Orbital Dzyaloshinskii-Moriya Exchange Interaction

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Supercexchange calculation is performed for multi-orbital band models with broken inversion symmetry. Orbital-changing hopping terms allowed by the symmetry breaking electric field lead to a new kind of orbital exchange interaction closely resembling the Dzyaloshinskii-Moriya spin exchange. Inversion symmetry breaking as present in surfaces and interfaces and a strong on-site repulsion, but not the spin-orbit interaction, are the requirements to observe the proposed effect. Mean-field phase diagram exhibits a rich structure including anti-ferro-orbital, ferro-orbital, and both single and multiple spiral-orbital phases in close analogy with the Skyrmion spin crystal phase recently discovered in thin-film chiral magnets.

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I. INTRODUCTION

Strong on-site repulsion transforms the Bloch bands of nearly-free electrons into an insulator where the residual low-energy dynamics is that of the spin degrees of freedom interacting with each other via the superexchange mechanism\textsuperscript{1}. For spin-orbit-coupled bands, the spin-flip hopping processes result in another type of spin exchange called the Dzyaloshinskii-Moriya (DM) interaction\textsuperscript{2} under the superexchange process. Symmetry-wise, local inversion symmetry breaking such as the bond distortion for a pair of adjacent magnetic orbitals, in addition to the spin-orbit interaction (SOI), is the pre-requisite for the DM interaction to make its appearance in a given system. Ordered magnetic ground state is modified from the DM interaction to make its appearance in a given system. Ordered magnetic ground state is modified from being collinear as a result of the DM exchange to favor spiral structure.

Meanwhile, inversion symmetry breaking (ISB) on the global scale takes place for surfaces and interfaces and affects the band structure with new effects such as the Rashba interaction\textsuperscript{3}. The situation was recently reviewed carefully in Refs. 4,5 where it was shown that the symmetry-breaking electric field along the surface-normal z-direction modifies the band structure within the xy-plane by allowing previously forbidden hopping processes. Examples are $p_x(p_y)\leftrightarrow p_z$ orbital hopping in the $p$-band, and $d_{xy}\leftrightarrow d_{xz}$ orbital hopping in the $t_2g$-band. It was further shown\textsuperscript{5} that the new hopping terms arising from ISB can be cast in the form $-\gamma \sum_{\mathbf{k}} \Psi_{\mathbf{k}}^\dagger \mathbf{L} \cdot (\mathbf{k} \times \hat{z}) \Psi_{\mathbf{k}}$ around the $\Gamma$ ($\mathbf{k}=0$) point, where $\Psi_{\mathbf{k}}$ is the collection of, say, $p$-orbital operators $(X_k, Y_k, Z_k)^T$ in momentum coordinates $\mathbf{k}$, $\gamma$ is a parameter measuring the degree of ISB, and $\mathbf{L}$ is the spin-1 orbital angular momentum (OAM) operator. From the structure of the new Hamiltonian it readily follows that each band will carry polarized OAM proportional to $h(\mathbf{k} \times \hat{z})$ with the respective helicities $h=+1,0,-1$. The enlargement of effective spin size from 1/2 (as in electrons’ spin) to 1 (as in degenerate $p$-orbital bands) results in the appearance of the third band that remains unpolarized. The chiral structure of the OAM, dubbed the “orbital Rashba effect”, can occur even in the complete absence of SOI, and has been confirmed by circular dichroism ARPES work on the weak-SOI material, Cu\textsuperscript{6}.

The new hopping processes allowed by ISB are in fact the orbital analogues of spin-flip hoppings in spin-orbit-coupled bands. Therefore, the two necessary conditions for the emergence of spin-DM interaction - ISB and SOI - are both effectively fulfilled for orbital magnetism when a symmetry-breaking electric field acts perpendicular to the two-dimensional surface. The purpose of this paper is to review this situation carefully in the limit of strong on-site interaction regime to ask if an orbital analogue of spin-DM interaction exists. In Sec. II superexchange calculation is carried out for multi-orbital tight-binding Hamiltonian embodying the ISB. The emergence of the orbital DM interaction is demonstrated in Sec. III together with the phase diagram exhibiting spiral and multi-spiral structures. Possible observation of orbital DM-induced orbital-spiral phases in magnetic thin films is discussed in Sec. IV.

II. SUPEREXCHANGE WITH ISB

Assuming three degenerate $p$-orbital states at each site, a square lattice Hamiltonian with nearest-neighbor hopping $H_t = \sum_{i\sigma}(H_{i,i+\hat{x},\sigma} + H_{i,i+\hat{y},\sigma})$ is constructed,

$$
H_{i,i+\hat{x},\sigma} = t_a X_i^\dagger Y_i + t_b Y_i^\dagger Z_i + t_c Z_i^\dagger X_i + h.c. ,
$$

$$
H_{i,i+\hat{y},\sigma} = t_a Y_i^\dagger X_i + t_b X_i^\dagger Z_i + t_c Z_i^\dagger Y_i + h.c. .
$$

Two hopping integrals $t_a$ and $t_b$ are introduced for $\sigma$- and $\pi$-bonding orbital hoppings, respectively. Inter-orbital hopping becomes possible when the ISB parameter $\gamma$ is nonzero\textsuperscript{4,5}. All hopping parameters are real due to the assumed time-reversal invariance. Three-component spinor
can be formed, \( \psi_i = (X_i, Y_i, Z_i)^T \), representing the \( p_x \), \( p_y \), and \( p_z \)-orbitals at the site \( i \). Multi-orbital Hubbard interaction is\(^7\)

\[
H_U = U \sum_{i,\alpha} n_{i\alpha\uparrow} n_{i\alpha\downarrow} + \left( U - \frac{5}{2} J_H \right) \sum_{i,\alpha<\beta,\sigma,\sigma'} n_{i\alpha\sigma} n_{i\beta\sigma'} - 2J_H \sum_{i,\alpha<\beta} S_{i\alpha\uparrow} S_{i\beta\downarrow} + J_H \sum_{i,\alpha\neq\beta} S_{i\alpha\uparrow} S_{i\beta\downarrow}, \tag{2}
\]

where \( U \) and \( J_H \) are Coulomb and Hund’s exchange elements, \( \alpha, \beta \) and \( \sigma, \sigma' \) are orbital and spin indices, respectively, and \( n_{i\alpha\sigma} \) is a number operator counting the electron number in \( \alpha \)-orbital with spin \( \sigma \) at site \( i \).

Shekhtman \textit{et al.} showed how to carry out the superexchange calculation efficiently for spin-orbit-coupled bands by introducing unitary rotations for operators to absorb spin-flip hoppings\(^8\). We may adopt similar unitary rotations, in the orbital subspace, to remove orbital-changing hoppings from the Hamiltonian (1) with two unitary matrices

\[
U_x = \begin{pmatrix} \cos \frac{\theta}{2} & 0 & \sin \frac{\theta}{2} \\ 0 & 1 & 0 \\ -\sin \frac{\theta}{2} & 0 & \cos \frac{\theta}{2} \end{pmatrix}, \quad U_y = \begin{pmatrix} 1 & 0 & 0 \\ 0 & \cos \frac{\theta}{2} & \sin \frac{\theta}{2} \\ 0 & -\sin \frac{\theta}{2} & \cos \frac{\theta}{2} \end{pmatrix}. \tag{3}
\]

Two distinct rotations are required since the orbital-changing hopping mixes \( (p_x,p_z) \) orbitals along the \( x \)-direction, but \( (p_y,p_z) \) orbitals for the \( y \)-direction. As the new operators

\[
\tilde{\psi}_{i,x}(y) = U_x^{\dagger} \psi_{i,y},
\]

\[
\tilde{\psi}_{i,x} = U_x \tilde{\psi}_{i,x},
\]

\[
\tilde{\psi}_{i,y} = U_y \tilde{\psi}_{i,y}, \tag{4}
\]

are inserted in Eq. (1) one obtains a new hopping Hamiltonian \( \tilde{H}_t = \sum_{\sigma}(\tilde{H}_{i,i+\hat{x},\sigma} + \tilde{H}_{i,i+\hat{y},\sigma}), \)

\[
\tilde{H}_{i,i+\hat{x},\sigma} &= (t + t_2) \tilde{X}_{i\alpha\sigma} \tilde{X}_{i+\hat{x},\alpha\sigma} + t_b \tilde{Y}_{i\alpha\sigma} \tilde{Y}_{i+\hat{x},\alpha\sigma} + (t - t_2) \tilde{Z}_{i\alpha\sigma} \tilde{Z}_{i+\hat{x},\alpha\sigma} + \text{h.c.}, \\
\tilde{H}_{i,i+\hat{y},\sigma} &= t_b \tilde{X}_{i\alpha\sigma} \tilde{X}_{i+\hat{y},\alpha\sigma} + (t + t_2) \tilde{Y}_{i\alpha\sigma} \tilde{Y}_{i+\hat{y},\alpha\sigma} + (t - t_2) \tilde{Z}_{i\alpha\sigma} \tilde{Z}_{i+\hat{y},\alpha\sigma} + \text{h.c.}, \tag{5}
\]

where \( t(\cos \theta, \sin \theta) = (t_1, 1), t_2 = (t_a + t_b)/2, t_1 = (t_a - t_b)/2 \). Although bearing the same notation, the meaning of the tilde operators appearing in \( \tilde{H}_{i,i+\hat{x},\sigma} \) is distinct from that in \( H_{i,i+\hat{x},\sigma} \) due to different sets of rotations involved.

Superexchange calculation at 1/6-filling (one particle per site) can proceed now via standard methods with the Hamiltonian \( \tilde{H} = \tilde{H}_t + H_U \), where the orbital-changing hopping terms are seemingly absent. The exchange Hamiltonian thus obtained, writing \( t + t_2 = t_{\hat{x}}, t_b = t_{\hat{y}} \), and \( t - t_2 = t_{\hat{z}} \), reads \( \mathcal{H} = \mathcal{H}_{\hat{x}} + \mathcal{H}_{\hat{y}} \)

\[
\mathcal{H}_{\hat{x}} = J_T \sum_i \left( S_i \cdot S_{i+\hat{x}} + \frac{3}{4} \right) \left( \hat{A}_{i,i+\hat{x}} + \hat{B}_{i,i+\hat{x}} - \hat{N}_{i,i+\hat{x}} \right) + J_E \sum_i \left( S_i \cdot S_{i+\hat{x}} - \frac{1}{4} \right) \left( -\hat{A}_{i,i+\hat{x}} + \hat{B}_{i,i+\hat{x}} + \hat{N}_{i,i+\hat{x}} \right) + J_{A_1} \sum_\alpha \left( S_i \cdot S_{i+\hat{x}} - \frac{1}{4} \right) \left( \frac{2}{3} \hat{A}_{i,i+\hat{x}} + \frac{2}{3} \hat{C}_{i,i+\hat{x}} \right) + J_{T_2} \sum_\alpha \left( S_i \cdot S_{i+\hat{x}} - \frac{1}{4} \right) \left( \frac{1}{4} \hat{A}_{i,i+\hat{x}} - \frac{2}{3} \hat{C}_{i,i+\hat{x}} \right), \tag{6}
\]

where \( J_T = 2/(U - 3J_H) \), \( J_E = J_{T_2} = 2/(U - J_H) \), and \( J_{A_1} = 2/(U + 2J_H) \). Orbital exchange parts are given by

\[
\hat{A}_{i,i+\hat{x}} = \sum_\alpha t_{\alpha}^2 n_{i\alpha n_{i\alpha}}, \\
\hat{B}_{i,i+\hat{x}} = \sum_{\alpha<\beta} t_{\alpha} t_{\beta} (\alpha^\dagger_\beta \beta^\dagger_{i+\hat{x}} \alpha_{i+\hat{x}} + \beta^\dagger_\alpha \alpha^\dagger_{i+\hat{x}} \beta_{i+\hat{x}}), \\
\hat{C}_{i,i+\hat{x}} = \sum_{\alpha<\beta} t_{\alpha} t_{\beta} (\alpha^\dagger_\beta \beta^\dagger_{i+\hat{x}} \alpha_{i+\hat{x}} + \beta^\dagger_\alpha \alpha^\dagger_{i+\hat{x}} \beta_{i+\hat{x}}), \\
\hat{N}_{i,i+\hat{x}} = \frac{1}{2} \sum_{\alpha,\beta} (t_{\alpha}^2 n_{i\alpha} + t_{\beta}^2 n_{i+\hat{x},\beta}), \tag{7}
\]

where \( \alpha^\dagger_\beta \) annihilates (creates) electrons in the \( \alpha \)-orbital at \( i \)-site. \( \mathcal{H}_{\hat{y}} \) is easily obtained from \( \mathcal{H}_{\hat{x}} \) by switching \( t_{\hat{x}} \leftrightarrow t_{\hat{y}} \) and replacing \( i + \hat{x} \) by \( i + \hat{y} \). This Hamiltonian resembles the spin-orbital model describing LaTiO\(_3\) system studied by Khaliullin and his colleagues\(^9\) in the sense that in LaTiO\(_3\) system, each site has one active electron occupying one of three \( t_{2g} \)-orbitals. Yet, there are differences coming from the fact that while our model (6) after the rotation is dealing with three unequal hopping integrals between adjacent orbitals, in LaTiO\(_3\) system the \( \pi \)-hopping is ignored and the other two integrals have equal strengths\(^9\). By ignoring \( \pi \)-hopping and taking the limit \( t_1 + t_2 = t_1 - t_2 \) in Eq. (6), our superexchange Hamiltonian becomes identical to that of LaTiO\(_3\). Equations (6) and (7) constitute the main technical findings of the present work.

Some comments about the limiting cases are in order. The exchange Hamiltonian (6) simplifies greatly in the \( J_H = 0 \) limit,

\[
\mathcal{H}_{\hat{x}} = \frac{1}{4} \sum_\alpha \sum_{\alpha,\beta} \left( S_i \cdot S_{i+\hat{x}} + \frac{1}{4} \right) t_{\alpha} t_{\beta} (\alpha^\dagger_\beta \beta^\dagger_{i+\hat{x}} \alpha_{i+\hat{x}} \right), \\
\mathcal{H}_{\hat{y}} = \mathcal{H}_{\hat{x}}(t_{\hat{x}} \leftrightarrow t_{\hat{y}}, i + \hat{x} \leftrightarrow i + \hat{y}), \tag{8}
\]

where \( U \) is taken to be unity. Furthermore, in the isotropic limit \( t_{\hat{x}} = t_{\hat{y}} = t_{\hat{z}} \) the pairwise exchange interaction in Eq. (8) possesses the SU(6) symmetry in the combined spin and orbital spaces\(^10\). The larger symmetry can be most easily seen by defining operators \( \Psi_{i,\alpha} = \)
\[ \varphi_i \alpha_\sigma \psi_j = \alpha_\sigma^\dagger \varphi_j, \] where \( \varphi_i \) (\( \varphi^\dagger_i \)) annihilates (creates) electrons with spin \( \sigma \) at \( i \)-site. The pairwise exchange Hamiltonian can be re-written in the manifestly SU(6)-invariant form \( H_{ij} = \sum_{K,K'} \Psi_j^K \Psi_i^K \Psi_{i,K'} \Psi_{j,K'} \), where \( K \) and \( K' \) run over six possible spin and orbital configurations. The SU(6) symmetry will remain for one-dimensional chain consisting of either \( H_\hat{z} \) or \( H_\hat{y} \) alone, but not for the two-dimensional model \( H_\hat{z} + H_\hat{y} \) due to the fact that two different sets of unitary rotations were used to arrive at the overall superexchange Hamiltonian, Eq. (8).

### III. ORBITAL DM AND PHASE DIAGRAM

In this section we explicitly point out the emergence of DM-type orbital exchange interaction in our model and study possible phase diagram using the site-factorization scheme. DM-type interactions will be recovered by un-rotating the Hamiltonian (7) to the original orbital basis as shown in Ref. 8. In doing so for our Hamiltonian (6) one encounters unwieldy expressions that simplify somewhat by taking the weaker \( \pi \)-bonding to zero: \( t_\theta = 0 \). This limit is usually taken in the superexchange calculation for \( d \)-orbital systems in transition metal oxides\(^9\), and more recently for \( p \)-orbital systems in an optical lattice\(^11\).

In such limit the orbital exchange operators in Eq. (7) can be re-expressed in terms of Gell-Mann matrices \( \lambda^\alpha \) (\( \alpha = 1 \cdots 8 \)) as follows:

\[
\mathcal{H}_{\hat{z}} = \frac{J_{\hat{z}}}{4} \sum_i \left( S_i \cdot S_{i\pm \hat{z}} + \frac{3}{4} \right) \left( t_{\hat{X}}^z [\lambda^\alpha + (\lambda^\alpha)^2] [\lambda^\alpha_{i\pm \hat{z}} + (\lambda^\alpha_{i\pm \hat{z}})^2] + t_{\hat{Z}}^z [\lambda^\alpha - (\lambda^\alpha)^2] [\lambda^\alpha_{i\pm \hat{z}} + (\lambda^\alpha_{i\pm \hat{z}})^2] \right) \\
- \frac{J_{\hat{\xi}}}{4} \sum_i \left( S_i \cdot S_{i\pm \hat{\xi}} - \frac{1}{4} \right) \left( t_{\hat{X}}^z [\lambda^\alpha + (\lambda^\alpha)^2] [\lambda^\alpha_{i\pm \hat{\xi}} + (\lambda^\alpha_{i\pm \hat{\xi}})^2] + t_{\hat{Z}}^z [\lambda^\alpha - (\lambda^\alpha)^2] [\lambda^\alpha_{i\pm \hat{\xi}} + (\lambda^\alpha_{i\pm \hat{\xi}})^2] \right) \\
+ \frac{J_{\xi}}{3} \sum_i \left( S_i \cdot S_{i\pm \hat{\xi}} - \frac{1}{4} \right) \left( t_{\hat{X}}^z [\lambda^\alpha + (\lambda^\alpha)^2] [\lambda^\alpha_{i\pm \hat{\xi}} + (\lambda^\alpha_{i\pm \hat{\xi}})^2] + t_{\hat{Z}}^z [\lambda^\alpha - (\lambda^\alpha)^2] [\lambda^\alpha_{i\pm \hat{\xi}} + (\lambda^\alpha_{i\pm \hat{\xi}})^2] \right) \\
- t_{\hat{X}}^z (n_{i,\hat{x}} + n_{i+\hat{z},\hat{x}}) - t_{\hat{Z}}^z (n_{i,\hat{z}} + n_{i+\hat{\xi},\hat{z}}),
\]

\[
\mathcal{H}_{\hat{y}} = \mathcal{H}_{\hat{z}}(t_{\hat{x}} \rightarrow t_{\hat{y}}, \lambda^\alpha \rightarrow \lambda^\beta, \lambda^\beta \rightarrow \lambda^\gamma, \lambda^\gamma \rightarrow \lambda^\delta, \lambda^\delta \rightarrow \lambda^\epsilon, \lambda^\epsilon \rightarrow \lambda^\zeta, i+\hat{x} \rightarrow i+\hat{y}, n_{i,\hat{x}} \rightarrow n_{i,\hat{y}}, n_{i+\hat{\xi},\hat{x}} \rightarrow n_{i+\hat{\xi},\hat{y}}).
\]

For convenience we introduced \( \lambda^\alpha = \text{diag}(1,0,-1) \) and \( \lambda^\beta = \text{diag}(0,1,-1) \) as combinations of \( \lambda^4, \lambda^5, \lambda^6 \) and the unit matrix. It is useful to note that \( \mathcal{H}_{\hat{z}} \) is entirely constructed from three matrices, \( T_{\hat{X}}^z = (\lambda^4, \lambda^5, \lambda^6) \), reducing to a set of Pauli matrices in the orbital subspace of \( (p_x, p_z) \). Similarly, \( \mathcal{H}_{\hat{y}} \) employs another set of three matrices and \( T_{\hat{y}}^z = (\lambda^4, \lambda^5, \lambda^6) \) reducing to Pauli matrices in the \( (p_y, p_z) \) orbital subspace.

Returning to the original basis \( (p_x, p_y, p_z) \) amounts to making the unitary replacements

\[
(\lambda^2, \lambda^6) \rightarrow (U_x \lambda^2 U_x^\dagger, U_x^\dagger \lambda^6 U_x), \\
(\lambda^3, \lambda^5) \rightarrow (U_y \lambda^3 U_y^\dagger, U_y^\dagger \lambda^5 U_y),
\]

in Eq. (9). After the rotation there appear terms linear in \( \gamma \),

\[
t_{\gamma} \sum_i \left( [\lambda^2 + (\lambda^2)^2] \lambda^6_{i+\hat{z}} - \lambda^6_{i\pm \hat{z}} [\lambda^2_{i\pm \hat{z}} + (\lambda^2_{i\pm \hat{z}})^2] \right)
\]

As mentioned earlier, \( (\lambda^4, \lambda^5, \lambda^6) \) are effectively replaced...
by the Pauli matrices $\tau = (\tau^x, \tau^y, \tau^z)$ within the $(p_x, p_z)$-orbital subspace, thus the first line of Eq. (11) becomes

$$t_\gamma \sum_i [\tau^x_i \tau^x_{i+\hat z} - \tau^y_i \tau^y_{i+\hat z}] = t_\gamma \sum_i \hat y \cdot [\tau_i \times \tau_{i+\hat z}].$$

This is the orbital analogue of the DM spin exchange, or “orbital DM” (ODM) exchange. The real-valued transition amplitudes obtained from the superexchange process necessarily excludes the imaginary $\tau_y$ operator, permitting $\hat y \cdot [\tau_i \times \tau_{i+\hat z}]$ as the only permissible form of DM interaction. Analogously, the second line of Eq. (11) reduces to the two-component $(p_y, p_z)$-orbital model with $(\lambda^0, \lambda^7, \lambda^y)$ acting as another set of Pauli matrices $\mu$ with the DM interaction, $2t_\gamma \sum_i \hat y \cdot [\mu_i \times \mu_{i+\hat y}]$. We emphasize that such Pauli matrix description (effective spin-1/2 model) must give way to the full Gell-Mann matrix formalism shown in Eq. (9) once $\mathcal{H}_z$ and $\mathcal{H}_y$ are combined in the two-dimensional lattice.

A similar Gell-Mann matrix expression appeared in the low-energy theory of hard-core three-component bosons confined in the optical lattice\textsuperscript{11} where, however, the influence of ISB on the superexchange process was not examined. Superexchange calculation for the $t_{2g}$-orbitals in the presence of GdFeO$_3$-type distortion was carried out by Ishihara et al.\textsuperscript{12} The oxygen distortions assumed in their work is staggered, in the sense that the net displacement vector of all the oxygen atoms is zero. On the contrary, we are dealing with the situation where displacement of the $p_z$ is uniform, due to external fields. Orbital analogue of the DM exchange as shown here might be anticipated on symmetry grounds, but has never been explicitly demonstrated before.

In proceeding to the mean-field analysis of the possible phases of our Hamiltonian we assume that $J_{Hi}$ is sufficiently large to favor the ferromagnetic spin state\textsuperscript{6}, \langle S_i \cdot S_j \rangle = 1/4. The approximation allows us to focus on the orbital sector, which makes Eq. (9) simple enough to be written down in original basis $\psi_i = (X_i \ Y_i \ Z_i)^T$ as Hamiltonian from those shown in Eq. (6), which we attempt to model with the extra parameter $A$. It is also an attempt to understand the possible phases of the orbital DM model within the wider perspective than might be allowed from second-order perturbation.

The zero-temperature phase diagram spanned by $(\gamma, A)$ with $t$ fixed to unity is shown in Fig. 1. Six Gell-Mann matrices, organized into two groups $T^y_i = (\lambda_i^0, \lambda_i^3, \lambda_i^7)$ and $T^y_i = (\lambda_i^0, \lambda_i^3, \lambda_i^7)$, are used to characterize the ground states by following their averages and their Fourier components: $T^y_k = \sum_i (T^y_i) e^{-ik \cdot r_i} (n = x, y)$. The Fourier analysis is particularly helpful in searching for modulated structures with long periods that often appear with the DM interaction.

When $\gamma = 0$ the Hamiltonian is expressed entirely in terms of two commuting matrices, $(\lambda_i^0, \lambda_i^y)$, with the ground state given by alternate occupations of $p_x$ and $p_y$ orbitals on the square lattice. This phase, called the antiferro-orbital (AFO) state, dominates the small-$\gamma$ region of the phase diagram. For $A$ small and $\gamma$ increasing beyond a critical value, one finds a first-order transition into a ferro-orbital (FO) state with $p_z$-orbital oc-
The emergence of orbital spiral phase is a natural consequence of the orbital DM exchange.

The new phase, indicated as MH in Fig. 1, is constructed as the equal-weight superposition of two helices with \( k_1 = \pm (\pi/2, \pi/2) \) and \( k_2 = \pm (\pi/2, -\pi/2) \), as well as two pairs of peaks at \( \pm (\pi, 0) \) and \( \pm (0, \pi) \). The intensities obtained from Fourier analysis of \( \langle T_1^+ \rangle \) (inset in Fig. 1) show stronger peaks at \( \pm (0, \pi) \) than at \( \pm (\pi, 0) \). On the other hand Fourier analysis of \( \langle T_2^+ \rangle \) revealed the Bragg peaks at \( \pm (\pi, 0) \) are brighter than at \( \pm (0, \pi) \). One can still draw a close parallel of the MH phase found in the present model to the square lattice of Skyrmions and anti-Skyrmions found in some models of spiral magnetism\(^{14,15} \), which also consists of multiple Bragg peaks at \( \pm (k, k) \) and \( \pm (k, -k) \) in its spin structure.

IV. CONCLUSION AND SUMMARY

Orbital ordering in multi-band Hubbard systems have been studied for several decades since the pioneering work of Kugel and Khomskii (KK)\(^{13} \). Extension of the original two-orbital KK model to three-orbital \( t_{2g} \) case has been thoroughly carried out by Khalilullin and collaborators\(^9 \). Recent works on the optical lattice of cold atoms also arrived at three-orbital exchange model\(^{11} \), without the spin degrees of freedom. Meanwhile, remarkable advances in the growth technique of ultrathin materials prompt considerations of the influence of ISB, \( \gamma \neq 0 \), on the electronic band structure and, as we discuss in this paper, on the orbital physics as well. With this background, we have derived the analogue of spin-DM exchange interaction as a natural consequence of ISB in the multi-orbital Hubbard model. Physical requirements for its appearance are the multi-orbital degeneracy and the loss of inversion symmetry, but not the spin-orbit interaction as in the spin-DM exchange.

Although the derivations presented in this paper are based on the \( p \)-orbital picture, the case of degenerate \( t_{2g} \)-orbitals with ISB can be worked out, without further calculation, by making the replacements \((p_x, p_y, p_z) \rightarrow (d_{yz}, d_{zx}, d_{xy}) \), and switching \( \sigma \leftrightarrow \pi \)-hopping integrals \( t_a \leftrightarrow t_b \) in all our results. It is thus expected that conclusions regarding the phase diagram as shown in Fig. 1 may be directly applicable to ultra-thin films made of transition-metal elements. Unlike the spin-DM interaction in materials, the governing factor \( \gamma \) responsible for the orbital-DM exchange can be imposed externally by the electric field in a controlled manner. Interesting quantum-orbital phases and transitions between them may be observed in a thin-film multi-orbital system subject to perpendicular electric field of variable strength.

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