Comment on "Proper and improper chiral magnetic interactions"

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In a recent paper by dos Santos Dias *et al.* [Phys. Rev. B **103**, L140408 (2021)], a critique of earlier works analyzing low-energy spin Hamiltonians is put forth. To be precise, it is the large noncollinear contributions to the Dzyaloshinskii-Moriya interaction (DMI) that is the main concern of dos Santos Dias *et al.* In this Comment, we clarify the microscopic mechanisms for the large DMI that can be found in noncollinear magnets. Furthermore, we outline the complementary nature of the different parametrizations of a spin Hamiltonian, with strengths and weaknesses of both approaches. Specifically, we stress the physical insight in the interpretation of the DMI, when decomposed in microscopic electron and spin densities and currents.

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A short background on spin Hamiltonians. There are several approaches to mapping results from electronic structure calculations to effective low-energy spin Hamiltonians. In doing this, it is possible to calculate from first-principles theory the exchange parameters, or alternatively the local Weiss field, that enter an effective atomistic spin Hamiltonian, enabling practical calculations of magnetic excitations, which may extend over hundreds of thousands of atomic spins. Approaches that have been used involve expressions of exchange parameters calculated from a fundamental electronic Hamiltonian [1–3], and the mapping of a spin Hamiltonian onto total energy calculations based on density functional theory (DFT) for spin spirals [4,5]. In addition, spin-cluster expansions have been explored [6–10], as has the method of considering energy variations of the magnetic ground state within first-principle approaches such as spin-polarized versions of DFT. The latter one is often referred to as the Liechtenstein-Katsnelson-Antropov-Gubanov (LKAG) approach, and was originally considered for collinear (ferromagnetic) ground states [11,12]. Extensions of the LKAG formalism to nonequilibrium [13–16], noncollinear [17–23], and finite temperature [24–26] states have also been considered. One should note that a mapping of noncollinear spin configurations to a Heisenberg model [21] or to a generalized Heisenberg model with a bilinear exchange tensor [13,19] is in general complicated, apparently requiring the inclusion of more complex exchange contributions [3,10,22,23,27]. It is also important to note that the spin-configuration dependence of the parameters is not exclusively a noncollinear issue. In fact, the calculated exchange parameters in general depend on the underlying magnetic configuration [28–30]. A simple example of this is a diatomic molecule with a ferromagnetic ground state, where a calculation of the exchange parameters of the ground state configuration and the antiferromagnetic configuration is expected to yield exchange parameters of opposite sign, but with equal strength. In practice the sign change of two such opposing configurations is found, but the strength of the interaction is seldom the same.

The LKAG approach of calculating Heisenberg exchange interactions from electronic structure theory was generalized to the relativistic case in which the spin-orbit interaction is considered for the electronic Hamiltonian [31–35]. This led to the possibility to evaluate anisotropic symmetric and antisymmetric exchange terms of an effective atomistic spin Hamiltonian. Here, the antisymmetric part represents the Dzyaloshinskii-Moriya (DM) interaction, $\mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j)$, in which $\mathbf{S_i}$ represents an atomic spin on site i. In two recent works [22,23], it was argued that for noncollinear magnetic configurations, electronic structure results can be mapped to an effective spin Hamiltonian with a significant DM interaction, the size of which can be much larger than the DM interaction from relativistic collinear configurations, even in the absence of a spin-orbit interaction. In Refs. [22,23] an exchange interaction that has the form $\mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j)$ and is nonrelativistic in origin, and instead is caused by an underlying noncollinear magnetic configuration, was referred to as a non-collinear (or nonrelativstic) DM (or DM-like) interaction. A critique of the analysis of Refs. [22,23,36] was recently put forth by the authors of Ref. [27], in which it was proposed that a more appropriate form of a spin Hamiltonian for noncollinear situations should contain a generalization of a four-spin interaction term, in the form of a biquadratic

interaction. It is the purpose of this Comment to Ref. [27], to clarify the microscopic mechanism of the noncollinear, nonrelativistic DM interaction suggested in Refs. [22,23], and to analyze the advantages and disadvantages of mapping electronic structure information to such a Hamiltonian, compared to, e.g., the model proposed in Ref. [27].

A microscopic analysis of noncollinear DM interactions. The most transparent demonstration of how noncollinear magnetic configurations have an electronic Hamiltonian that is similar in form to the inclusion of spin-orbit coupling (SOC), can be seen by starting from a general form of the Kohn-Sham equation for the electron states,

$$H_{\rm el} = [-\nabla^2 + v(\mathbf{r})]\sigma^0 + [\mathbf{b}(\mathbf{r}) + \xi\mathbf{l}]\cdot\boldsymbol{\sigma}, \qquad (1)$$

where v is the total nonmagnetic part of the effective potential, $\mathbf{b}$ is the magnetic part of the effective potential, ξ is the spin-orbit interaction strength, and $\mathbf{l}$ is the angular momentum operator, while $\boldsymbol{\sigma}$ and σ^0 are the vector (x, y, and z components) of Pauli matrices and the 2×2 identity, respectively. This form of the Kohn-Sham equation was proposed in Ref. [37] in discussing intra-atomic noncollinear magnetization densities, which for both collinear and noncollinear spin configurations may be a significant contribution to the effective potential the electrons move in. In practice this equation treats the noncollinear components of the exchange and correlation potential $\mathbf{b}$ and orbit interaction $\xi\mathbf{l}$ in a similar way by rewriting terms such as $\xi(l_x\sigma^x + l_y\sigma^y)$ as $\xi(l_+\sigma^- + l_-\sigma^+)/2$ and $b_x\sigma^x + b_y\sigma^y$ as $(b_+\sigma^- + b_-\sigma^+)/2$. Both terms thus enter the electronic Hamiltonian (typically considered in matrix form, when any basis-set expansion of electron states is considered) in a very similar way, in particular having a similar symmetry of the off-diagonal components of spin space. It is this similarity in the electronic Hamiltonian that governs that both relativity and noncollinearity result in quantum mechanical spin and charge currents that produce the significant values of the Green's functions G^{00}, G^{01}, $G^{\mu 0}$, $G^{\mu 1}$, defined and analyzed in Eqs. (4)–(9) in Refs. [22,23]. The subscript μ relates to a Cartesian component: x, y, or z. These Green's functions, in particular the spin- and charge-current terms, enter the expression for the DM interaction term [22,23]. The division of the DM interaction as coming from different charge and spin currents, naturally allows for a deepened understanding of the origin of DM-like interactions of a material. It is not clear if, or how, this type of analysis is possible in the model suggested in Ref. [27]. Based on this analysis, the $\mathbf{b}$ term of Eq. (1) is expected to result in physical phenomena typically associated with a spin-orbit interaction, the DM-like interaction discussed in Refs. [22,23], but also, e.g., the damping parameter of the Landau-Lifshitz-Gilbert equation [38], a significant spin-Hall effect, as well as Kerr and Faraday rotation.

In terms of the relative importance of the two contributions discussed above for the DM interaction, we note that the strength of the spin-orbit interaction of an element seldom varies significantly from compound to compound, and remains essentially an atomic, local property. For a $3d$ transition metal it adopts values in the range 30–100 meV. In contrast, the noncollinear contributions $\mathbf{b}$ to Eq. (1) can be an order of magnitude larger, which is consistent with DM-like interaction strengths that can be significantly larger than the spin-orbit-induced DM term. Another significant difference is that the noncollinear DM term does not favor a specific chirality, as shown in the result sections of Refs. [22,23].

Comparing parametrizations. In comparing the two types of parametrizations proposed in Refs. [22,23] and in Ref. [27], we note that the former suggests a form of $\mathbf{D}_{ij}\cdot(\mathbf{S}_i\times\mathbf{S}_j)$, while the latter advocates for a term $B_{ij}(\mathbf{S}_i\cdot\mathbf{S}_j)^2$. We proceed below with a comparison of these approaches, discussing advantages and disadvantages with both. The approach of Ref. [27] also constitutes in a fitting of the parameters of a bilinear/biquadratic spin model to total energies of the system (although the approach of using this spin Hamiltonian can be done from an LKAG-type approach). In Refs. [22,23] the parameters of the spin model were obtained from the electronic Hamiltonian (or one-electron Green's function) using, e.g., the LKAG formula and its extensions.

It is important to realize that the different parametrizations and methods used in Refs. [22,23] and in Ref. [27] complement each other and contain their own benefits and drawbacks. Reference [27] provides a description of the global magnetic structure in terms of bilinear and biquadratic spin-spin interactions which enables convenient analyses of, for instance, the system's magnetic ground state. However, a drawback with this approach is the absence of a unique mapping onto the multispin model. The restricted biquadratic form suggested in Ref. [27] gives a good fitting only when the variation of the magnetic structure is limited to the simple and symmetric variation of three independent moments confined to a coplanar structure, used in both Refs. [23,27]. For instance, if we extend this to a larger set of independent moments, say six sites, and allow for true noncollinearity, the generalized version of a four-spin interaction $B_{ijkl}(\mathbf{S}_i\cdot\mathbf{S}_j)(\mathbf{S}_k\cdot\mathbf{S}_l)$ as well as six-spin interactions come into play. The approach used in Refs. [22,23] allows for direct calculations of the magnetic interactions and, more importantly, points out the role of charge and spin densities as well as currents for the emergence of different types of interactions, such as the DM interaction. In addition, in Ref. [23] all possible bilinear interactions were calculated individually and therefore contain much more information than the restricted fits presented in Fig. 1 of Ref. [27] which only contain the summed up and hence averaged values for pairs of the same type in the variation of interactions.

Direct calculations of the magnetic interactions, and a parametrization of the DM form, allows us to analyze the nature of the interactions involved in a material. Considering that the spin-spin interactions can be calculated in terms of the Green's function loop $\mathrm{sp}\boldsymbol{\sigma}\mathbf{G}_{ij}\boldsymbol{\sigma}\mathbf{G}_{ji}$, where sp denotes the trace over spin-1/2 space, it is clear that the electronic structure, which is captured in the single-electron Green's function $\mathbf{G} = (G^{00} + G^{01})\sigma^0 + (G^{\mu 0} + G^{\mu 1})\sigma^\mu$, enables additional insight into the origin of the interactions. As discussed in Refs. [22,23] the asymmetric anisotropic interaction $\mathbf{D}_{ij}$, occurring in the product $\mathbf{D}_{ij}\cdot(\mathbf{S}_i\times\mathbf{S}_j)$, can be expressed as the integral over the terms $G^{00}_{ij}G^{\mu 1}$ and $G^{01}G^{\mu 0}$, where the former depend on the charge density and spin currents, whereas the latter on the charge currents and spin density. Further analysis of these terms suggests that the asymmetric anisotropy $\mathbf{D}_{ij}$ can emerge from any electronic structure in which both the inversion and time-reversal symmetries are

simultaneously broken. An example of this is illustrated in Ref. [15], where there is no spin-orbit interaction present, however, the interaction **D** is significant. Understanding the DM-like interaction in terms of currents opens up for designating the terms *relativistic* and *nonrelativistic* DM-like interactions, with contributions of different physical origin. While a spin-orbit interaction necessarily leads to both spin and charge current in the system which, hence, leads to that both contributions $G_{ij}^{00}G^{\mu 1}$ and $G^{01}G^{\mu 0}$ are finite, these two contributions are also finite in the presence of spin and charge currents which do not necessarily originate from a spin-orbit interaction. These contributions are also connected to the configuration dependence of the exchange-coupling parameter J_{ij} that have already been shown to significantly improve the comparison between theory and experiment in works such as Refs. [19,39]. It suggests that these interactions, labeled *improper* in Ref. [27], can be important when it comes to properties of excited states, such as, e.g., the magnon softening induced by temperature effects.

Another advantage of an LKAG-like approach to evaluating exchange interactions is that it enables us to analyze the microscopic origin of the exchange interaction by orbital decomposition of the Green's function. As an example, we mention the drastically different roles played by the $3d$ E_g and $3d$ T_{2g} orbitals, respectively, for the magnetic behavior of bcc Fe [21,40,41]. We note, however, that a drawback with the method used in Refs. [22,23] is the dependence on the reference frame, which leads to a difficulty in capturing the whole energy landscape, correctly pointed out by dos Santos *et al.* [27]. The method used in Refs. [22,23] is best suited for describing the energy landscape in a limited part of configuration space. This opens up for a discussion of local and global spin models, which we proceed with next.

Global and local energies. Recently, a distinction between local and global spin models was proposed [42]. Local models are valid for small magnetic fluctuations with respect to a magnetic reference state, in contrast to global spin models that are supposed to be valid for all magnetic states. Global spin models have by construction interaction parameters [27] that are independent of any reference state and they have to fulfill global symmetries because they aim to describe all magnetic states. While such magnetic interactions are required for global spin models, this does not apply to local models that are designed to map out a local Weiss field or torques obtained from gradients of an energy landscape in a smaller region of configuration space. This implies that these models do not have to fulfill global symmetry requirements, since they describe only small fluctuations with respect to a magnetic reference state, where the fluctuations can break global symmetries. A magnetic state dependence of the interaction coefficients arises naturally for local models due to their dependence on the reference state [42]. If the state dependence is taken into account for a local spin model, all global symmetries are recovered. Since the work in Refs. [22,23,36] belongs in the category of local spin models, the demand for global interaction parameters by Ref. [27] does not apply.

On the name convention. We end this Comment with a remark on classification. In Ref. [27] the term "improper" was used for the magnetic interaction discussed here. Since this term does not signify anything relating to the microscopic origin of the DM-like exchange interaction that results from the noncollinear electron states, as discussed here and in Refs. [22,23], we suggest that a noncollinear, or nonchiral, DM interaction is more appropriate terminology, since it alludes to physical properties and origins of this exchange interaction.

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