other interatomic exchange interactions are taken into account, the system is ferromagnetic in its ground state. Switching on negative second-neighbor interactions \( J_2 \), DMI \( \vec{D} \) and TCI \( J^{(3)} \) can change the magnetic texture in the system drastically. First, we consider the effects of the nearest neighbor DMI \( \vec{D} \) and three-spin interactions, while \( J_2 = 0 \, \text{meV} \), shown in Fig. 16. The DMI in the figure changes with the ratios \( D/J_1 = 0.3, 0.6 \) and 1.0 along a row. The three-spin interactions, on the other hand, change along a column with \( J^{(3)}/J_1 = 0.0, 0.4 \) and 0.5. One can see that the DMI stabilizes spin-spiral magnetic structures competing only with the FM isotropic exchange interactions \( J_1 \). However, the vortex lattice appears when the TCI is taken into account. When DMI are rather small, single vortexes can also be stabilized (Fig. 16, first column), while no magnetic texture was observed for \( D/J_1 = 0.0 \) (not shown in the figure).

The negative second-neighbor interaction term \( J_2 \) can also stabilize non-collinear magnetic structure competing with positive nearest-neighbor interaction. Therefore, we consider also the systems with \( |J_2|/J_1 = 0.5 \) (see Fig. 17). The first row corresponds to \( J^{(3)} = 0 \), while the second and third ones to \( J^{(3)}/J_1 = 0.4 \) and 0.5 respectively. The DMI changing along the rows takes the values \( D/J_1 = 0.0, 0.3 \), and 1.0. As one can see (first row) the spin spiral state is stabilized by accounting for a negative \( J_2 \) and DMI. On the other hand switching on in addition the TCI leads to the formation of a vortex texture that is modified upon a change of the exchange parameters TCI and DMI.

Obviously, it would be of great interest to see an impact of an external magnetic field, magnetic anisotropy and others multispin interactions. This however needs separate investigations beyond the main issue of the present work.

D. Monte Carlo simulations

In order to demonstrate the possible impact of the higher-order chiral interactions on the magnetic structure, Monte Carlo simulations have been performed for various model systems. In this section we focus only on the effect of the three-spin chiral interactions having different properties when compared to other chiral interactions discussed above and having relativistic origin (i.e. DMI-like interactions). Note also that the calculations

V. SUMMARY

To summarize, in the present work we present a general approach to calculate the multispin exchange interactions in order to extend the classical Heisenberg Hamiltonian. This approach allows first principles calculations of multispin interactions in real-space within the framework of the multiple scattering Green function formalism. We discussed some properties of different types of chiral interactions, with the main focus on the three-spin ex-
change interactions. A specific feature of TCI is its topological origin in contrast to the DMI. We demonstrated by means of MC simulations that this term can lead to a stabilization of vortex- or skyrmionic-like magnetic texture in centrosymmetric materials, in the absence of an external magnetic field.

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