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Anisotropic superexchange through nonmagnetic anions with spin-orbit coupling

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Abstract. Anisotropic superexchange interaction is one of the most important interactions in realizing exotic quantum magnetism, which is traditionally regarded to originate from magnetic ions and has no relation with the nonmagnetic ions. In our work, by studying a multi-orbital Hubbard model with spin-orbit coupling on both magnetic cations and nonmagnetic anions, we analytically demonstrate that the spin-orbit coupling on nonmagnetic anions alone can induce antisymmetric Dzyaloshinskii-Moriya interaction, symmetric anisotropic exchange and single ion anisotropy on the magnetic ions and thus it actually contributes to anisotropic superexchange on an equal footing as that of magnetic ions. Our results promise one more route to realize versatile exotic phases in condensed matter systems, long-range orders in low dimensional materials and switchable single molecule magnetic devices for recording and manipulating quantum information through nonmagnetic anions.

1 Introduction

Locking electron spin and momentum together, the relativistic spin-orbit coupling (SOC) plays a critical role in realizing a diversity of exotic phases in condensed matter systems, such as quantum spin liquid, spin-orbit coupled Mott insulator, Weyl semimetal and topological insulator [1–3]. In the absence of SOC, the magnetic interaction is isotropic with spin rotational invariance. However, SOC may lower the symmetry and lead to anisotropic interactions, which has been microscopically identified by Moriya by means of extending the Kramers-Anderson superexchange theory [4–7]. Importantly, the magnetic anisotropy is the key in bond-dependent Kitaev interaction and phase transition in low dimensional ($D \leq 2$) systems, which has been argued to be a great promise for quantum computation and information processing in addition to the fundamental interest [8–10].

Recently, magnetic orders induced by magnetic anisotropy have been reported in experiments on some two-dimensional (2D) materials or proposed by numerical simulations [11–16]. Since it is well known that the Hohenberg-Mermin-Wagner theorem forbids the spontaneous breaking of continuous symmetry at finite temperature in low dimensional systems [17], these findings are thus attributed to magnetic anisotropy in the materials. Commonly, the magnetic anisotropy is believed to dominantly result from the SOC on magnetic ions. Nevertheless, intuitively, the SOC on the nonmagnetic anions should also induce spin flip in the virtual hopping process of superexchange. Indeed, in ferromagnetic CrI_3 and $CrGeTe_3$ monolayers, anisotropic exchange coupling and single ion anistropy are found to be dominated by the SOC from the nonmagnetic $5p$ Iodine or Tellurium anions rather than the magnetic $3d$ Cr ions [14,15]. Although evidences have been provided numerically by comparing the relevant magnetic couplings with and without SOC [14,15], from the theoretical point of view, an insight into the microscopic mechanism to understand how the SOC on nonmagnetic ions induces the magnetic anisotropic superexchange on magnetic ions is of both importance and interest [3,10,18]. Actually, some relavent studies have been started since 1980s, which focuse on the RKKY interaction via the conduction electron with spin-orbit coupling [19,20].

In this paper, we consider a general system in which magnetic ions interact through the superexchange via nonmagnetic ions with SOC on the latter. Starting from a multi-orbital model with orbital hybridization, onsite Coulomb interaction and SOC on both magnetic cations and nonmagetic anions, we first transform this model into a multiorbital model with both spin-isotropic and spin-anisotropic electron hopping integrals between magnetic and nonmagnetic ions. Next, applying the degenerate

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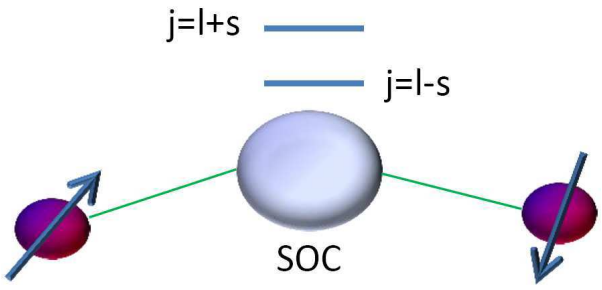

Fig. 1. Superexchange between two magnetic ions via a nonmagnetic ligand with spin-orbit interaction. Apart from the crystalline electric field splitting, the SOC on the nonmagnetic ligands could further split the energy levels in two fine structures with the spin moment parallel and anti-parallel to the orbital moment, respectively. The SOC on ligands could induce spin flip in the virtual hopping process of superexchange.

perturbation theory, we obtain the isotropic exchange, aniso-tropic exchange or pseudodipolar interaction, antisymmetric Dzya-loshinskii-Moriya (DM) interaction and single-ion anisotropy on magnetic ions. We show that the SOC on nonmagnetic ions contributes to magnetic anisotropy in a similar way as that on the magnetic ions. Moreover, the anisotropic magnetic couplings increase sharply with the atomic number. Therefore, the heavy nonmagnetic ligands may contribute dominantly to the magnetic anisotropy rather than the light magnetic ions.

2 Microscopic Hamiltonian

We model a general system containing magnetic cations M surrounded by nonmagnetic anions or ligands NM, as shown in Figure 1. The central interactions include the spin-orbit angular momentum coupling $\mathbf{L}\cdot\mathbf{s}$ on both M and NM ions, and the onsite Coulomb interaction U_d and U_p between electrons on the M and NM ions, respectively. The perturbation term is assumed to be the electron transfer integral t_{imkn} between the mth orbital of M ion at site i and the nth orbitals of the ligand NM at site k. We neglect the direct hopping between M sites. Then, the Hamiltonian can be written as

$$H = H_d + H_p + H_{pd}, \tag{1}$$

where

$$H_{c=p,d} = H_{c0} + \frac{1}{2}\sum_{imn\alpha\beta} U_{cm\alpha n\beta} n_{cim\alpha} n_{cin\beta}, \tag{2}$$

$$H_{pd} = \sum_{ikmn\alpha}\left(t_{imkn} d^{\dagger}_{im\alpha} p_{kn\alpha} + H.c.\right), \tag{3}$$

with

$$H_{c0} = \sum_{im\alpha n\beta} E^{ci}_{m\alpha,n\beta} c^{\dagger}_{im\alpha} c_{in\beta}, \tag{4}$$

$$E^{ci}_{m\alpha,n\beta} = \varepsilon_{cm0}\delta_{mn}\delta_{\alpha\beta} + \lambda_c \left\langle l_c m\alpha \left| \mathbf{L}_{ci}\cdot\mathbf{s} \right| l_c n\beta \right\rangle, \tag{5}$$

where $d^{\dagger}_{im\alpha}$ is the creation operator of an electron with spin α on the mth orbital of ith M ion, and ε_{dm0} is the energy of the mth orbital. We assume that the crystalline electric field effect has been taken into account by lifting the degeneracy of the orbital energy levels. $p^{\dagger}_{kn\alpha}$ is the creation operator of an electron on the mth orbital with spin α. n_d and n_p are the particle number operators of the electrons on M and NM sites, respectively. $\mathbf{L}$ and $\mathbf{s}$ denote the orbital and spin angular momenta. λ_d and λ_p are the spin-orbit coupling constants on the M and NM ions.

It is easy to diagonalize the single-site Hamiltonian matrix E^{ci} firstly [9,21]. In general, we have a site-dependent diagonal eigenvalue matrix $\varepsilon_{ic} = W^{ci\dagger} E^{ci} W^{ci}$ with the unitary eigenvector matrix W^{ci}. Choosing real orbital wave functions, then the $\mathbf{L}_c$ matrix elements are purely imaginary. ε_{ci} has l_c eigenvalues with the orbital angular momentum quantum number l_c and every single-particle energy of ε_{ci} has at least a twofold degeneracy due to time reversal symmetry, characterized by pseudospin quantum numbers, $\pm 1/2$. Consequently, a new set of electron creation and annihilation operators is introduced for p and d electron operators

$$q_{kn\alpha} = \sum_{l\beta} W^{pk}_{n\alpha,l\beta} p_{kl\beta}, a_{im\alpha} = \sum_{n\beta} W^{di}_{m\alpha,n\beta} d_{in\beta}. \tag{6}$$

Let's express the microscopic Hamiltonian in terms of $q_{km\alpha}$ and $a_{im\alpha}$ as

$$H_p = \sum_{kn\alpha} \varepsilon_{pkn} n_{qkn\alpha} + \frac{1}{2}\sum_{if\rho h\sigma l\tau r\upsilon} V^{pk}_{f\rho h\sigma l\tau r\upsilon} q^{\dagger}_{kf\rho} q_{kh\sigma} q^{\dagger}_{kl\tau} q_{kr\upsilon}, \tag{7}$$

$$H_d = \sum_{im\alpha} \varepsilon_{dim} n_{aim\alpha} + \frac{1}{2}\sum_{if\rho h\sigma l\tau r\upsilon} V^{di}_{f\rho h\sigma l\tau r\upsilon} a^{\dagger}_{if\rho} a_{ih\sigma} a^{\dagger}_{il\tau} a_{ir\upsilon}, \tag{8}$$

$$H_{pd} = \sum_{imkn\alpha} a^{\dagger}_{im\alpha}(b_{imkn}\delta_{\alpha\beta} + \mathbf{C}_{imkn}\cdot\boldsymbol{\sigma}_{\alpha\beta}) q_{kn\beta} + H.c. \tag{9}$$

with

$$V^{pk}_{f\rho h\sigma l\tau r\upsilon} = \sum_{mn\alpha\beta} U_{pm\alpha n\beta} W^{pk}_{m\alpha,f\rho} W^{pk\dagger}_{m\alpha,h\sigma} W^{pk}_{n\beta,l\tau} W^{pk\dagger}_{n\beta,r\upsilon},$$

$$V^{di}_{f\rho h\sigma l\tau r\upsilon} = \sum_{mn\alpha\beta} U_{dm\alpha n\beta} W^{di}_{m\alpha,f\rho} W^{di\dagger}_{m\alpha,h\sigma} W^{di}_{n\beta,l\tau} W^{di\dagger}_{n\beta,r\upsilon},$$

the spin-isotropic hopping matrix elements

$$b_{imkn} = \frac{1}{2}\sum_{\alpha\gamma m' n'} W^{di}_{m\alpha,m'\gamma} t_{im'kn'} W^{pk\dagger}_{n'\gamma,n\alpha}, \tag{10}$$

and the spin-anisotropic matrix elements

$$\mathbf{C}_{imkn} = \frac{1}{2} \sum_{\alpha\beta\gamma m'n'} W^{di}_{m\alpha,m'\gamma} t_{im'kn'} W^{pk\dagger}_{n'\gamma,n\beta} \boldsymbol{\sigma}_{\alpha\beta}, \quad (11)$$

where $\boldsymbol{\sigma}$ is the vector of the three Pauli matrix. Clearly, if we ignore the SOC on nonmagnetic anions, namely, setting W^{pk} a unit matrix, Moriya's result is reproduced [4,9,21]. On the other hand, ignoring the SOC on the magnetic cations, i.e. setting W^{di} a unit matrix, we still reach a similar result. Formally, the SOC on nonmagnetic ions contributes to both the spin-isotropic and spin-anisotropic hopping matrix elements in a similar way to that on the magnetic ions. In the weak SOC limit, the spin-anisotropic hopping matrix elements are in the linear order of $\lambda_c/\Delta\varepsilon_c|_{c=p,d}$ with the energy difference $\Delta\varepsilon_c$ between the ground state and the excited state.

3 Superexchange interaction

For magnetic materials, it is often assumed that localized electrons yield both orbital and spin magnetic moments on each ions or part of ions. The direction of a magnetic moment is described by a classical unit vector. It is natural to define an effective spin to match up with the local magnetization. Here, we define effective spin operators in terms of the creation $a^\dagger_{im}$ and annihilation a_{im} operators as

$$\mathbf{S}_i = \frac{1}{2} \sum_{\alpha\beta m} a^\dagger_{im\alpha} \boldsymbol{\sigma}_{\alpha\beta} a_{im\beta}. \quad (12)$$

Due to the time-reversal symmetry, the fully occupied pseudospin levels give no contribution to magnetization. Therefore, the magnetization is dominated by the half-occupied effective spin levels. Moreover, on magnetic atoms, the electron kinetic energy or H_{pd} is often weak enough to be regarded as perturbation, and then the generalized Hubbard Hamiltonian H in equation (1) is reduced to an effective spin interaction Hamiltonian of magnetic ions, mediated by the nonmagnetic ions. To obtain the spin interaction between magnetic ions M, it is customary to appeal to the degenerate perturbation theory by taking the hybridization H_{pd} or hopping as the perturbation. We consider the case that the hopping is much less than onsite Coulomb interactions $U_{p,d}$ and the charge transfer energy between NM and M ions. A common way is to use the limit $U_d \to \infty$ to forbid double occupancy in the magnetic atom orbitals. According to Goodenough-Kanamori rule, half-occupied orbitals dominate the exchange coupling and fully occupied orbitals' contribution is much weaker [6,7]. We define a projection operator $\hat{P}_g$, which projects onto the subspace with only single electron locating on each pseudospin levels of magnetic ions, and another projector $\hat{P}_e = \hat{I} - \hat{P}_g$ with the unit operator $\hat{I}$. We take the hopping term H_{pd} as the perturbation to derive an effective Hamiltonian [21,22]. The nth order perturbation reads,

$$H^{(n)} = \hat{P}_g H_{pd} \left(\frac{1}{E - (H_d + H_p)} \hat{P}_e H_{pd} \right)^{n-1} \hat{P}_g, \quad (13)$$

with the ground state energy E. One finds the first and the third order perturbations vanish because odd-order hops introduce double or empty occupancy on the originally half-filled magnetic atom orbitals, i.e. $H^{(1)} = H^{(3)} = 0$. The second-order contribution is dominated by the magnetic coupling between electrons on NM and M sites, e.g. $\mathbf{S}_{pk} \cdot \mathbf{S}_{di}$. However, this term gives no contribution because the total magnetic moment on each NM ion is zero in the ground state, or $\langle \mathbf{S}_{pk} \rangle = 0$. Consequently, the leading contribution to superexchange is the fourth order in hopping between M and NM ions, which consists of the "hops" described by spin-isotropic matrix elements b_{ik}, and the "hops" given by anisotropic C_{ik} matrix elements. We have assumed that the electron number fluctuation on the NM ions is weak enough so that it can be ignored.

In time-reversal invariant systems, the magnetic Hamiltonian consists of two-spin interactions between nearest and further neighbors, four spin interactions, and so forth [4,5,21,23]. In this paper, we confine to the two-spin interactions. A general bilinear two-spin interaction can be written as

$$H_{ex} = \sum_{ij} \mathbf{S}_i \cdot \mathbf{M}_{ij} \cdot \mathbf{S}_j. \quad (14)$$

Usually, the 3×3 interaction-matrix $\mathbf{M}_{ij}$ is separated into an antisymmetric and a symmetric matrix

$$\mathbf{M}^{\mp}_{ij} = \frac{\mathbf{M}_{ij} \mp \mathbf{M}^T_{ij}}{2}. \quad (15)$$

Further, the symmetric $\mathbf{M}^+_{ij}$ is often split into an isotropic coupling matrix $J_{ij}\mathbf{I}$, and a symmetric traceless one

$$\boldsymbol{\Gamma}_{ij} = \mathbf{M}^+_{ij} - J_{ij}\mathbf{I} \quad (16)$$

where $J_{ij} = Tr(\mathbf{M}_{ij})/3$ and $\mathbf{I}$ is a unit matrix. Alternatively, the eigenvalues of the symmetric matrix $\mathbf{M}^+_{ij}$ are obtained by diagonalization, and then separated into the isotropic exchange and Kitaev coupling [14]. The antisymmetric exchange is commonly written as DM interaction $\mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j)$ since each antisymmetric 3×3 matrix can be linearly mapped onto a 3D vector, $(\mathbf{M}^-_{ij})_{\mu\nu} = \sum D_{ij,\lambda} \varepsilon_{\lambda\mu\nu}$ with Levi-Civita symbol $\varepsilon_{\lambda\mu\nu}$ and Cartesian components λ, μ and ν. Thus, the exchange Hamiltonian is rewritten in terms of the isotropic Heisenberg exchange, DM interaction, and symmetric anisotropic exchange as

$$H_{ex} = \sum_{ij} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j) + \mathbf{S}_i \cdot \boldsymbol{\Gamma}_{ij} \cdot \mathbf{S}_j. \quad (17)$$

Casting off some constants and hopping terms, the 4th order perturbation $H^{(4)}$ in equation (13) is rearranged into

H_{ex} form. The interaction matrix tensors in H_{ex} include the isotropic Heisenberg coupling

$$J_{ij} = 4 \sum_{knk'n'} s_{ijkn} g_{knk'n'} s_{jik'n'}, \tag{18}$$

Dzyaloshinskii-Moriya vectors

$$\mathbf{D}_{ij} = -4i \sum_{knk'n'} g_{knk'n'} \left(\mathbf{v}_{ijkn} s_{jik'n'} - s_{ijkn} \mathbf{v}_{jik'n'} \right), \tag{19}$$

and symmetric anisotropic exchange

$$\begin{aligned} \mathbf{\Gamma}_{ij} = & \ 4 \sum_{knk'n'} g_{knk'n'} \left(\mathbf{v}_{ijkn} \mathbf{v}_{jik'n'} + \mathbf{v}_{jikn} \mathbf{v}_{ijk'n'} \right) \\ & - g_{knk'n'} \mathbf{I} \left(\mathbf{v}_{ijkn} \cdot \mathbf{v}_{jik'n'} \right) \end{aligned} \tag{20}$$

with unit matrix $\mathbf{I}$. To simplify the expressions of the coupling matrix, the effective spin-isotropic hopping matrix elements have been introduced between i and j magnetic ions after the definition by Chen and Balents [9]

$$s_{ijkn} = b_{ikn} b_{knj} + \mathbf{C}_{ikn} \cdot \mathbf{C}_{knj}, \tag{21}$$

the effective spin-anisotropic hopping matrix elements

$$\mathbf{v}_{ijkn} = \mathbf{C}_{ikn} b_{knj} + b_{ikn} \mathbf{C}_{knj} + i \left(\mathbf{C}_{ikn} \times \mathbf{C}_{knj} \right), \tag{22}$$

and the coefficients originating from the third power of $(E - H_d - H_p)^{-1}$ in the 4th order perturbation $H^{(4)}$

$$\begin{aligned} g_{knk'n'} = & \ \frac{\left(1 - \frac{1}{2}\delta_{kk'}\delta_{nn'}\right)}{E_{knk'n'}} \left(E_{kn}^{-1} + E_{k'n'}^{-1} \right)^2 \\ & + \frac{1}{E_{kn} E_j E_{k'n'}}, \end{aligned} \tag{23}$$

where E_{kn} is the energy difference between the ground state and the intermediate excited state with one electron or hole hopping to the nth orbital on k site from the i or j site. Commonly, as a function of ε_d and ε_p as well as the onsite Coulomb interaction U_d and U_p, the detail value of E_{kn} depends on the electron configuration. Similarly, E_j is the single particle excitation energy for the j ions. $E_{knk'n'}$ corresponds to the double-electron or hole activation energy for two electrons or holes hopping to the nth orbital on the k site and the n'th orbital on the k' site from i and j site.

In addition, the SOC on nonmagnetic ligands could also induce the local spins on magnetic ions to align along a specific local axis, i.e. the single-ion anisotropy or magnetocrystalline anisotropy. The single-ion anisotropy could be of the same order in magnitude as the pseudodipole or Kitaev interactions [14]. It is written as a lattice-dependent onsite tensor

$$\begin{aligned} \mathbf{\Gamma}_{ii} = & \ 4 \sum_{knk'n'} 2\mathbf{v}_{iikn} g_{knk'n'} \mathbf{v}_{iik'n'} \\ & - g_{knk'n'} \mathbf{I} \left(\mathbf{v}_{iikn} \cdot \mathbf{v}_{iik'n'} \right). \end{aligned} \tag{24}$$

This provides a novel route to engineer the single-molecule magnetic switch devices for storing or manipulating information with the aid of spins.

4 Discussion and conclusion

DMI and anisotropic exchange as well as the single ion anisotropy depend on the geometrical symmetry of lattice. The introduction of the SOC effect of ligands actually contributes to anisotropy on an equal footing as that of magnetic ions via the hopping matrix, and maintains the symmetries of both the hopping matrix elements and crystal field levels. Therefore, the dependence of the magnetic anisotropy on the crystal symmetry is just the same as that of the SOC effects on the magnetic ions. The DMI and anisotropic exchange are still anti-symmetric and symmetric, respectively, e.g. $D_{ij} = -D_{ji}$ and $\Gamma_{ij} = \Gamma_{ji}$. For example, in a system without inversion symmetry, both DMI and anisotropic exchange could emerge, depending on the detail geometric lattice symmetry. On the other side, in a system with inversion symmetry, DMI disappears but the anisotropic exchange could be preserved. If the symmetry of the hopping matrix elements alters, then the magnetic anisotropy may change accordingly. For instance, a magnetic ion has no single ion anisotropy in a cubic lattice surrounded by 6 same ligands. When one ligand is replaced by a nonmagnetic ion with higher SOC and even the crystalline electric field is kept the same, the single ion anisotropy is induced with the magnetic moment vector alone the line connecting the magnetic ion and the substituted ligand. The original geometric symmetry is broken due to the different SOC of the substituted ligand and hence the hopping matrix elements.

Although we assume that the single-site energies ε_{ic} may depend on the site index, in certain situations, for example, with the same crystalline electric field for each magnetic ion, it is possible to define site-dependent transformations W^{ic} such that the single-particle energies are site independent. However, the overlaps between orbital states, e.g. the spin-anisotropic matrix elements $\mathbf{C}_{imjn}$ and the symmetric anisotropic exchange $\mathbf{\Gamma}_{ij}$ could be bond-dependent due to the orbital anisotropy and crystal lattice geometry, including frustration [24]. Therefore, taking advantage of the properties of the heavy nonmagnetic ligands, additional methods are provided to design potential exotic spin models, such as the Kitaev model with bond-dependent anisotropic interaction.

Within Moriya's theoretical framework, the DM interaction and the anisotropic exchange are proportional to λ_c and their quadratic forms, respectively. Moreover, λ_c increases sharply with the atomic number Z_c, namely, $\lambda_c \propto Z_c^4$. Since the SOC on both the nonmagnetic ligands and the magnetic ions contributes in the same way to the magnetic anisotropy, the magnetic anisotropy is thus dominated by the heavier ions. For example, in the ferromagnetic CrI_3 and $CrGeTe_3$ monolayers, the leading contribution to the spin-anisotropic exchanges is from the SOC on the nonmagnetic $5p$ Iodine or Tellurium anions rather than from the magnetic $3d$ Cr ions because the atomic number $Z_I = 53$ and $Z_{Te} = 52$ are much larger

than that of Cr ions with $Z_{cr} = 24$. Although the nearest neighbor DM interaction vanishes due to the inversion lattice symmetry the DM term may appear between the next-nearest neighbors [11]. It is also intriguing to revisit the magnetic properties in iron-based superconductors since the atomic number of the surrounding nonmagnetic anions is comparable to that of the irons [25].

Finally, although we neglect the direct hoppings between magnetic ions, it is straightforward to include them in our formula [9].

To conclude, we have presented an analytical study of the superexchange of magnetic cations through nonmagnetic anions with SOC. We show that the SOC on nonmagnetic ligands could induce the anisotropic exchange, DM interaction and single-ion anisotropy on their neighboring magnetic ions. The nonmagnetic ligands contribute to magnetic anisotropy in a similar way as the magnetic ions. Our work demonstrates that exotic quantum states in condensed matter systems, order phases in low dimensional systems and single-molecular magnetic device could be engineered by the SOC on nonmagnetic ligands.

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Author contribution statement

J.C. performed the theoretical calculations and analysis with assistance from J.Z. and Y.D. All authors contributed to the discussion and the writing of the manuscript.

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