

Bilayer triple-Q state driven by interlayer higher-order exchange interactions

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Using first-principles calculations and an atomistic spin model, we predict the stabilization of a bilayer triple-Q state in an atomic Mn bilayer on Ir(111) due to interlayer higher-order exchange interactions. Based on density functional theory (DFT) we study the magnetic interactions and ground state in a Mn monolayer and bilayer on the Ir(111) surface. We calculate the energy dispersion of spin spirals (single-Q states) to scan a large part of the magnetic phase space and to obtain constants of pairwise exchange interactions. By including spin-orbit coupling, we determine the strength of the Dzyaloshinskii-Moriya interaction. To reveal the role of higher-order exchange interactions in these films, we consider multi-Q states obtained by a superposition of spin spirals. For the Mn monolayer in fcc stacking on Ir(111), the triple-Q state exhibits the lowest total energy in DFT, while the Néel state is most favorable for hcp stacking. For the Mn bilayer on Ir(111), two types of the triple-Q state are possible. In both magnetic configurations, a triple-Q state occurs within each of the Mn layers. However, only in one of them, the spin alignment between the layers is such that nearest neighbor spins of different layers also exhibit the tetrahedron angles that characterize the triple-Q state. We denote this state—which has the lowest total energy in our DFT calculations—as the ideal bilayer triple-Q state. This state exhibits significant topological orbital moments within each of the two Mn layers which are aligned in parallel resulting in a large topological orbital magnetization. We interpret the DFT results within an atomistic spin model, which includes pairwise Heisenberg exchange, the Dzyaloshinskii-Moriya interaction, as well as higher-order exchange interactions. We classify the different types of higher-order interactions into intralayer terms, i.e., acting only between spins within one of the layers, and interlayer interactions, in which spins in both layers are involved. We point out the role of an odd or even distribution of spins in the multispin interactions between the layers. Finally, we demonstrate that the stabilization of the ideal bilayer triple-Q state in a Mn bilayer on Ir(111) can only be explained upon taking the effect of interlayer higher-order exchange interactions into account.

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I. INTRODUCTION

Noncollinear spin structures have recently moved into the focus of research in spintronics and nanomagnetism due to their promise for future applications in logic, memory, or novel computing devices [1–3]. The magnetic properties of a material are often described within an atomistic spin model which contains the Heisenberg pairwise exchange interaction, the Dzyaloshinskii-Moriya interaction [4,5] as well as the magnetocrystalline anisotropy energy. However, in itinerant magnets such as 3d transition metals it is *a priori* not clear whether pairwise exchange terms are sufficient for an adequate description of magnetic order. It has been demonstrated that higher-order exchange terms such as biquadratic

or four-spin interactions arise in a perturbative expansion of the Hubbard model with respect to the hopping term t over Coulomb repulsion U [6–8]. In the last decade, compelling evidence has been given for the relevance of such multispin interactions in ultrathin transition-metal films by combined experimental and theoretical studies. In these investigations, a number of intriguing magnetic ground states have been identified such as the triple-Q state [9–12], conical spin spirals [13,14], the *uudd* state [15,16], and nanoskyrmion lattices [17–20]. Higher-order interactions can also play an important role for the stabilization of topological spin structures such as isolated magnetic skyrmions or antiskyrmions [21].

So far, mainly ultrathin films consisting of a single atomic magnetic layer have been considered. For such systems, a spin model containing higher-order exchange interactions has been developed [8] and successfully applied to explain nontrivial magnetic ground states observed in various 3d transition-metal monolayers on surfaces [10,12,15,20,22,23]. However, spin models including the effect of higher-order interactions between layers in magnetic bilayers or multilayers are missing even though a conical spin spiral driven by higher-order exchange has been observed in a Mn bilayer on W(110) [13]. An understanding of magnetic ground states and higher-order interactions in bilayers is nontrivial as interactions with vertical

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components and truly three-dimensional hopping processes come into play.

In this work, we apply density functional theory (DFT) to explore the complex magnetism of both a hexagonal Mn monolayer (ML) and a hexagonal Mn bilayer (BL) grown pseudomorphically on the Ir(111) surface and directly compare the results to a freestanding Mn BL. We perform total energy DFT calculations for various collinear and noncollinear magnetic states with and without including spin-orbit coupling. We find strong antiferromagnetic (AFM) exchange interactions for the Mn ML and a stacking dependent magnetic ground state. For hcp Mn stacking, we obtain the Néel state with 120° angles between adjacent spins as the ground state consistent with recent experimental [24] and theoretical [25] work. For fcc Mn stacking, we predict a triple-Q state with tetrahedron angles between adjacent spin moments.

For the Mn BL on Ir(111), we obtain AFM exchange interactions within the top and bottom Mn layer as well as AFM exchange between the two layers. As a result a rowwise AFM state with an effective AFM coupling between the layers is most favorable among all spin spiral states. However, upon considering superposition states of spin spirals, i.e., multi-Q states, we find that a triple-Q state is the energetically lowest magnetic configuration in the bilayer. For a magnetic bilayer two types of the triple-Q state are possible. In both of them the spins of adjacent Mn atoms within a layer exhibit tetrahedron angles. However, only one of the two triple-Q states possesses in addition tetrahedron angles between adjacent spins of different layers which we denote in the following as the ideal bilayer triple-Q state. This ideal bilayer triple-Q state is favored for the Mn BL on Ir(111). For this state the spin moments exhibit an effective AFM coupling between the layers and the total spin moment vanishes. However, significant topological orbital moments are induced in the triple-Q state due to its noncoplanarity [11,12,26]. In the ideal bilayer triple-Q state, the topological orbital moments of the two layers couple ferromagnetically resulting in a significant total topological orbital magnetization [27].

The core of our computational DFT approach is the calculation of the energy dispersion of flat spin spirals propagating either in only one or simultaneously in two magnetic layers. By mapping the total energies to a Heisenberg model, we can determine both intra- and interlayer pairwise exchange constants. While the treatment of spin spiral states in magnetic MLs represents a well-established method to extract the intralayer Heisenberg exchange parameters, the calculation of the spin spiral energy dispersion of two interacting magnetic layers is a computationally and conceptually much more demanding task. We present a detailed introduction to the underlying framework for the model of pairwise Heisenberg exchange in hexagonal magnetic bilayers.

We transfer the concept of multi-Q states—such as the up-up-down-down (*uudd*) [28] and triple-Q [9] state—from magnetic MLs to BLs. Similar to their ML analogues, the prototypical multi-Q states of hexagonal BLs are energetically strictly degenerate with their corresponding 1Q building blocks regarding the pairwise Heisenberg exchange. We develop an atomistic spin model which includes not only intralayer higher-order exchange interactions (HOI) [8] but also interlayer HOI. By classifying the HOIs into even and

odd terms with respect to the distribution of spins in the top and bottom layer we demonstrate that the ideal bilayer triple-Q (3Q) state in the Mn BL on Ir(111) is stabilized by the interplay of AFM interlayer pairwise exchange and interlayer higher-order exchange.

The paper is organized in the following way. Section II serves as an introduction to the calculation methodology of spin spiral (1Q) and multi-Q states for magnetic mono- and bilayers from first principles. We discuss how the obtained total energies can be mapped to an atomistic spin model which includes both intra- and interlayer pairwise Heisenberg and higher-order exchange interactions. In Sec. III, the applied computational DFT methods are described. The main part of the paper (Sec. IV) presents DFT total energy calculations of collinear and noncollinear spin states for Mn MLs on Ir(111), a freestanding Mn bilayer, and Mn BLs on Ir(111) as well as interpretation of the data within the context of the atomistic spin model developed in our work including intra- and interlayer HOI. The key aspects and results of the present work are summarized in Sec. V.

II. ATOMISTIC SPIN MODEL

In order to interpret the total energy DFT calculations of different collinear and noncollinear magnetic states including spin-orbit coupling, we apply an atomistic spin model that is based on the interactions between normalized magnetic moments $\mathbf{m}_i$ localized at atoms at lattice sites i . The general form of the Hamiltonian is given by

$$H = - \sum_{ij} J_{ij} (\mathbf{m}_i \cdot \mathbf{m}_j) - \sum_{ij} \mathbf{D}_{ij} \cdot (\mathbf{m}_i \times \mathbf{m}_j) - \sum_{ijkl} K_{ijkl} (\mathbf{m}_i \cdot \mathbf{m}_j) (\mathbf{m}_k \cdot \mathbf{m}_l) - \sum_i K_u (m_i^z)^2 \quad (1)$$

and includes pairwise Heisenberg exchange, the Dzyaloshinskii-Moria interaction (DMI) arising from spin-orbit coupling (SOC) [4,5], higher-order exchange interactions [6–8], and the uniaxial magnetocrystalline anisotropy. The strength of these terms is given by the respective constants J_{ij} , $\mathbf{D}_{ij}$, K_{ijkl} , and K_u . The magnitude of the magnetic moment at site i is denoted by M_i and the magnetic moment is given by $\mathbf{M}_i = M_i \mathbf{m}_i$. The sum of the higher-order exchange is performed over (i, j, k, l) , including equal indices. Therefore all terms of fourth order are contained, i.e., the four-spin three-site, four-spin four-site, and biquadratic exchange interactions. Note that we do not consider higher-order interactions of sixth order such as bicubic or topological chiral magnetic interactions [11,29] or higher-order terms due to spin-orbit coupling [30–32].

To parametrize the model, the total energies of different collinear and noncollinear magnetic states are computed by DFT with and without including spin-orbit coupling (details in Sec. III) and fitted to the spin model by minimizing the residual sum of squares. Depending on the geometry of the system and the interactions that need to be calculated, different magnetic states must be considered within DFT. In the next two subsections we describe how this spin model is applied to the cases of magnetic mono- and bilayers.

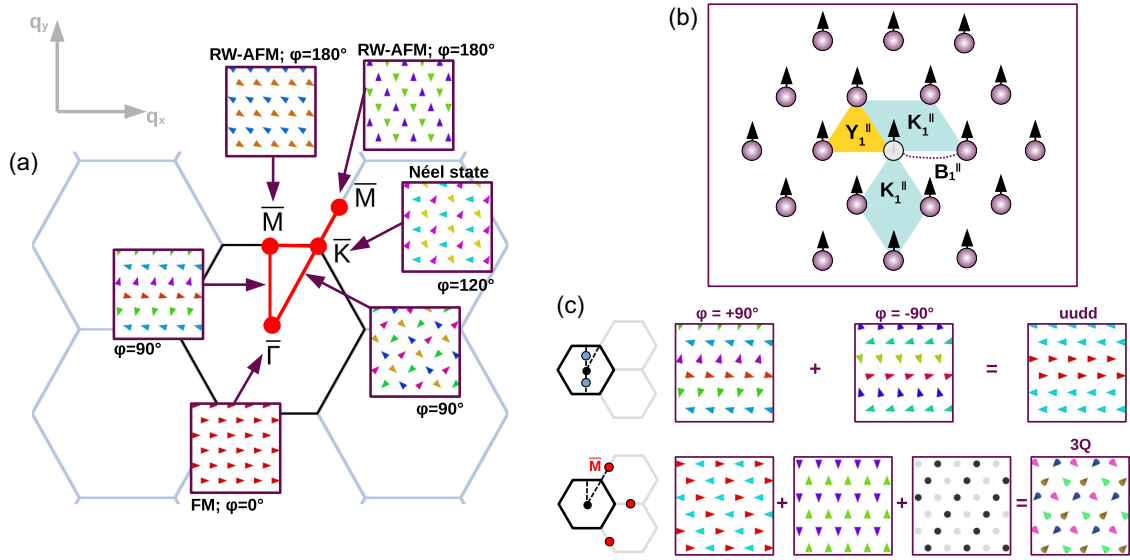


FIG. 1. Sketch of the 2D hexagonal Brillouin zone (BZ) with a selection of possible flat spin spiral states, illustration of intralayer higher-order exchange interactions (HOIs) on the 2D hexagonal lattice, and examples for the construction of multi-Q states for a magnetic ML. (a) Irreducible part of the hexagonal 2D-BZ (encircled in red) with its highsymmetry paths as well as the spin structures arising at the highsymmetry points $\bar{\Gamma}$, $\bar{K}$, and $\bar{M}$. (b) Minimal hopping paths within the NN approximation for an electron taking part in the biquadratic interaction denoted by its respective strength $B_1^{\parallel}$, the four-spin three-site ($Y_1^{\parallel}$) and the four-spin four-site interaction ($K_1^{\parallel}$). For the latter, two out of 12 possible diamonds are depicted in light blue color and one out of six possible triangles for $Y_1^{\parallel}$ is illustrated in orange color. (c) Formation of a collinear *uudd* state from a superposition of two counterpropagating 90° spin spirals at $\mathbf{q} = \pm \frac{1}{2} \bar{\Gamma}\bar{M}$ (in a similar fashion, the other *uudd* state is constructed from a superposition of left- and right-rotating 90° spin spirals at $\mathbf{q} = \pm \frac{3}{4} \bar{\Gamma}\bar{K}$) and the noncollinear 3Q state originating from the superposition of three $\mathbf{q}$ vectors at the symmetry-equivalent $\bar{M}$ -points.

A. Magnetic monolayers

The largest contribution to the energy of a magnetic state usually stems from the first term of Eq. (1), the pairwise Heisenberg exchange, emerging from the interplay of Pauli principle and Coulomb interaction. The sign of the exchange constant J_{ij} determines whether the system at hand prefers a parallel orientation, i.e., a ferromagnetic (FM) coupling ($J_{ij} > 0$), or an antiparallel alignment, i.e., an antiferromagnetic (AFM) coupling ($J_{ij} < 0$), between two magnetic moments $\mathbf{m}_i$ and $\mathbf{m}_j$. However, often the situation does not turn out to be that trivial and bilinear exchange beyond nearest neighbors with different signs of J_{ij} needs to be taken into account resulting in the formation of noncollinear magnetic ground states such as spin spirals on account of exchange frustration.

The second term in Eq. (1) describes the antisymmetric part of the exchange interaction, the DMI. Necessary ingredients for this relativistic effect to occur are both SOC and an inversion-asymmetric environment. In contrast to the symmetric pairwise Heisenberg exchange, the DMI prefers canted spin structures with a well-defined rotational sense owing to the anticommutativity of the cross product between two involved magnetic moments.

Due to the high symmetry of a single magnetic hexagonal layer, interactions between two atoms with the same distance always share the same interaction constant. Therefore the exchange constants J_{ij} as well as the magnitudes of the DM vectors D_{ij} can be organized in shells based on the distance of the involved atoms i and j (see Appendix A). By ordering the shells with increasing distance, each shell is given a number $\sigma \geq 1$ that uniquely identifies the interaction constants J_σ and the DM vectors magnitude D_σ .

Flat spin spirals with varying spiral vector $\mathbf{q}$ along high-symmetry directions of the Brillouin zone provide a set of magnetic states that make it possible to determine J_σ and D_σ by means of their energy dispersion $E(\mathbf{q})$ obtained via DFT. For the case of the pairwise Heisenberg exchange, the individual contributions of each interaction constant J_σ to $E(\mathbf{q})$ are given in Appendix A. At first, all J_σ are calculated by computing the energy of spin spirals without SOC since DMI arises from the latter. After that, the contribution of SOC on the energy is determined for each spin spiral state and mapped onto the DMI in the atomistic spin model.

Because of the periodicity of the atomic lattice only $\mathbf{q}$ vectors in the first two-dimensional (2D) Brillouin zone (BZ) need to be taken into account for the calculation of $E(\mathbf{q})$. As shown in Fig. 1(a), our DFT calculations are restricted to spin spiral vectors along the highsymmetry paths $\bar{\Gamma}\bar{M}$ and $\bar{\Gamma}\bar{K}\bar{M}$ around the irreducible wedge of the hexagonal BZ. In a flat spin spiral, the direction of a magnetic moment at a specific lattice site $\mathbf{R}_i$ is given by $\mathbf{m}_i = (\cos(\mathbf{q} \cdot \mathbf{R}_i), \sin(\mathbf{q} \cdot \mathbf{R}_i), 0)^T$ where the dot product of the reciprocal space vector $\mathbf{q}$ and the real space vector $\mathbf{R}_i$ yields the angle φ_i , denoting the spin orientation in the xy plane. Note, that the rotation plane of the spin spiral can be chosen arbitrarily as long as SOC is neglected.

Figure 1(a) illustrates important spin spiral states of a hexagonal magnetic ML arising at the highsymmetry points of the 2D BZ: at the $\bar{\Gamma}$ point corresponding to $\mathbf{q} = 0$ the FM state with a relative angle of $\varphi = 0^\circ$ between adjacent moments occurs, whereas at the $\bar{M}$ point the rowwise antiferromagnetic (RW-AFM) ($\varphi = 180^\circ$) and at the $\bar{K}$ point the Néel state with angles of 120° between adjacent moments can be found.

Note that due to the symmetry of the pairwise Heisenberg exchange [see first term of Eq. (1)] the energies of left-(- $\mathbf{q}$) and right-rotating (+ $\mathbf{q}$) spin spirals with the same angle φ between neighboring spins are degenerate if SOC and thereby DMI is neglected.

Spin spiral states also denoted as single-Q (1Q) states with symmetry-equivalent $\mathbf{q}$ vectors can be coupled to so-called multi-Q states by higher-order interactions (HOI) beyond the model of pairwise Heisenberg exchange arising in fourth-order perturbation theory from a multiband Hubbard model [8] [third term in Eq. (1)]. For a magnetic ML, we restrict the discussion to the following HOI terms [8] throughout this paper:

$$H_{\text{HOI}}^{\parallel} = - \sum_{(ij)} B_1^{\parallel} (\mathbf{m}_i \cdot \mathbf{m}_j)^2 - 2 \sum_{(ijk)} Y_1^{\parallel} (\mathbf{m}_i \cdot \mathbf{m}_j) (\mathbf{m}_i \cdot \mathbf{m}_k) - \sum_{ijkl} K_1^{\parallel} [(\mathbf{m}_i \cdot \mathbf{m}_j) (\mathbf{m}_k \cdot \mathbf{m}_l) + (\mathbf{m}_i \cdot \mathbf{m}_l) (\mathbf{m}_j \cdot \mathbf{m}_k) - (\mathbf{m}_i \cdot \mathbf{m}_k) (\mathbf{m}_j \cdot \mathbf{m}_l)]. \quad (2)$$

The coupling strengths within the magnetic layer are denoted by $B_1^{\parallel}$, $Y_1^{\parallel}$, and $K_1^{\parallel}$ for the biquadratic, four-spin three-site, and four-spin four-site interaction, respectively. In a similar way, the intralayer pairwise Heisenberg exchange constants J_{σ} and the magnitudes of the DMI vectors D_{σ} will obtain the superscript $\parallel$, denoting an interaction between moments within one layer, in this work. Figure 1(b) illustrates the interacting moments of minimal distances of the above-mentioned interactions: for the four-spin three-site (four-site) interaction the moments lie on an equilateral triangle (diamond). The interacting moments are nearest neighbors (NN), except for the four-spin four-site interaction on the diamond, which contains a next-nearest neighbor (NNN) connection leading to the minus sign in the last term of Eq. (2), i.e., if the NNN spin moments appear in one of the two scalar products [8].

On the hexagonal lattice of a magnetic ML, three prototypical multi-Q states can arise from the superposition of spin spiral (1Q) states due to HOI. These are two collinear up-up-down-down (*uudd*) states [28] generated from a superposition of two 90° spin spirals with opposite rotational sense (at $\mathbf{q} = \pm \frac{1}{2} \Gamma\overline{\text{M}}$ and $\mathbf{q} = \pm \frac{3}{4} \Gamma\overline{\text{K}}$) and one three-dimensional noncollinear 3Q state [9,10] originating from a superposition of three $\mathbf{q}$ vectors at symmetry-equivalent $\overline{\text{M}}$ points [see Fig. 1(c)]. If only pairwise Heisenberg exchange was present, the multi-Q states and their respective single-Q states would be energetically degenerate. However, the HOIs introduced above lift this degeneracy according to the following set of coupled equations [8]:

$$\Delta E_{\overline{\text{M}}} = E_{\overline{\text{M}}}^{3\text{Q}} - E_{\overline{\text{M}}}^{1\text{Q}} = \frac{16}{3} (2K_1^{\parallel} + B_1^{\parallel} - Y_1^{\parallel}), \quad (3)$$

$$\Delta E_{\frac{1}{2}\Gamma\overline{\text{M}}} = E_{\frac{1}{2}\Gamma\overline{\text{M}}}^{\text{uudd}} - E_{\frac{1}{2}\Gamma\overline{\text{M}}}^{1\text{Q}} = 4(2K_1^{\parallel} - B_1^{\parallel} - Y_1^{\parallel}), \quad (4)$$

$$\Delta E_{\frac{3}{4}\Gamma\overline{\text{K}}} = E_{\frac{3}{4}\Gamma\overline{\text{K}}}^{\text{uudd}} - E_{\frac{3}{4}\Gamma\overline{\text{K}}}^{1\text{Q}} = 4(2K_1^{\parallel} - B_1^{\parallel} + Y_1^{\parallel}). \quad (5)$$

Here, $E_{\frac{1}{2}\Gamma\overline{\text{M}}}^{\text{uudd}}$, $E_{\frac{3}{4}\Gamma\overline{\text{K}}}^{\text{uudd}}$, and $E_{\overline{\text{M}}}^{3\text{Q}}$ denote the total energies of the *uudd* states along $\overline{\text{M}}$ - and $\overline{\text{K}}$ -direction, and the 3Q state

[cf. Fig. 1(c)], respectively, whereas $E_{\frac{1}{2}\Gamma\overline{\text{M}}}^{1\text{Q}}$, $E_{\frac{3}{4}\Gamma\overline{\text{K}}}^{1\text{Q}}$, and $E_{\overline{\text{M}}}^{1\text{Q}}$ are the total energies of the corresponding 1Q states.

B. Magnetic bilayers

With the introduction of a second magnetic layer, the distance of two atoms is no longer enough to determine if interactions share the same interaction constant since it becomes important which layer (top or bottom) the two interacting atoms belong to. The four possible interactions top-top, top-bottom, bottom-top, and bottom-bottom each share their own sets J_{σ}^{T} , J_{σ}^{TB} , J_{σ}^{BT} , and J_{σ}^{B} , respectively.

These considerations lead us to the pairwise exchange Hamiltonian of a magnetic bilayer, describing interactions of two magnetic moments $\mathbf{m}_i^{\text{T}}$ and $\mathbf{m}_j^{\text{B}}$ lying in the top (T) or bottom (B) layer:

$$H_{\text{BL}} = - \sum_{i \neq j} J_{ij}^{\text{T}} (\mathbf{m}_i^{\text{T}} \cdot \mathbf{m}_j^{\text{T}}) - \sum_{i \neq j} J_{ij}^{\text{B}} (\mathbf{m}_i^{\text{B}} \cdot \mathbf{m}_j^{\text{B}}) - \sum_{ij} J_{ij}^{\text{TB}} (\mathbf{m}_i^{\text{T}} \cdot \mathbf{m}_j^{\text{B}}) - \sum_{ij} J_{ij}^{\text{BT}} (\mathbf{m}_i^{\text{B}} \cdot \mathbf{m}_j^{\text{T}}). \quad (6)$$

Here, the first two contributions describe the interaction of spins within the individual magnetic layers, i.e., similar to a magnetic ML, and are hence referred to as intralayer exchange, while the latter two denote the interlayer exchange between the top and bottom magnetic layer and vice versa. As the corresponding exchange parameters are symmetric and the dot product of the moments is commutative, i.e., $\mathbf{m}_i^{\text{T}} \cdot \mathbf{m}_j^{\text{B}} = \mathbf{m}_j^{\text{B}} \cdot \mathbf{m}_i^{\text{T}}$, the two terms yield the same value thereby sharing the same exchange constant $J_{\sigma}^{\text{TB}} = J_{\sigma}^{\text{BT}}$ denoted $J_{\sigma}^{\perp}$ in the following. For a symmetric magnetic bilayer, e.g., a freestanding Mn bilayer considered as a reference system in our DFT calculations presented below, both magnetic layers are equivalent. In that case, the exchange constants of top and bottom layer coincide and are denoted for the shells as $J_{\sigma}^{\parallel} = J_{\sigma}^{\text{T}} = J_{\sigma}^{\text{B}}$.

Hence, one can rewrite the Hamiltonian for the symmetric bilayer as

$$H_{\text{s-BL}} = - \sum_{i \neq j, L} J_{ij}^{\parallel} (\mathbf{m}_i^L \cdot \mathbf{m}_j^L) - \sum_{ij, L \neq L'} J_{ij}^{\perp} (\mathbf{m}_i^L \cdot \mathbf{m}_j^{L'}) \quad (7)$$

with the layer indices $L, L' \in \{\text{T}, \text{B}\}$. The pairwise intra- and interlayer Heisenberg exchange constants can be determined separately via DFT calculations by taking different spin spiral setups into account. In case of two magnetic layers, the definition of the direction of a magnetic moment in a spin spiral state as introduced in the previous section needs to be adapted according to

$$\mathbf{m}_i^L = (\cos(\varphi_i^L) \sin(\theta_L), \sin(\varphi_i^L) \sin(\theta_L), \cos(\theta_L))^T \quad (8)$$

for a spin at site i in layer $L \in \{\text{T}, \text{B}\}$ and $\varphi_i^L = \mathbf{q} \cdot \mathbf{R}_i^L$. Only the cone angle θ_L , which describes the canting of the spins with respect to their rotation axis in layer L , varies between the two magnetic layers.

For $\theta_{\text{T}} = 90^\circ$ and $\theta_{\text{B}} = 0^\circ$ [see Fig. 2(a)], the flat spin spiral propagates only in the top magnetic layer while the moments of the bottom layer are aligned perpendicular to the overlying noncollinear spin structure thus making the interlayer

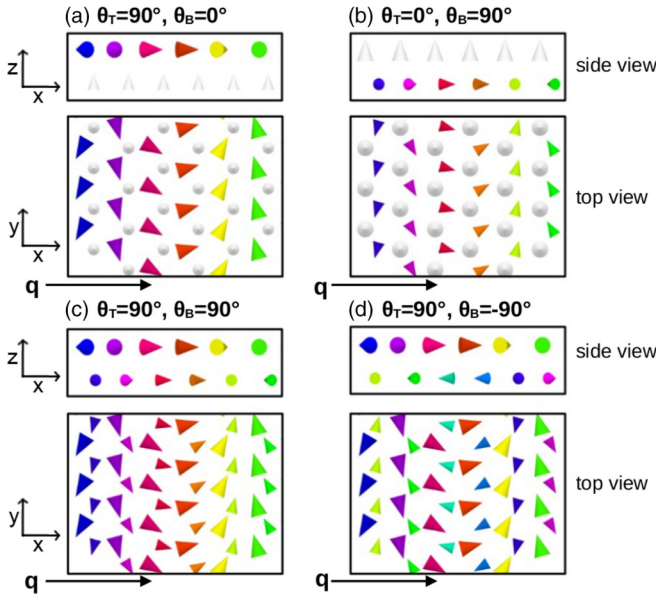


FIG. 2. Spin spiral setups used for the DFT calculation of pairwise Heisenberg intra- and interlayer exchange constants in atomic hexagonally arranged magnetic bilayers. Magnetic moments belonging to the top layer are displayed larger compared to the bottom layer magnetic moments. [(a) and (b)] Spin spiral propagating along the wave vector $\mathbf{q}$ either only in the top or the bottom layer, respectively, while the magnetic moments of the other layer are oriented perpendicular to the rotation plane of the spin spiral. [(c) and (d)] Spin spirals propagating simultaneously in both magnetic layers with parallel and antiparallel alignment of spins between the layers, respectively. For every magnetic configuration the upper (lower) image illustrates its respective side (top) view. The magnetic configurations can be described by Eq. (8) and the angles θ_T and θ_B are given above each panel.

Heisenberg exchange for the whole setup vanish. As the intralayer exchange of the bottom layer remains constant, the only term left is the intralayer exchange of the top magnetic layer. With the same reasoning, the intralayer exchange constants of the bottom layer are obtained by letting the spin spiral propagate solely in this layer [Fig. 2(b)]. These types of DFT calculations can be restricted to $\mathbf{q}$ vectors along the highsymmetry directions of the 2D hexagonal BZ as shown in Fig. 1(a) since the distances between neighboring moments in the individual layers correspond to those of the hexagonal magnetic ML. Consequently, the symmetry zone of spin spirals for the computation of the intralayer Heisenberg exchange is the usual hexagonal 2D BZ.

For the case that a flat spin spiral propagates simultaneously in both magnetic layers, i.e., $\theta_T = 90^\circ$ and $\theta_B = \pm 90^\circ$, two alignments between the spins of the layers need to be taken into account for the bilayer magnetization. In the first case, $\theta_B = 90^\circ$, the same magnetization is present in the two layers, neglecting the phase-shift due to the hexagonal stacking. The second case introduces a spin flip of the bottom layer, through $\theta_B = -90^\circ$.

For spin spirals propagating in two layers, the stacking sequence of the magnetic layers is relevant. As visible from Figs. 2(c) and 2(d), in a magnetic bilayer the atomic positions

of the layers are shifted against each other in the xy plane, due to the hexagonal stacking, resulting in different distances between neighboring interlayer spins compared to the ML (cf. Fig. 11) [33]. With the introduction of the second layer, the rotational symmetry of the hexagonal ML around the z axis in real space is partially broken, but from the atomic positions of both layers a new symmetric lattice (cf. top views in Fig. 2) with a smaller hexagonal cell can be constructed.

In reciprocal space, the symmetry zone of spin spirals corresponding to such an effective honeycomb lattice is extended by a factor of $\sqrt{3}$ into every direction and rotated by an angle of 30° compared to the BZ of the hexagonal lattice of a magnetic ML [cf. Fig. 3(a)]. Due to the extended 2D BZ new highsymmetry points occur which correspond to spin structures of the bilayer and the alignment of spins between them. In addition, the computational effort in DFT increases significantly due to the longer highsymmetry directions $\overline{\Gamma M'}$ and $\overline{\Gamma K' M'}$ containing a larger number of $\mathbf{q}$ values. The spin structures arising in spin spiral calculations at the highsymmetry points are determined by the alignment of the magnetic moments: in the case of $\theta_T = 90^\circ$ and $\theta_B = 90^\circ$ [Fig. 2(c)], the FM state emerges at the center of the symmetry zone [$\overline{\Gamma}$ point, see Fig. 3(a)], whereas it is the layered AFM (LAFM) state for $\theta_T = 90^\circ$ and $\theta_B = -90^\circ$ [Fig. 2(d)].

At the $\overline{M'}$ point at the zone boundary, one obtains two types of RW-AFM states depending on the alignment between the layers. In both magnetic configurations, there is a RW-AFM state within each of the two magnetic layers as for the ML case (cf. Fig. 1). However, there are two possibilities of the net alignment of the moments between the two layers: in the net AFM alignment two of the three NN moments of bottom and top layer are antiparallel [Fig. 4(a)], while two of the three NN moments are parallel for the net FM alignment [Fig. 4(b) and sketches in Fig. 3(a)]. The RW-AFM state with a net AFM coupling between the layers results at the $\overline{M'}$ point for spin spirals with $\theta_T = 90^\circ$ and $\theta_B = 90^\circ$ and the RW-AFM state with a net FM coupling between the layers occurs for $\theta_T = 90^\circ$ and $\theta_B = -90^\circ$. To emphasize the net coupling between the two layers, we denote these magnetic states of the bilayer in the following as the RW-AFM $_{\rightleftharpoons}$ state and the RW-AFM $_{\rightarrow}$ state.

At the $\overline{K'}$ point corresponding to the center of the second periodic image of the hexagonal BZ of a magnetic ML, one recovers a FM state within each layer with moments rotated by 120° or 60° between the layers depending on the spin alignment between the layers [Fig. 3(a)]. Moreover, at the $\frac{2}{3}\overline{\Gamma M'}$ point, corresponding to the $\overline{K}$ point of the BZ for a hexagonal monolayer, the Néel state is still present [not shown in Fig. 3(a)].

The following discussions in our work focus on the two types of RW-AFM states in the bilayer and the corresponding 3Q states (Fig. 4). This motivates us to choose the following notation for spin spiral energy dispersions based on the alignment of the moments between the layers at the $\overline{M'}$ point of the BZ. We denote the energy dispersions of the spin spirals as $E_{\rightleftharpoons}(\mathbf{q})$ [$\theta_T = 90^\circ$, $\theta_B = 90^\circ$, Fig. 2(c)] and $E_{\rightarrow}(\mathbf{q})$ [$\theta_T = 90^\circ$, $\theta_B = -90^\circ$, Fig. 2(d)]. The corresponding states at the $\overline{M'}$ point are the RW-AFM $_{\rightleftharpoons}$ state [Fig. 4(a)] and the RW-AFM $_{\rightarrow}$ state [Fig. 4(b)], respectively, and the energies are

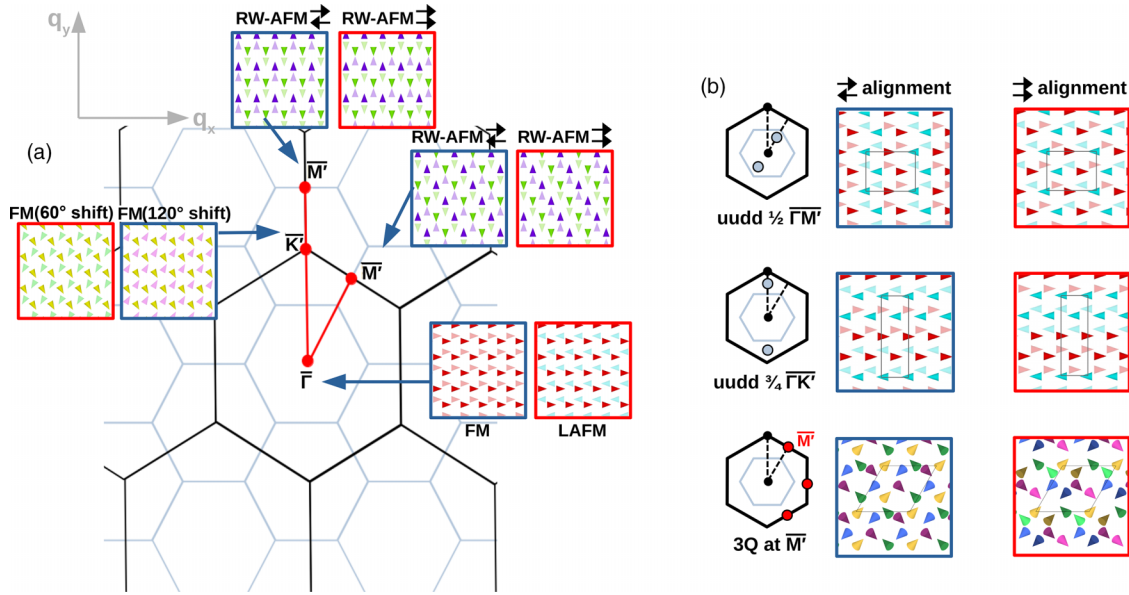


FIG. 3. Sketch of the enlarged 2D hexagonal symmetry zone, a selection of spin spiral states arising at the highsymmetry points and images of the prototypical multi-Q states of a magnetic bilayer. (a) depicts the enlarged 2D hexagonal symmetry zone in reciprocal space which needs to be taken into account for the calculation of spin spirals propagating simultaneously in both magnetic layers. For the highsymmetry points, spin structures are visualized for the case of $\rightleftharpoons$ (blue) and $\Rightarrow$ (red) alignment of the layers at the $\bar{M}'$ -point; bold (faded) colors indicate moments in the top (bottom) layer. The 2D BZ which suffices to treat spin spirals of a magnetic ML is depicted by a faded hexagon. (b) shows the definition and construction of the prototypical multi-Q states of a magnetic BL analogous to the one of a ML; for each coupling type of the layers, two collinear *uudd* states and one noncollinear 3Q state are possible. For each spin structure the respective magnetic unit cell is indicated by a thin black line.

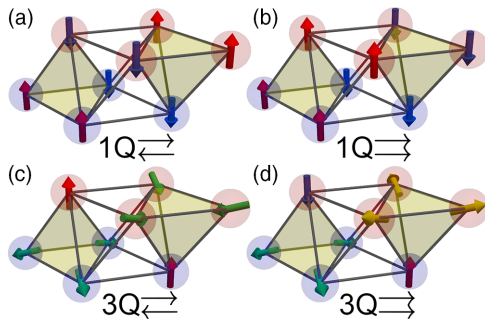


FIG. 4. Illustration of the two types of [(a) and (b)] RW-AFM (1Q) and [(c) and (d)] 3Q states in a magnetic bilayer. In every panel four magnetic atoms are shown in the top (red spheres) and in the bottom layer (blue spheres). Grey lines indicate the NN lattice connections within and between the top and bottom layer. Arrows denote the magnetic moment directions. (a) RW-AFM $_{\rightleftharpoons}$ (1Q $_{\rightleftharpoons}$) state: the layers exhibit a net antiparallel ($\rightleftharpoons$) alignment, i.e., one moment of a yellow tetrahedron lies in the hollow site of three moments of the other layer with one aligned parallel and two aligned antiparallel. (b) RW-AFM $_{\Rightarrow}$ (1Q $_{\Rightarrow}$) state: magnetic moments of the upper layer are flipped with respect to the 1Q $_{\rightleftharpoons}$ state and the layers are effectively aligned ferromagnetically ($\Rightarrow$). (c) 3Q $_{\rightleftharpoons}$ state: magnetic moments form tetrahedron angles within and between the layers and the layers are net antiparallel aligned (see yellow tetrahedrons). Hence we refer to this state as the ideal bilayer triple-Q state. (d) 3Q $_{\Rightarrow}$ state: net parallel alignment of moments between the layers. In this state tetrahedron angles occur only for NN moments within one layer and not between them.

given by $E_{\text{RW-AFM}\rightleftharpoons} = E_{\rightleftharpoons}(\mathbf{q}_{\bar{M}'})$ and $E_{\text{RW-AFM}\Rightarrow} = E_{\Rightarrow}(\mathbf{q}_{\bar{M}'})$ [Fig. 3(a)]. At the $\bar{\Gamma}$ point, on the other hand, we obtain the FM and the LAFM state [Fig. 3(a)] for the $\rightleftharpoons$ and $\Rightarrow$ alignment, respectively, and the energies are given by $E_{\text{FM}} = E_{\rightleftharpoons}(0)$ and $E_{\text{LAFM}} = E_{\Rightarrow}(0)$.

For spin spirals propagating in both magnetic layers, energy contributions from intra- and interlayer pairwise exchange arise, which we denote as $E_{\parallel}(\mathbf{q})$ and $E_{\perp}(\mathbf{q})$, respectively. Based on the atomistic spin model, Eq. (6), the energy contributions can be written as

$$E_{\rightleftharpoons}(\mathbf{q}) = E_{\parallel}(\mathbf{q}) + E_{\perp}(\mathbf{q}), \quad E_{\Rightarrow}(\mathbf{q}) = E_{\parallel}(\mathbf{q}) - E_{\perp}(\mathbf{q}). \quad (9)$$

Hence, by subtracting the DFT energy dispersion $E_{\rightleftharpoons}(\mathbf{q})$ from $E_{\Rightarrow}(\mathbf{q})$ or adding the two dispersions the curves of the interlayer exchange $E_{\perp}(\mathbf{q})$ or intralayer exchange $E_{\parallel}(\mathbf{q})$ can be filtered out.

The enlargement of the magnetic phase space of spin spirals is accompanied by an increase of the number of prototypical multi-Q states. While there are two *uudd* states and one 3Q state possible for a magnetic ML (cf. Sec. II A), twice the number can be constructed for the BL, based on the $\rightleftharpoons$ and $\Rightarrow$ alignment between the layers. Indeed, these states are defined and constructed in analogy to those of the ML; $\mathbf{q} = \pm \frac{1}{2} \bar{\Gamma} \bar{M}'$ and $\mathbf{q} = \pm \frac{3}{4} \bar{\Gamma} \bar{K}'$ represent 90° spin spirals in each layer from which *uudd* states can be formed by superposition. The first mentioned $\mathbf{q}$ value is identical to the one of the *uudd*- $\bar{\Gamma} \bar{K}'$ state of a magnetic ML, whereas the latter is located outside the first BZ [see Fig. 3(b)]. The resulting *uudd* states for both alignment types of the magnetic layers repre-

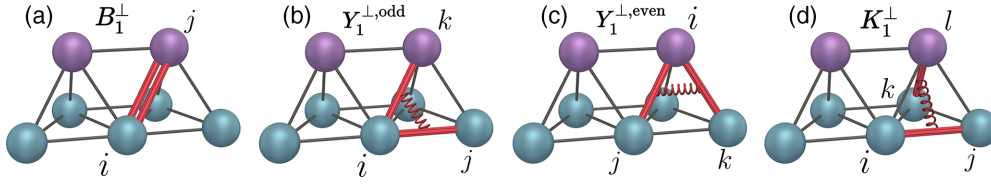


FIG. 5. Illustration of nearest neighbor interlayer higher-order exchange interactions in a hexagonal magnetic bilayer. In every panel only five atoms of the bottom layer (blue spheres) and two atoms of the top layer (purple spheres) are displayed with nearest neighbor atoms connected by thin gray lines. The interacting atoms are marked with an index (i, j, k, l) matching to Eq. (10). (a) Biquadratic exchange interaction between the two layers with constant $B_1^\perp$. (b) Odd type of interlayer four-spin three-site interaction with three of the four spin moments in the two scalar products from the bottom layer and constant $Y_1^{\perp, \text{odd}}$. (c) Even type of interlayer four-spin three-site interaction with evenly distributed spin moments in the scalar products from top and bottom layer and constant $Y_1^{\perp, \text{even}}$. (d) Interlayer four-spin four-site interaction with spin moments on the corners of a tetrahedron and constant $K_1^\perp$. The higher-order interactions are depicted through solid large red tubes, denoting the scalar products between spin moments, which are connected by red springs [cf. Eq. (10)]. For clarity spin moments are not depicted in the sketches. The three types of four-spin interactions are also illustrated in top view sketches in the Appendix (Fig. 12) showing for one reference atom i every pair of interacting moments.

sent a combination of single-layer $uudd$ states with opposite coupling between the layers [see sketches in Fig. 3(b)].

Similarly, we can construct a 3Q state in the bilayer either from the RW-AFM $_{\rightleftharpoons}$ states at the three equivalent $\bar{M}'$ points or from the RW-AFM $_{\rightarrow}$ states [Fig. 3(b)]. From the superposition of the three RW-AFM $_{\rightleftharpoons}$ states we obtain a 3Q state with a net AFM coupling [Fig. 4(c)], while a net FM coupling arises [Fig. 4(d)] upon using the RW-AFM $_{\rightarrow}$ state. We denote these two 3Q states as the 3Q $_{\rightleftharpoons}$ and the 3Q $_{\rightarrow}$ state, respectively [Figs. 4(c) and 4(d)]. In both of these 3Q states, tetrahedron angles occur between adjacent spins within one of the layers. However, only in the 3Q $_{\rightleftharpoons}$ state [Fig. 4(c)] tetrahedron angles are also spanned for nearest neighbor spins of different layers. Therefore we denote this spin structure as the ideal bilayer 3Q state.

Since the multi-Q states of the bilayer are obtained from a superposition of spin spiral (1Q) states as for the monolayer they are also energetically degenerate if we consider only pairwise Heisenberg exchange interactions, H_{BL} [Eq. (6)]. In order to explain the total energy differences found in DFT calculations as reported below, we need to take higher-order interactions into account. Considering HOIs in a magnetic bilayer introduces additional terms to the Hamiltonian. In the nearest neighbor approximation for a symmetric bilayer, the following additional higher-order terms arise:

$$\begin{aligned}
 H_{\text{HOI}}^\perp = & - \sum_{\langle ij \rangle} B_1^\perp (\mathbf{m}_i^L \cdot \mathbf{m}_j^{L'})^2 \\
 & - \sum_{\langle ijk \rangle} Y_1^{\perp, \text{odd}} (\mathbf{m}_i^L \cdot \mathbf{m}_j^L) (\mathbf{m}_i^L \cdot \mathbf{m}_k^{L'}) \\
 & - \sum_{\langle ijk \rangle} Y_1^{\perp, \text{even}} (\mathbf{m}_i^L \cdot \mathbf{m}_j^{L'}) (\mathbf{m}_i^L \cdot \mathbf{m}_k^{L'}) \\
 & - \sum_{\langle ijkl \rangle} K_1^\perp (\mathbf{m}_i^L \cdot \mathbf{m}_j^L) (\mathbf{m}_k^L \cdot \mathbf{m}_l^{L'}). \quad (10)
 \end{aligned}$$

Here the summations are performed over unit cells i, j, k , and l and the superscripts L and L' denote the top and bottom layer ($L, L' \in \{B, T\}$) with $L \neq L'$ in all terms.

Four interlayer HOIs are introduced in Eq. (10) which are schematically depicted in Fig. 5. The first term is the bi-

quadratic exchange interaction between two nearest interlayer neighbors i and j of the top and bottom layer [Fig. 5(a)] with the interaction constant $B_1^\perp$. There are two different types of interlayer four-spin three-site interactions which occur since there are two possibilities to distribute three spin moments on the two scalar products. In the first type, characterized by the interaction constant $Y_1^{\perp, \text{odd}}$, there is a scalar product of the two spin moments of the same layer L and the second scalar product contains one spin of the other layer L' [Fig. 5(b)]. This term exhibits a change of sign if the spin moments in one of the two layers are inverted, i.e., for the operation $\mathbf{m}_i^L$ to $-\mathbf{m}_i^L$. Therefore we denote it as an odd four-spin three-site interaction, due to the odd distribution 3:1 on the layers $L : L'$. The even interlayer four-spin three-site interaction denoted by the HOI constant $Y_1^{\perp, \text{even}}$ is obtained if both scalar products contain the spin moment of the other layer L' [Fig. 5(c)]. Note that the summations are performed over triangles of nearest neighbors within the two layers and all permutations of the indices i, j, k .

Finally, we consider the interlayer four-spin four-site interaction in which the spin moments are distributed on a tetrahedron [Fig. 5(d)]. In this case, all four sites are nearest neighbor atoms. The interlayer four-spin four-site interaction is characterized via the constant $K_1^\perp$ and is odd with respect to inverting the spin moments either in the top or bottom layer.

For simplicity, we consider a symmetric bilayer in our discussion of the interlayer HOI terms. For the Mn bilayer on the Ir(111) surface discussed below based on DFT calculations, additional terms arise since the two Mn layers are not equivalent. Thereby, we would obtain two additional four-spin three-site interaction constants and one additional four-spin four-site interaction constant depending on whether L or L' denote the top or bottom layer in Eq. (10). While it is straightforward to generalize the spin model in this way it is not required below for the discussion of the intra- vs. interlayer HOI contributions to the DFT total energies.

The presented interlayer HOIs can be derived from a multiband Hubbard model with spin ≥ 1 using fourth-order perturbations [8]. Our bilayer case would change the geometry of the model through new atomic sites featuring different hopping and onsite parameters (t, U). Introducing one atom of a

TABLE I. Relaxed structural parameters for a Mn monolayer in fcc and hcp stacking on the Ir(111) surface in the RW-AFM state. Structural optimizations of the interlayer distances have been carried out using the VASP code and a symmetric slab comprising nine Ir substrate layers and a Mn ML on each side. All distances between the Mn layer and the three topmost Ir layers (Ir1 to Ir3) and between Ir layers in bulk are given in angstroms.

System	$d_{\text{Mn-Ir1}}$	$d_{\text{Ir1-Ir2}}$	$d_{\text{Ir2-Ir3}}$	$d_{\text{Ir,bulk}}$
fcc-Mn/Ir(111)	2.076	2.282	2.213	2.25
hcp-Mn/Ir(111)	2.058	2.263	2.221	2.25

second species into the four-spin three-site energy of a triangle of spins, Eq. (36) of Ref. [8], would split the three terms up into the different interlayer four-spin three-site interactions ($Y^{\perp,\text{odd}}$, $Y^{\perp,\text{even}}$).

III. COMPUTATIONAL METHODS

In order to investigate the magnetic exchange interactions of a freestanding Mn BL as well as Mn MLs and BLs on the Ir(111) surface, we resort to first-principles calculations based on DFT. Similar to previous work on ultrathin transition metal films on surfaces [19,23], we make use of a combination of the full-potential linearized augmented plane wave (FLAPW) method implemented in the FLEUR code [34,35] and the projector augmented wave (PAW) method incorporated in the VASP code [36–39]. While all calculations on the freestanding Mn bilayer rely on the in-plane lattice constant of Ir determined within the local density approximation (LDA) of the exchange correlation functional taking a value of 2.70 Å [40], the in-plane lattice constant of Ir obtained from the generalized gradient approximation (GGA) with a value of 2.75 Å [41] serves as a basis for Mn MLs and BLs on the Ir(111) substrate.

In a first step, geometry optimizations for both fcc and hcp stacking of the Mn ML as well as the Mn BLs on Ir(111) have been performed via the VASP code. For Mn BLs, in accordance with experimental observations [24,27], only an hcp stacking of the subsurface Mn layer is considered, whereas for the top magnetic layer both stacking sequences are possible. Here we have considered only pseudomorphic stackings of Mn MLs or BLs on Ir(111). For structural relaxations, we have used symmetric films comprising nine Ir layers and Mn ML or Mn BL on both sides of the film. The initial interlayer distance, 2.25 Å, was set using the equilibrium lattice parameter of the Ir bulk obtained in DFT within GGA. Only the z coordinates of the Mn ML (BL) as well as the first two Ir layers were allowed to relax, while the five central Ir layers were kept fixed to the bulk value. The relaxed structural parameters are tabulated in Table I and in Table II for Mn ML on Ir(111) and Mn BL on Ir(111), respectively. The two-dimensional (2D) Brillouin zone (BZ) of both magnetic unit cells was sampled by a $15 \times 15 \times 1$ Γ -centered k mesh. We have used a 400 eV energy for the plane wave basis set cutoff. The force tolerance for the structural minimization were used as 0.01 eV/Å. We calculate the force by using the Hellmann-Feynman theorem [42]. For the freestanding Mn bilayer no structural optimization has been carried out; instead, the interlayer distance,

TABLE II. Relaxed structural parameters for Mn bilayers in fcc-hcp and hcp-hcp stacking on the Ir(111) surface in the RW-AFM $_{\pm}$ state. Structural optimizations of the interlayer distances have been carried out using the VASP code and a symmetric slab comprising nine Ir substrate layers and a Mn BL on each side. All distances between the two Mn layers of the BL (Mn1 and Mn2) and the three topmost Ir layers (Ir1 to Ir3) are given in angstroms.

System	$d_{\text{Mn1-Mn2}}$	$d_{\text{Mn2-Ir1}}$	$d_{\text{Ir1-Ir2}}$	$d_{\text{Ir2-Ir3}}$
fcc-Mn/hcp-Mn/Ir(111)	2.092	2.053	2.259	2.220
hcp-Mn/hcp-Mn/Ir(111)	2.060	2.043	2.265	2.222

2.25 Å, was set using the equilibrium lattice parameter of the Ir bulk obtained in DFT within GGA.

Employing the geometry of the structural relaxation, we perform subsequent self-consistent calculations of the energy dispersion $E(\mathbf{q})$ of flat spin spirals for the freestanding Mn BL, the Mn ML and BL on Ir(111) using the FLEUR code. For the Mn BL on the Ir substrate, we restrict ourselves to the calculation of the electronic and magnetic properties of just one stacking of the top Mn layer—the hcp sequence—since for this system the energetically lowest spin structure after geometry optimization—the RW-AFM $_{\pm}$ state (see also Ref. [27])—turns out to be lower in energy than for an fcc stacking of the upper Mn layer. Due to symmetry, no significant change of the energy dispersion is expected upon choosing the fcc stacking sequence for the top Mn layer. These spin spiral calculations can be performed within the chemical unit cell by exploiting the generalized Bloch theorem [35]. Exchange correlation effects are included in LDA by using the Vosko, Wilk, and Nusair (VWN) functional [43]. Note that our approach of using GGA for structural relaxations and LDA for the total energy calculations of various collinear and non-collinear spin states has previously allowed to explain many complex magnetic structures observed in 3d-transition-metal films on 4d and 5d transition-metal surfaces [10,12,13,15–17,44].

While for the Mn ML on Ir(111) the computation of just one set of flat spin spirals with $\mathbf{q}$ vectors chosen along the two highsymmetry directions of the 2D hexagonal BZ is sufficient for each stacking (see Sec. II A), the situation turns out to be computationally much more demanding for the magnetic BL systems. As described in Sec. II B, four sets of calculations are necessary for both the freestanding Mn bilayer and the hcp-Mn/hcp-Mn/Ir(111) film system in order to extract the relevant intra- and interlayer Heisenberg exchange constants.

For these types of calculations, asymmetric films comprising nine Ir layers and one Mn ML (BL) on one side of the film are used. The muffin tin radii are chosen as 2.30 a.u. for the Mn atoms, whereas a slightly larger value of 2.31 a.u. is used for the nonmagnetic Ir atoms. To obtain well-converged total energies we have used a high number of k points, which amounts to 1936 (2025) in the full 2D BZ and the cutoff for the basis functions is taken as $k_{\text{max}} = 4.1$ a.u. $^{-1}$ (4.3 a.u. $^{-1}$) for Mn/Ir(111) and hcp-Mn/hcp-Mn/Ir(111) (freestanding Mn bilayer). Starting from the self-consistent results for spin spirals, the energy contributions of the DMI for the film systems are calculated by including SOC in first-order perturbation theory [45]. The magnetocrystalline anisotropy energy

(MAE) is computed self-consistently [46] via the FLEUR code as well by adding SOC to the RW-AFM state in the two-atomic unit cell. To obtain accurate results for this small quantity, the cutoff for the basis functions is further increased to $k_{\max} = 4.3 \text{ a.u.}^{-1}$ and the number of k -points to 8024 in the full 2D BZ.

For the Mn BL on Ir(111), we obtain the total energies of all considered multi-Q superposition states using the FLEUR code. We used the same relaxed interlayer distances, exchange-correlation potential, cutoff for the basis functions and muffin tin radii as for the spin spiral energy dispersion to determine the energies of the $uudd$ states with respect to the $\rightleftharpoons$ or $\Rightarrow$ alignment within their four-atomic unit cells per layer. Only the number of k points needs to be adapted since the size of the BZ in reciprocal space changes: for the multi-Q states, i.e., $uudd$ states along $\overline{\Gamma\text{M}}$ ($\overline{\Gamma\text{K}}$) and $\overline{\Gamma\text{K}}$ ($\overline{\Gamma\text{M}}$) direction, the 3Q state and the reference 1Q state (RW-AFM), we chose 288 k points in the irreducible part of the 2D BZ. Due to the smaller 2D BZ, this number of k points is equivalent to 2304 k points in spin spiral calculations. The total energies for the $uudd$ states for the Mn ML on Ir(111) were also obtained using the FLEUR code with this setup.

For the self-consistent calculation of the total energies of the noncollinear 3Q state of the freestanding Mn BL and the Mn ML on Ir(111) we resort to the VASP code using the same relaxed interlayer distances as in the associated FLEUR calculations, the LDA VWN potential [43] and the constrained magnetic moment approach. The energy cutoff is set to 400 eV in all cases and the 2D BZ is sampled by a $15 \times 15 \times 1$ Monkhorst-Pack (MP) k -point mesh. The total energies of the two $uudd$ states of the freestanding Mn BL are also obtained via VASP; a $22 \times 28 \times 1$ MP k -point mesh is applied for the multi-Q state along $\overline{\Gamma\text{M}}$ direction and a $14 \times 44 \times 1$ MP k -point mesh for the respective state along $\overline{\Gamma\text{K}}$.

IV. RESULTS

A. Mn monolayers on Ir(111)

Before turning to the more complex topic of magnetic bilayers, we first focus on the magnetic properties of Mn MLs on the Ir(111) surface in this section. Figure 6(a) displays the energy dispersion $E(\mathbf{q})$ of flat spin spirals in the scalar-relativistic approximation, i.e., neglecting the effect of SOC, for both fcc and hcp stacking of the Mn ML. While filled circles represent total energies calculated via DFT, the solid lines depict a fit to the model of pairwise Heisenberg exchange including shells up to 8th nearest neighbors. One immediately notices that $E(\mathbf{q})$ for both stackings shows the same qualitative behavior with the RW-AFM state at the BZ boundary ($\overline{\text{M}}$ point) having a much lower energy (on the order of 290 to 330 meV/Mn atom) than the FM state at the $\overline{\Gamma}$ point. At the center of the BZ the magnetic moment of Mn in fcc stacking on Ir(111) (fcc-Mn) amounts to $3.35 \mu_B$, the respective value of hcp-Mn is found to be given by $3.26 \mu_B$. In both cases the magnitude changes by only 2% upon varying $\mathbf{q}$ along the high-symmetry directions of the 2D BZ.

As seen from Fig. 6(a), for both Mn ML stackings on Ir(111) the Néel state at the $\overline{\text{K}}$ point represents the state of

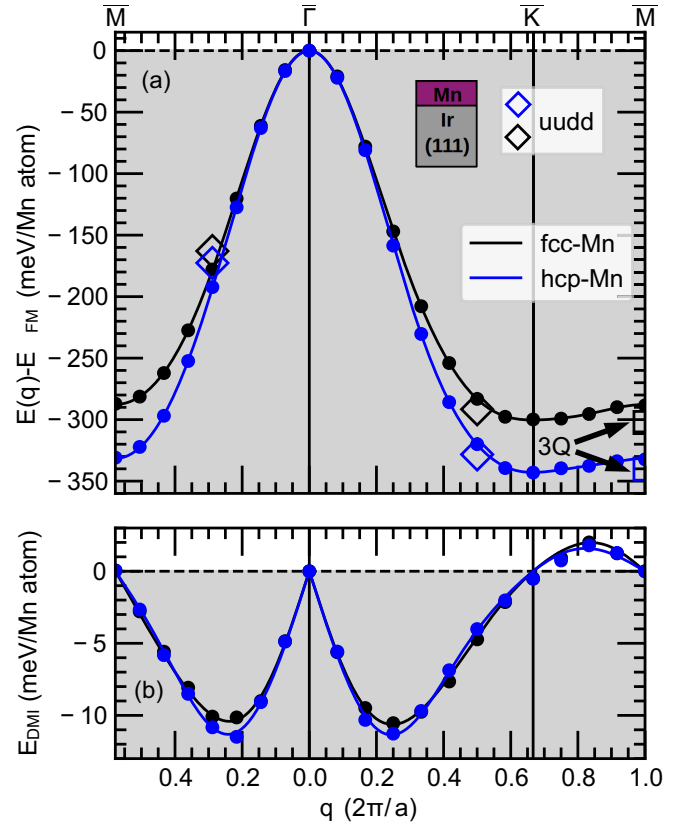


FIG. 6. DFT calculated energy dispersion $E(\mathbf{q})$ of flat spin spirals for fcc- (black symbols) and hcp-stacked (blue symbols) Mn monolayers (MLs) on Ir(111). (a) Filled circles depict scalar-relativistic total DFT energies, while solid lines represent a fit to the model of intralayer pairwise Heisenberg exchange beyond nearest neighbors. The energies of the $uudd$ states and the 3Q state are depicted as empty diamonds and squares at the absolute values of $\mathbf{q}$ of the constituting spin spiral states, respectively. (b) Total DMI contributions for cycloidal spin spirals of the two Mn stackings. The solid lines denote a fit to the Dzyaloshinskii-Moriya interaction. $E_{\text{DMI}} < 0$ (> 0) indicates the preference of (counter)clockwise rotation of cycloidal spin spirals.

lowest energy in the phase space of spin spirals. This can be understood by extracting the pairwise intralayer Heisenberg exchange constants from the fit of the energy dispersion (see Table III): in both systems, $J_1^{\parallel}$ turns out to be the dominant exchange parameter mediating a strong antiferromagnetic exchange coupling between nearest neighbor (NN) spins. $J_2^{\parallel}$ is of

TABLE III. Heisenberg exchange constants for first to third nearest neighbors of fcc- and hcp-stacked Mn MLs on Ir(111), $J_1^{\parallel}$ to $J_3^{\parallel}$, as extracted from fitting the respective spin spiral DFT energy dispersion $E(\mathbf{q})$, i.e., neglecting higher-order terms, and higher-order exchange constants $B_1^{\parallel}$, $K_1^{\parallel}$, and $Y_1^{\parallel}$ calculated from Eqs. (3)–(5). All values are given in meV/Mn atom.

System	$J_1^{\parallel}$	$J_2^{\parallel}$	$J_3^{\parallel}$	$B_1^{\parallel}$	$K_1^{\parallel}$	$Y_1^{\parallel}$
fcc-Mn/Ir(111)	-31.22	-4.29	-1.72	-3.19	-1.22	-3.02
hcp-Mn/Ir(111)	-36.78	-4.47	-1.02	-3.19	-0.89	-3.50

negative sign as well with a ratio of $0.14J_1^\parallel$ in case of fcc-Mn and $0.12J_1^\parallel$ for hcp-Mn. As shown in Ref. [47], under the condition that either only $J_1^\parallel$ is present in an antiferromagnetic material or $J_2^\parallel$ prefers AFM coupling as well with a maximum strength of $\frac{1}{8}J_1^\parallel$, the Néel state emerges as the ground state due to geometric frustration of the exchange interactions on the triangular lattice. For fcc-Mn/Ir(111), the value of $J_2^\parallel$ exceeds the limit of $1/8=0.125$, thus one needs to take into account the contribution of third NN exchange $J_3^\parallel$. The condition then becomes $J_2^\parallel > 9/8J_3^\parallel + 1/8J_1^\parallel$ [9], which is fulfilled by the exchange constants extracted for fcc-Mn.

The situation is different for the closely related system of a Mn ML on the Re(0001) surface [10]. Although in this case the Mn MLs are characterized by strong AFM intralayer exchange between nearest neighbors as well, the energy minimum in the spin spiral phase space turns out to be the RW-AFM state due to exchange frustration of $J_1^\parallel$ and $J_2^\parallel$ having a large ratio of 0.23 up to 0.32 for the two Mn stackings.

Figure 6(b) further shows the total DMI contributions for every cycloidal spin spiral state calculated along the high-symmetry directions of the 2D hexagonal BZ for the Mn/Ir(111) film system. Just like the scalar-relativistically computed DFT energies, they appear rather similar favoring a clockwise rotational sense of cycloidal spin spirals. Although the DMI parameter D_1 for nearest neighbors takes significant values of around 2.8 meV/Mn atom (see Table VIII), the effect of SOC becomes only relevant in the vicinity of the $\bar{\Gamma}$ point where the slope of the dispersion is steep. As expected, the DMI shows (nearly) no influence on the energetically close Néel and RW-AFM states at the $\bar{K}$ and $\bar{M}$ points, respectively.

Focusing on the last aspect of our analysis on Mn MLs on Ir(111), the prototypical multi-Q states, we notice that the total energies of the two collinear *uudd* states and of the non-collinear 3Q state differ significantly from their corresponding single-Q (spin spiral) states [see Fig. 6(a)]. As a result, we obtain large values for the intralayer HOI constants according to Eqs. (3) to (5) (see Table III); while the biquadratic term $B_1^\parallel$ and the four-spin three-site interaction strength $Y_1^\parallel$ are on the order of 3 meV/Mn atom, notably smaller values of about 1 meV/Mn atom result for the four-spin four-site term $K_1^\parallel$.

Similar to the HOI constants calculated for Mn/Re(0001) [10], all of them exhibit a negative sign. From Fig. 6(a), it becomes obvious that these terms actually favor the 3Q superposition state over its constituting 1Q spin spiral state, the RW-AFM state, for both stackings of the Mn layer. For fcc-Mn/Ir(111) the 3Q state gains 2.5 meV per Mn atom compared to the energetically lowest spin spiral state, i.e., the Néel state, making it the lowest energy state. For hcp-Mn/Ir(111), in contrast, the 3Q state remains by 3 meV per Mn atom higher in energy than the Néel state consistent with the experimental observation of the Néel state in this ultrathin film system [24].

B. Mn bilayers on Ir(111)

1. Intralayer exchange

Figures 7(a) and 7(b) show the energy dispersion $E(\mathbf{q})$ neglecting SOC of flat spin spirals propagating only in

one magnetic layer of a freestanding Mn bilayer and the Mn/Mn/Ir(111) film system. Similar to Mn MLs on Ir(111), the two BL systems overall display a typical AFM coupling within the layers, with the RW-AFM state at the $\bar{M}$ -point being energetically favored over the FM state at the center of the BZ by 130 to 230 meV/magnetic unit cell (u.c.), i.e., per two Mn atoms. However, allowing the Mn bilayer to grow pseudomorphically on the Ir(111) surface has a drastic impact on the spin spiral state of lowest energy due to hybridization of Mn states with the nonmagnetic substrate.

For the freestanding Mn bilayer the global minimum of the energy dispersion is located at $q = |\mathbf{q}| \approx 0.51 \times 2\pi/a$ along $\bar{\Gamma M}$ direction and hence only by 2 meV lower in energy than the RW-AFM state [Fig. 7(a)]. Note that for this system the calculation of one spin spiral setup and one set of intralayer Heisenberg exchange constants (top or bottom magnetic layer) is sufficient since the two Mn layers are symmetry equivalent due to the vacuum on both sides. As revealed in Table IV, the Mn bilayer is characterized by strong intralayer exchange frustration of AFM interactions between nearest and next-nearest neighbor spins.

In the case of the Mn BL on Ir(111), spin spirals in the top magnetic layer [red symbols and curve in Fig. 7(b)] exhibit a similar energy dispersion as for the freestanding bilayer [cf. Fig. 7(a)]. The dispersion clearly shows the RW-AFM state as the 1Q state of lowest energy. However, spin spirals propagating only in the bottom Mn layer [blue symbols and curve in Fig. 7(b)], which is in direct contact with the Ir substrate, possesses a 120° Néel ground state similar to the case of the Mn ML on Ir(111) [cf. Fig. 6(a)]. Again, this observation can be attributed to the ratio of the extracted intralayer Heisenberg exchange constants (see Table IV): the respective value between the AFM nearest and next-nearest neighbor constant $J_1^\parallel$ and $J_2^\parallel$, respectively, amounts to roughly $\frac{1}{4}$ for the top Mn layer thereby causing the formation of a RW-AFM ground state due to exchange frustration. The AFM coupling constant $J_1^\parallel$ turns out to be the dominant Heisenberg exchange parameter for the bottom Mn layer resulting in the emergence of the Néel state owing to geometric frustration on the hexagonal lattice, $|J_2^\parallel| < 1/8|J_1^\parallel|$.

Figure 13(a) in Appendix C further displays the energy contributions of the intralayer DMI computed for spin spiral states of the Mn/Mn/Ir(111) film system. As for the Mn ML on Ir(111), the preferred rotational sense of cycloidal spin spirals is clockwise for both Mn layers. As expected, the subsurface Mn layer experiences a larger impact of SOC compared to the top Mn layer which is well reflected in the DMI constant $D_1^\parallel$ for nearest neighbors (see Table IV): with about 1.8 meV/Mn atom it is by a factor of about 6 larger than for the top layer.

Figures 7(c)–7(f) display another interesting aspect of Mn bilayer systems: the variation of the magnetic moments of the individual layers with the spin spiral vector $\mathbf{q}$. While the values for the freestanding Mn BL are on the order of $3.30 \mu_B$ and vary only little with $\mathbf{q}$ [Figs. 7(c) and 7(e)], a different situation emerges for the Mn/Mn/Ir(111) film system. Here, for the case of a flat spin spiral propagating only in the top Mn layer [Fig. 7(f)], its moments are quite constant upon the variation of $\mathbf{q}$ taking a maximum deviation of 3% from the

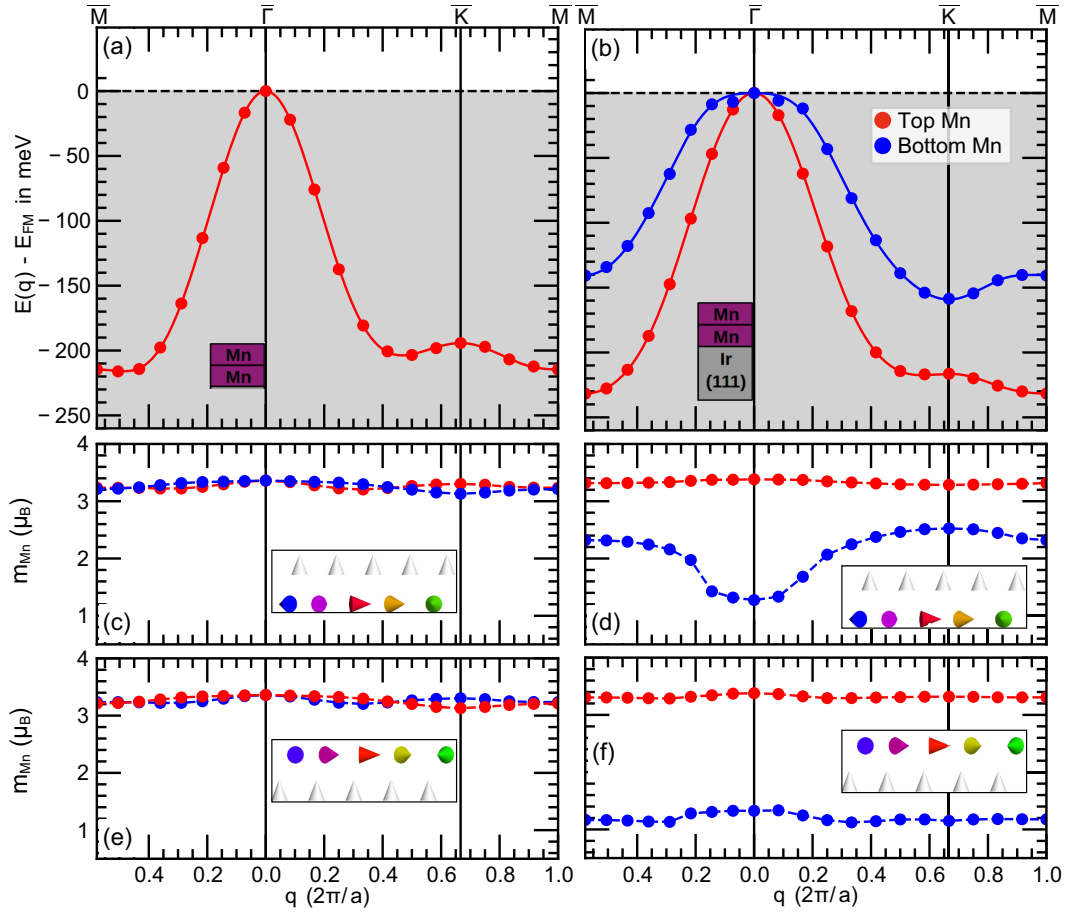


FIG. 7. DFT calculated energy dispersion $E(\mathbf{q})$ of a flat spin spiral propagating in (a) one of the equivalent magnetic layers of a freestanding Mn bilayer and (b) either in the top or in the bottom magnetic layer of a Mn BL on Ir(111). The energy differences are given in meV per two Mn atoms. Total DFT energies in scalar-relativistic approximation, i.e., neglecting SOC, are depicted by filled circles whereas solid lines denote fits to the model of intralayer pairwise Heisenberg exchange including up to eight nearest neighbor shells (see Table IV). [(c)–(f)] Magnetic moments of the Mn layers obtained for every $\mathbf{q}$ value calculated along the highsymmetry lines of the 2D hexagonal BZ shown in (a) and (b). Note that (d) show Mn moments for a flat spin spiral propagating in the bottom (interface) magnetic layer of hcp-Mn/hcp-Mn/Ir(111) and (f) show Mn moments for a flat spin spiral propagating in the top (surface) magnetic layer of hcp-Mn/hcp-Mn/Ir(111) are displayed. For the free-standing Mn bilayer [(c) and (e)], the two layers are equivalent. Dashed lines between the DFT data points serve as a guide to the eye.

TABLE IV. Intralayer Heisenberg exchange constants ($J_i^{\parallel}$) and Dzyaloshinskii-Moriya interaction (DMI) constants ($D_i^{\parallel}$) calculated via DFT for a freestanding Mn bilayer and the hcp-Mn/hcp-Mn/Ir(111) film system. The parameters have been determined individually for each Mn layer via a separate energy dispersion $E(\mathbf{q})$ as indicated in Figs. 7(a), 7(b), and 13(a) (Appendix C); the respective Mn layer is marked in bold letters. Note that in case of the freestanding Mn layer the calculation of the energy dispersion in one Mn layer suffices due to symmetry reasons and DMI contributions do not occur. Possible intralayer HOI terms are not taken into account here. $J_i^{\parallel} > 0$ ($J_i^{\parallel} < 0$) denotes ferromagnetic (antiferromagnetic) coupling and $D_i^{\parallel} > 0$ ($D_i^{\parallel} < 0$) the preference of a clockwise (counterclockwise) rotation of cycloidal spin spirals. All values are given in meV/atom of the respective layer.

System	$J_1^{\parallel}$	$J_2^{\parallel}$	$J_3^{\parallel}$	$J_4^{\parallel}$	$J_5^{\parallel}$	$J_6^{\parallel}$	$J_7^{\parallel}$	$J_8^{\parallel}$
Mn/Mn	−18.73	−8.17	−2.53	−0.19	0.34	−0.23	0.07	−0.04
hcp-Mn /hcp-Mn/Ir(111)	−23.37	−5.80	−0.74	−0.13	0.17	−0.13	0.15	0.03
hcp-Mn/ hcp-Mn /Ir(111)	−18.91	0.19	0.46	0.39	0.52	0.25	−0.06	0.11
	$D_1^{\parallel}$	$D_2^{\parallel}$	$D_3^{\parallel}$	$D_4^{\parallel}$	$D_5^{\parallel}$			
hcp-Mn /hcp-Mn/Ir(111)	0.33	0.22	0.11	0.07	−0.06			
hcp-Mn/ hcp-Mn /Ir(111)	1.82	−0.23	0.12	−0.10	−0.26			

TABLE V. Interlayer Heisenberg exchange constants ($J_i^\perp$) calculated via DFT for a freestanding Mn bilayer and the hcp-Mn/hcp-Mn/Ir(111) film system. The parameters have been determined from the energy dispersions shown in Figs. 8(c) and 8(d). $J_i^\perp > 0$ ($J_i^\perp < 0$) denotes ferromagnetic (antiferromagnetic) coupling between the moments of the two magnetic layers. All values are given in meV/magnetic u.c., i.e., per two Mn atoms.

System	$J_1^\perp$	$J_2^\perp$	$J_3^\perp$	$J_4^\perp$	$J_5^\perp$	$J_6^\perp$	$J_7^\perp$	$J_8^\perp$
Mn/Mn	4.58	-6.20	-4.39	1.22	1.16	0.39	-0.27	-0.34
hcp-Mn/hcp-Mn/Ir(111)	-19.65	-3.57	-2.93	-0.27	0.41	0.26	0.06	-0.23

value at the $\bar{\Gamma}$ point. On the other hand, the Mn moments of the bottom layer which are always oriented perpendicular to the rotation plane of the overlying spin spiral are strongly reduced to $1.33 \mu_B$ with minimum values of $1.12 \mu_B$ being taken for $\mathbf{q}$ vectors along $\bar{\Gamma}\bar{M}$ [Fig. 7(f)].

If the flat spin spiral propagates only in the bottom Mn layer [Fig. 7(d)], the moments in the top magnetic layer are again quite stable. Surprisingly, for the spin spiral in the bottom Mn layer an increase of up to 98% from the value of the $\bar{\Gamma}$ point occur: while the minimum of $1.27 \mu_B$ is given exactly at the FM state, the Néel state at the $\bar{K}$ point holds the maximum of $2.52 \mu_B$ ([Fig. 7(d)]). This behavior can be attributed to the energy gain upon the formation of a magnetic moment for the ground state of the system. Note, that variations in the magnitude of the magnetic moments are mapped effectively into the interaction constants of the classical Heisenberg model. Therefore the spin model accounts for the fluctuations of the magnetic moment in the bottom Mn layer of the present system, resulting in a good fit of the energy dispersion in Fig. 7(b). While the spin fluctuations could limit the general applicability of the spin model, e.g., in spin dynamics simulations, a good description is achieved for spin configurations close to the spin spiral energy minimum. Large variations of the magnetic moment for spin spirals along the highsymmetry lines of the 2D BZ have also been reported for the bottom layer of a Mn BL on the W(110) surface [48].

2. Interlayer exchange

The energy dispersions of flat spin spirals propagating only in one of the magnetic layers as presented in the previous section are perfectly suited to obtain the intralayer exchange constants and thereby provide valuable insight into the magnetism of the bilayer. However, the interlayer exchange contributions are missing and these DFT results cannot predict the possible ground state of our Mn bilayer on Ir(111). Therefore additional calculations for spin spirals propagating simultaneously in both layers are required. Figures 8(a) and 8(b) illustrate a comparison of the DFT data of the just mentioned spin spiral setups for the freestanding Mn bilayer and the Mn BL on Ir(111). Total DFT energies for a $\rightleftharpoons$ ($\Rightarrow$) alignment are denoted by blue (red) circles and solid lines of the respective color indicate fits to the model of pairwise Heisenberg exchange including both intra- and interlayer exchange. Note that these lines just serve as a guide to the eye and that the exchange parameters of both systems listed in Tables IV and V are determined separately from Figs. 7(a), 7(b) and 8(c), 8(d) [49]. The energy dispersion of the $\rightleftharpoons$ aligned layers, $E_{\rightleftharpoons}(\mathbf{q})$, is shifted in both plots with respect to the energy at the $\bar{\Gamma}$ point in the $\Rightarrow$ alignment,

$E_{\rightleftharpoons}(\mathbf{q} = 0)$, by the difference $\Delta E = E_{\text{LAFM}} - E_{\text{FM}}$ obtained from the respective collinear calculations.

From Figs. 8(a) and 8(b), one clearly sees that the general trend of the energy dispersion of the freestanding Mn bilayer is preserved upon placing it on the Ir(111) surface. $\Delta E = E_{\text{LAFM}} - E_{\text{FM}}$ takes a large negative value of about -90 meV/magnetic u.c. in case of the freestanding Mn bilayer and about -180 meV/magnetic u.c. for the ultrathin film system, showing a strong AFM tendency in both systems consistent with previous observations on the intralayer spin spiral dispersion. The FM state with the same orientation of the magnetic moments between the two layers [see Fig. 3(a)] arising at the $\bar{\Gamma}$ point [$E_{\text{FM}} = E_{\rightleftharpoons}(0)$] represents the global energy maximum of the enlarged hexagonal symmetry zone in both cases. Another local energy maximum can be found at the highsymmetry $\bar{K}'$ point, which corresponds to the $\bar{\Gamma}$ point of the second periodic image of the original hexagonal BZ [see Fig. 3(a)], corresponding to a FM state within each layer with moments rotated by 120° or 60° between the layers depending on the spin alignment between the two layers.

However, the global and local energy minima of the spin spiral dispersion in the enlarged hexagonal symmetry zone turn out to be different for the two systems mainly due to hybridization between the magnetic bilayer and the nonmagnetic Ir substrate. As visible from Fig. 8(a), the state of lowest energy for the freestanding Mn bilayer is a 135° spin spiral in the $\rightleftharpoons$ aligned layers at $|\mathbf{q}| \approx 0.43 \times 2\pi/a$ along $\bar{\Gamma}\bar{K}'$ (blue curve) and another local minimum is given by the RW-AFM $\rightleftharpoons$ state at the boundary of the enlarged symmetry zone ($\bar{M}'$ point, red curve). On the other hand, for the Mn BL on Ir(111) the inclusion of DMI [see Fig. 13(b) in the Appendix] does not alter the computed energetically lowest 1Q state, which is the RW-AFM $\rightleftharpoons$ state ($\bar{M}'$ point, blue curve) characterized by a net AFM coupling between the magnetic moments of the two layers [see Fig. 8(b)] [50].

Figures 8(c) and 8(d) show the dispersion of the interlayer Heisenberg exchange calculated according to $E_\perp(\mathbf{q}) = \frac{1}{2}(E_{\rightleftharpoons}(\mathbf{q}) - E_{\Rightarrow}(\mathbf{q}))$ for the two systems under discussion. Here, the subtle differences from the energy dispersion of $\rightleftharpoons$ and $\Rightarrow$ aligned layers between the simple freestanding Mn bilayer and the more complex film system as mentioned above become more apparent. The sign of $E_\perp(\mathbf{q})$ gives the tendency of aligning the layers to each other, i.e., in the $\Rightarrow$ ($\rightleftharpoons$) alignment for a positive (negative) contribution of $E_\perp(\mathbf{q})$. For the freestanding Mn bilayer [Fig. 8(c)], two deep energy minima arise along the two highsymmetry directions of the enlarged hexagonal symmetry zone with another shallow local minimum showing up in the vicinity of the $\bar{K}'$ point. The interlayer exchange energy of the Mn BL on Ir(111) [Fig. 8(d)]

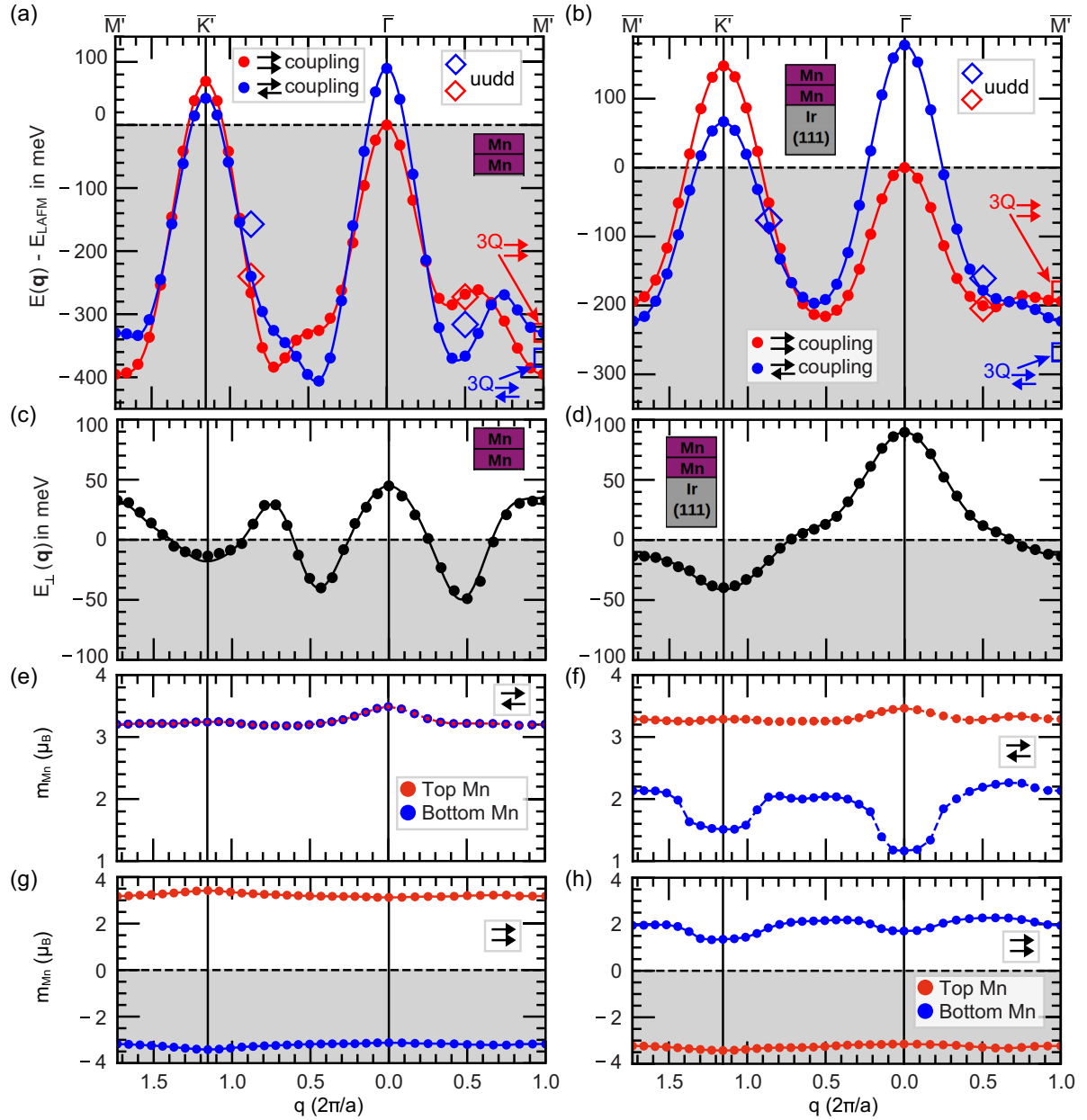


FIG. 8. Energy dispersion $E(\mathbf{q})$ of flat spin spirals propagating in both magnetic layers of (a) a freestanding Mn bilayer and (b) the hcp-Mn/hcp-Mn/Ir(111) film system given in meV per two Mn atoms with respect to the LAFM state. Both $\rightleftharpoons$ (blue) and $\Rightarrow$ (red) alignment between the layers at the $\bar{M}'$ point are taken into account. Total scalar-relativistic DFT energies are marked by filled circles of the respective color whereas solid lines denote fits to the Heisenberg model including intra- and interlayer exchange up to eight nearest neighbor shells. Energies of *uudd* states and the 3Q states are depicted as empty diamonds and squares at the $\mathbf{q}$ values of their constituting spin spiral states for both alignments. Note that the DMI contributions to spin spirals in the Mn BL on Ir(111) are given in the Appendix [see Fig. 13(b)]. [(c) and (d)] Dispersion of the interlayer exchange calculated according to $E_{\perp}(\mathbf{q}) = \frac{1}{2}(E_{\rightleftharpoons}(\mathbf{q}) - E_{\Rightarrow}(\mathbf{q}))$ for the freestanding Mn bilayer and the hcp-Mn/hcp-Mn/Ir(111) film system, respectively. [(e)–(h)] Magnetic moments of the Mn layers for both spin alignments ($\rightleftharpoons$ and $\Rightarrow$) along the highsymmetry lines of the enlarged hexagonal BZ. Note that positive and negative values of the magnetic moments refer to the alignment at the $\bar{\Gamma}$ point, i.e., FM and LAFM state for $\rightleftharpoons$ and $\Rightarrow$ alignments, respectively.

is varying less, no local minima are observed, and the global minimum forms at the $\bar{K}'$ point.

This different behavior can be explained with the interlayer Heisenberg exchange constants obtained from a fit to the energy dispersions (see Table V). Surprisingly, for the freestanding Mn bilayer $J_1^{\perp}$ mediates a FM coupling between neighboring spins of the two magnetic layers com-

peting with AFM interactions of second and third nearest neighbors, $J_2^{\perp}$ and $J_3^{\perp}$, respectively, which are also on the same order of magnitude thereby revealing a strong interlayer exchange frustration. For the Mn BL on the Ir(111) surface, however, the AFM interlayer exchange constant $J_1^{\perp}$ with a value of ≈ -20 meV/magnetic u.c. is the dominant exchange parameter resulting in strong AFM bonds between

the magnetic layers. Therefore the LAFM state is favored over the FM state at the $\bar{\Gamma}$ point, which leads to $E_{\perp}(0) > 0$ and at the $\bar{M}'$ point the RW-AFM $_{\rightleftharpoons}$ state is preferred and thus $E_{\perp}(\mathbf{q}_{\bar{M}'} < 0$.

Turning our focus to the magnetic moments of spin spiral states in two magnetic layers of both $\rightleftharpoons$ and $\Rightarrow$ alignment [Figs. 8(e)–8(h)], we observe a similar behavior as for the case of the intralayer exchange. Upon variation of $\mathbf{q}$ along the high-symmetry directions of the BZ, the spin moment remains quite stable on the order of about 3 to 3.2 μ_B with increasing values up to 3.4 μ_B near the $\bar{\Gamma}$ and $\bar{K}'$ points for spin spirals with $\rightleftharpoons$ and $\Rightarrow$ in the freestanding Mn bilayer, respectively [Figs. 8(e) and 8(g)]. A similar trend is observed in the top Mn layer of the Mn BL on Ir(111) for which the maximum fluctuation with $\mathbf{q}$ amounts to 9% for both alignments [Figs. 8(f) and 8(h)]. However, as in the case of a flat spin spiral propagating only in one magnetic layers [cf. Figs. 7(b) and 7(d)], a different situation emerges for the bottom Mn layer, which is in direct contact with the Ir(111) surface. For the $\rightleftharpoons$ alignment of the Mn layers [Fig. 8(f)], the moment at the $\bar{\Gamma}$ point, i.e., in the FM state, is not only strongly reduced to 1.16 μ_B , but also varies by 94% for $\mathbf{q}$ vectors along the highsymmetry lines of the enlarged symmetry zone for spin spirals with the maximum value of 2.26 μ_B being taken at $|\mathbf{q}| = 0.66 \times 2\pi/a$ along $\bar{\Gamma}\bar{M}'$ (corresponding to a Néel state in each magnetic layer). A similar observation is made for the bottom Mn layer in the $\Rightarrow$ alignment between the two magnetic layers [Fig. 8(h)]. Note that the $\bar{\Gamma}$ point corresponds to the LAFM state in this alignment based on our definition.

3. Multi- Q states

Increasing the explored part of the magnetic phase space of the freestanding Mn bilayer and the Mn BL on Ir(111) to the prototypical multi- Q states, we obtain further surprising results. As recognizable in Fig. 8(a), the $uudd$ states along the $\bar{\Gamma}\bar{K}'$ and $\bar{\Gamma}\bar{M}'$ directions for the $\Rightarrow$ alignment of the freestanding Mn bilayer are quite close in energy to the corresponding 90° spin spirals. However, the energies of the respective superposition states for the $\rightleftharpoons$ alignment deviate significantly from those of the corresponding 1Q states (see Table X for exact values).

For the more complex film system of a Mn bilayer on Ir(111) all four $uudd$ states turn out to be energetically close to their constituents [see Fig. 8(b) and Table X]. In contrast to the $uudd$ states and 90° spin spirals, the multi- Q states at the $\bar{M}'$ point show a distinctively different trend. For a net FM alignment of spins in the two layers, the 3Q $_{\rightleftharpoons}$ state [cf. Fig. 4(d)] rises in energy with respect to its respective 1Q state, the RW-AFM $_{\rightleftharpoons}$ state. However, for a net AFM alignment, i.e., for the 3Q $_{\Rightarrow}$ state [cf. Fig. 4(c)], it is energetically favored over the RW-AFM $_{\rightleftharpoons}$ state. This effect is particularly pronounced in Mn/Mn/Ir(111) [Fig. 8(b)] with the 3Q $_{\rightleftharpoons}$ state gaining 45 meV/magnetic u.c. compared to its corresponding 1Q state, i.e., the RW-AFM $_{\rightleftharpoons}$ state.

Figure 9 compares the spin-dependent local density of states (LDOS) of the top and bottom Mn atoms for the two types of RW-AFM (1Q) and 3Q states in the Mn BL on Ir(111). One can see the stronger hybridization in the Mn interface (bottom) layer—sandwiched between the top Mn

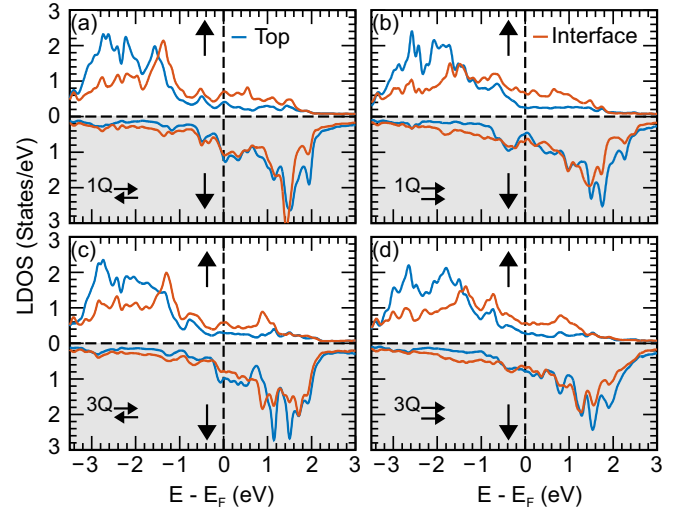


FIG. 9. Spin-decomposed local density of states (LDOS) for the Mn atoms for the two types of [(a) and (b)] RW-AFM (1Q) and [(c) and (d)] 3Q states in the Mn BL on Ir(111). (a)–(d) show the RW-AFM $_{\rightleftharpoons}$ (1Q $_{\rightleftharpoons}$) state, the RW-AFM $_{\Rightarrow}$ (1Q $_{\Rightarrow}$) state, the 3Q $_{\rightleftharpoons}$ state, and the 3Q $_{\Rightarrow}$ state, respectively. The blue and orange colors indicate the LDOS of the Mn atom from the top layer and interface layer, respectively. The black up arrow indicates the spin-up channel. The gray shaded area with black down arrow indicates the spin-down channel.

layer and the Ir surface—from the spreading of its LDOS, e.g., higher LDOS in the spin-up channel above E_F and lower LDOS at energies far below E_F . The hybridization between the two Mn layers is also apparent in all cases from peaks of similar shape occurring at the same energy.

The spin mixing between the Mn top and bottom layer, i.e., hybridization between states in the spin-up and -down channels, increases in the 3Q $_{\rightleftharpoons}$ and 1Q $_{\rightleftharpoons}$ state due to the net antiparallel alignment of spins between the layers. The hybridization and spin mixing between the two Mn layers is larger in the 3Q states than in the corresponding RW-AFM (1Q) states. This effect is visible, e.g., for the 3Q $_{\rightleftharpoons}$ vs. the 1Q $_{\rightleftharpoons}$ [Figs. 9(a) and 9(c)] in the spin-down channel for the unoccupied states. Here, a splitting of the peaks occurs in the 3Q $_{\rightleftharpoons}$ state. This spin mixing can be attributed to the tetrahedron angles between the spins in the 3Q state vs. the opposite alignment of spins in the RW-AFM state. Note that the enhanced hybridization between the layers promotes the effect of higher-order interactions that scale with the hopping parameter when derived from the Hubbard model [8].

4. Higher-order exchange interactions

One can understand the surprising energetic order of the two 3Q and the two RW-AFM states of the bilayer by taking into account the interlayer HOI terms introduced in Eq. (10) of Sec. II B. The Hamiltonian is of the form

$$H = H_{s\text{-BL}} + H_{\text{HOI}}^{\parallel} + H_{\text{HOI}}^{\perp} \quad (11)$$

with the separate terms corresponding to the pairwise Heisenberg exchange of a symmetrical bilayer, $H_{s\text{-BL}}$, the intralayer HOIs within each layer, $H_{\text{HOI}}^{\parallel}$, and the interlayer HOIs of the

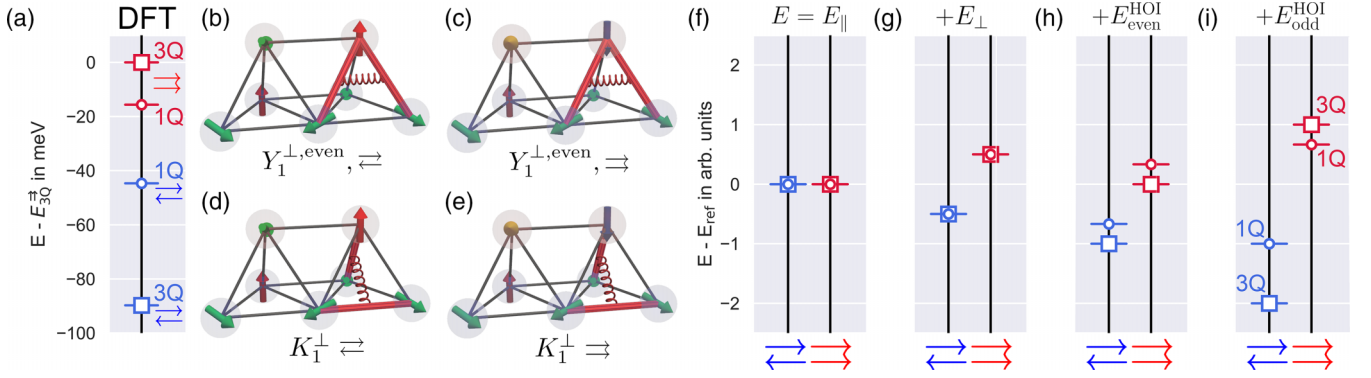


FIG. 10. (a) DFT total energy differences for the Mn bilayer on Ir(111) at the $\bar{M}'$ point of the Brillouin zone with respect to the $3Q_{\rightleftharpoons}$ state in meV per 2 Mn atoms. The energies of the RW-AFM (1Q, circle) and the 3Q state (square) are shown for $\rightleftharpoons$ (blue) and $\Rightarrow$ (red) alignment of the spins in the two layers (for states cf. Fig. 4). [(b) and (c)] The evenly distributed interlayer four-spin three-site interaction $Y_1^{\perp, \text{even}}$ is displayed schematically for the 3Q state in $\rightleftharpoons$ and $\Rightarrow$ alignment, respectively. The same spin structures are shown in (d) and (e) with the oddly distributed $K_1^{\perp}$ interaction. [(f)–(i)] Energies of 1Q (circle) and 3Q (square) states in $\rightleftharpoons$ (blue) and $\Rightarrow$ (red) alignments are shown qualitatively on an arbitrary energy scale. From (f) to (i), the energy contributions of Eq. (15) are shown. (f) Only bilinear intralayer exchange contributes with $E_{\parallel} = -1/2$. (g) Bilinear interlayer exchange is added with a value of $E_{\perp} = -1/2$. (h) Even intra- and interlayer HOI are added with a value of $E_{\text{even}}^{\text{HOI}} = -1/2$ for the 3Q states. (i) Odd interlayer HOI is added setting $E_{\text{odd}}^{\text{HOI}} = -1$ for the 3Q states. The HOI energy contributions ($E_{\text{even}}^{\text{HOI}}$, $E_{\text{odd}}^{\text{HOI}}$) to the 1Q states in (h),(i) are arbitrarily chosen to be 1/3 of the chosen values for the 3Q states.

bilayer, $H_{\text{HOI}}^{\perp}$ given in Eqs. (7), (2), and (10), respectively. We limit our further discussions to a Hamiltonian of a symmetric bilayer for easier comprehensibility, however, generalizing the HOIs to an asymmetrical bilayer is straightforward. For the symmetric bilayer, the intralayer pairwise and higher-order exchange constants of the top and bottom layers coincide and the interlayer higher-order interactions are symmetric. This reduces significantly the number of terms one needs to distinguish.

The energies of the RW-AFM $\rightleftharpoons$ and of the RW-AFM $\Rightarrow$ state, $E_{\text{RW-AFM}}^{\rightleftharpoons}$ and $E_{\text{RW-AFM}}^{\Rightarrow}$, are given by

$$E_{\text{RW-AFM}}^{\rightleftharpoons, \rightleftharpoons} = 2J_1^{\parallel} \pm J_1^{\perp} - 6B_1^{\parallel} + 4Y_1^{\parallel} - 12K_1^{\parallel} - 3B_1^{\perp} \pm 2Y_1^{\perp, \text{odd}} + 2Y_1^{\perp, \text{even}} \mp 6K_1^{\perp} \quad (12)$$

showing the energy contributions from the discussed NN exchange interactions. Note, that the change of spin alignment between the two layers introduces a sign flip of oddly distributed interactions.

The energy contributions to the respective $3Q_{\rightleftharpoons}$ and the $3Q_{\Rightarrow}$ state are given by

$$E_{3Q}^{\rightleftharpoons, \rightleftharpoons} = 2J_1^{\parallel} \pm J_1^{\perp} - \frac{2}{3}B_1^{\parallel} - \frac{4}{3}Y_1^{\parallel} - \frac{4}{3}K_1^{\parallel} - \frac{1}{3}B_1^{\perp} \mp \frac{2}{3}Y_1^{\perp, \text{odd}} - \frac{2}{3}Y_1^{\perp, \text{even}} \mp \frac{2}{3}K_1^{\perp}. \quad (13)$$

The first point to notice upon comparing Eqs. (12) and (13) is that the bilinear exchange interaction constants $J_1^{\parallel}$ and $J_1^{\perp}$ show the energetic degeneracy of the RW-AFM and corresponding 3Q states in the pairwise Heisenberg exchange Hamiltonian $H_{\text{s-BL}}$. If we consider the energy difference between these states, the pairwise exchange terms cancel and we obtain

$$\Delta E_{3Q}^{\rightleftharpoons, \rightleftharpoons} = E_{3Q}^{\rightleftharpoons, \rightleftharpoons} - E_{\text{RW-AFM}}^{\rightleftharpoons, \rightleftharpoons} = \frac{8}{3}(2B_1^{\parallel} - 2Y_1^{\parallel} + 4K_1^{\parallel} + B_1^{\perp} \mp Y_1^{\perp, \text{odd}} - Y_1^{\perp, \text{even}} \pm 2K_1^{\perp}). \quad (14)$$

HOI lift the degeneracy of 1Q and 3Q states, as seen from the difference between $E_{3Q}^{\rightleftharpoons, \rightleftharpoons}$ and $E_{\text{RW-AFM}}^{\rightleftharpoons, \rightleftharpoons}$ and only contributions from intra- and interlayer HOI remain in Eq. (14). Note that the pairwise exchange terms also cancel if we go beyond the NN approximation and consider inter- and intralayer exchange up to an arbitrary neighbor shell. We have performed similar calculations for the 90° spin spirals along the two highsymmetry directions and the corresponding $uudd$ states and the results are given in Eqs. (D1)–(D4) in Appendix D.

The DFT total energies of the RW-AFM (1Q) and 3Q states are displayed for the Mn bilayer on Ir(111) for both $\rightleftharpoons$ and $\Rightarrow$ spin alignment in Fig. 10(a) with respect to the energy of the $3Q_{\rightleftharpoons}$ state. Note, that these energies can also be seen at the $\bar{M}'$ point in Fig. 8(b). The RW-AFM $\rightleftharpoons$ (1Q $\rightleftharpoons$) state with a net antiferromagnetic coupling between the layers is clearly favorable by about 25 meV/unit cell than the RW-AFM $\Rightarrow$ (1Q $\Rightarrow$) state with ferromagnetic coupling. However, the energy gain of the 3Q state with respect to its corresponding 1Q state is very different for the two spin alignments between the layers. While the $3Q_{\rightleftharpoons}$ state for the case of net ferromagnetic alignment lies roughly 15 meV/uc above the RW-AFM $\Rightarrow$, the $3Q_{\rightleftharpoons}$ state shows a large energy gain of 45 meV/uc with respect to the corresponding RW-AFM $\rightleftharpoons$ state in the net antiferromagnetic alignment (for the four spin structures see Fig. 4).

This unexpected result can be understood based on Eq. (14). All intralayer HOI terms ($B_1^{\parallel}$, $Y_1^{\parallel}$, $K_1^{\parallel}$) and two of the interlayer HOI terms ($B_1^{\perp}$, $Y_1^{\perp, \text{even}}$) contribute in the same way to both energy differences. However, two of the interlayer HOI terms ($Y_1^{\perp, \text{odd}}$, $K_1^{\perp}$) have opposite signs for the two spin alignments. The different behavior of these HOI terms is explained from their symmetry (even or odd) if the spin moments of one of the two layers are flipped, i.e., switching between the $\rightleftharpoons$ and the $\Rightarrow$ state [going from Figs. 10(b) to 10(c) or from Figs. 10(d) to 10(e)]. All intralayer HOI terms are even, i.e., there is no change of sign in the energy terms, as

TABLE VI. Energy contributions from even and odd HOI terms to the energy difference of 1Q and multi-Q states as obtained via DFT for the Mn bilayer on Ir(111). The three points in the BZ are considered (3Q and RW-AFM state, $uudd$ and 90° spin spiral in the $\bar{\Gamma}\bar{M}'$ and $\bar{\Gamma}\bar{K}'$ directions). The values are given in meV per two Mn atoms.

$\mathbf{q}$	states	$E_{\text{even}}^{\text{MQ}} - E_{\text{even}}^{\text{1Q}}$	$E_{\text{odd}}^{\text{MQ}} - E_{\text{odd}}^{\text{1Q}}$
$\bar{M}'$	3Q/RW-AFM	-15.46	-31.08
$\frac{1}{2}\bar{\Gamma}\bar{M}'$	$uudd/90^\circ$ -spiral	10.67	6.67
$\frac{3}{4}\bar{\Gamma}\bar{K}'$	$uudd/90^\circ$ -spiral	-7.64	18.08

well as the biquadratic interlayer HOI and the even four-spin three-site interaction [Figs. 10(b) and 10(c)]. On the other hand, the four-spin four-site interaction with spin moments on a tetrahedron is odd [cf. Eq. (10) and Figs. 10(d) and 10(e)] just as the odd four-spin three-site term [cf. Fig. 5(b)].

Therefore the difference in the energy gain of the 3Q state to its respective RW-AFM state in the two alignments originates from the odd interlayer HOI terms. The contribution from even HOI terms moves the 3Q states of Fig. 10(a) down in energy, in the $\Rightarrow$ as well as the $\Leftarrow$ alignment of the Mn bilayer. The odd HOI terms favor the 3Q state in $\Leftarrow$ alignment even further and move the $3Q_{\Rightarrow}$ state up in energy. The contributions from odd and even HOIs are quantitatively different by a factor of approximately two, as seen in Table VI, driving the surprising behavior of energetically disfavored $3Q_{\Rightarrow}$ state to its RW-AFM $_{\Rightarrow}$ state and a huge energy gain of the $3Q_{\Leftarrow}$ state to the RW-AFM $_{\Leftarrow}$ state of 45 meV. This effect is even larger than the energy splitting of the two RW-AFM states due to interlayer exchange interactions. As a result the ideal bilayer 3Q state with tetrahedron angles between neighboring intra- and interlayer spin moments becomes the ground state of the Mn bilayer on Ir(111) through effects of interlayer higher-order interactions. Note, that for different signs of the odd interlayer HOI terms, e.g., in another material system, one could also obtain a situation in which the other 3Q state is stabilized in a bilayer.

5. Generalization to arbitrary multi-Q states

Following this classification of HOI terms, we can generalize the result discussed above for the 3Q state to an arbitrary multi-Q state and its corresponding 1Q state. One can write the energy in the Heisenberg model extended by intra- and interlayer HOI terms of an arbitrary spin state within a layer and parallel and antiparallel alignment between the two layers as

$$E^{\Leftarrow, \Rightarrow} = E_{\parallel} \pm E_{\perp} + E_{\text{even}}^{\text{HOI}} \pm E_{\text{odd}}^{\text{HOI}}, \quad (15)$$

where $E_{\parallel}$ and $E_{\perp}$ correspond to the energy of bilinear intra- and interlayer Heisenberg exchange interactions and $E_{\text{even}}^{\text{HOI}}$ and $E_{\text{odd}}^{\text{HOI}}$ are contributions from even and odd HOIs. For a multi-Q state (MQ) and its respective 1Q state (1Q) the energy for $\Leftarrow$ and $\Rightarrow$ alignment follows as

$$\begin{aligned} E_{1Q}^{\Leftarrow, \Rightarrow} &= E_{\parallel} \pm E_{\perp} + E_{\text{even}}^{\text{1Q}} \pm E_{\text{odd}}^{\text{1Q}}, \\ E_{MQ}^{\Leftarrow, \Rightarrow} &= E_{\parallel} \pm E_{\perp} + E_{\text{even}}^{\text{MQ}} \pm E_{\text{odd}}^{\text{MQ}} \end{aligned} \quad (16)$$

indicating the degeneracy in the bilinear exchange ($E_{\parallel}, E_{\perp}$) and the sign flip of odd interactions ($E_{\perp}, E_{\text{odd}}^{\text{MQ}}$) regarding the $\Leftarrow$ and $\Rightarrow$ alignment as discussed above. Here, the superscript denotes the different HOI contributions to the multi-Q and 1Q states.

The effect of the energy contributions to the 1Q and the corresponding multi-Q state in the $\Leftarrow$ and in the $\Rightarrow$ alignment as given by Eq. (15) is illustrated in Figs. 10(f)–10(i) for the example of the RW-AFM (1Q) and 3Q states. If only the interlayer bilinear exchange, $E_{\parallel}$, is considered [Fig. 10(f)] the 1Q and the 3Q states are energetically degenerate and their energies do not depend on the spin alignment between the layers. Upon taking the interlayer bilinear exchange, $E_{\perp}$, into account the energies for the two spin alignments are shifted with respect to each other [Fig. 10(g)]. Going beyond the Heisenberg bilinear exchange to the HOI, we find that the degeneracy of the 1Q and 3Q states is lifted by the even terms, $E_{\text{even}}^{\text{HOI}}$ [Fig. 10(h)]. Finally, the contribution from the odd HOI terms, $E_{\text{odd}}^{\text{HOI}}$, favors the $\Leftarrow$ alignment and disfavors the $\Rightarrow$ alignment for the chosen sign of $E_{\text{odd}}^{\text{HOI}}$ [Fig. 10(i)]. Taking the odd HOI into account—which all originate from interlayer HOI—allows to explain the DFT result for the Mn BL on Ir(111) [Fig. 10(a)]. Note that we have chosen arbitrary values for the different energy contributions here to illustrate the effect in a qualitative way.

We can further derive equations of energy differences, in which only the odd or the even HOI terms play a role. Taking the energy difference between the $\Leftarrow$ and $\Rightarrow$ alignment of one multi-Q state only bilinear interlayer exchange and odd HOI terms remain. Subtracting the same energy difference for the 1Q state leads to

$$(E_{MQ}^{\Leftarrow} - E_{MQ}^{\Rightarrow}) - (E_{1Q}^{\Leftarrow} - E_{1Q}^{\Rightarrow}) = 2(E_{\text{odd}}^{\text{MQ}} - E_{\text{odd}}^{\text{1Q}}) \quad (17)$$

in which the contribution from bilinear interlayer exchange vanishes and only odd HOI terms are present.

Now by adding the energies of $\Leftarrow$ and $\Rightarrow$ aligned layers and subtracting the 1Q state from the multi-Q state, one finds

$$(E_{MQ}^{\Leftarrow} + E_{MQ}^{\Rightarrow}) - (E_{1Q}^{\Leftarrow} + E_{1Q}^{\Rightarrow}) = 2(E_{\text{even}}^{\text{MQ}} - E_{\text{even}}^{\text{1Q}}) \quad (18)$$

showing only contributions from even HOIs, due to their indifference regarding the two alignments.

The contributions from odd and even HOI terms can be calculated via Eqs. (17) and (18), respectively, using the DFT total energies of the 1Q state and the corresponding multi-Q state. For the Mn bilayer on Ir(111), the values are displayed in Table VI. For the 3Q state, we find that the energy differences of even and odd HOI terms have the same sign and the odd HOI terms contribute with about twice the strength. Subtracting these two values explains the nearly 15 meV energy difference between the $3Q_{\Rightarrow}$ state and the RW-AFM $_{\Rightarrow}$ state observed in Fig. 10(a). For the opposite alignment between the two layers ($\Leftarrow$) the odd and even HOI energy contributions add up which results in the large energy gain of the $3Q_{\Leftarrow}$ state of about 45 meV with respect to the RW-AFM $_{\Leftarrow}$ state. We can apply the approach also for the $uudd$ states along the two highsymmetry directions of the 2D BZ (last two lines of Table VI).

The contributions from odd and even HOI terms to multi-Q states in the freestanding Mn BL are given in Table XI.

Here the contribution at the $\bar{M}'$ point of the BZ is even larger for the odd HOIs but does not surpass the energy difference of the RW-AFM states in the two alignment. Therefore the RW-AFM $_{\rightleftharpoons}$ is still the lowest in energy [as visible at the $\bar{M}'$ point in the dispersion Fig. 8(a)]. Note, that it is not possible to obtain the seven intra- and interlayer HOI constants from the six energy differences of the two 3Q and four *uudd* states, i.e., using the system of Eqs. (12), (13), and (D1)–(D4).

Recently, the 3Q state was also discovered in the bulk crystal $\text{Co}_{1/3}\text{TaS}_2$, in which hexagonal layers of Co atoms are separated by van der Waals spacer layers [51,52]. Based on neutron scattering and transport measurements it was concluded that a 3Q state exists within each of the Co layers with an $\rightleftharpoons$ alignment between layers as discussed in our work for Mn bilayers. Reference [51] discussed their findings also through a classical spin model containing intralayer Heisenberg and higher-order exchange ($J_1^{\parallel}, J_2^{\parallel}, B_1^{\parallel}$) for realizing the 3Q spin structure within the layers and a NN interlayer exchange ($J_1^{\perp} < 0$) aligns the layers in the net antiferromagnetic alignment of the ideal bilayer 3Q state. However, this spin model only takes contributions of $E_{\parallel}$, $E_{\perp}$, and $E_{\text{even}}^{\text{HOI}}$ from Eq. (15) into account, shown qualitatively in Figs. 10(f)–10(h) and does not consider interlayer HOI.

In contrast, we find that the stabilization of the 3Q $_{\rightleftharpoons}$ state [Fig. 4(c)] in Mn bilayers on Ir(111) [Fig. 10(a)] cannot be explained by intralayer HOI and interlayer Heisenberg exchange. We show that interlayer HOI acting between spins in the two layers must be taken into account, specifically $E_{\text{odd}}^{\text{HOI}}$ [Fig. 10(i)]. We introduce interlayer higher-order interactions in a NN framework and in a general way, which were to our knowledge not studied previously. These terms can favor either the bilayer 3Q state 3Q $_{\rightleftharpoons}$ [Fig. 4(c)] or the net ferromagnetically aligned 3Q state 3Q $_{\rightarrow}$ [Fig. 4(d)] depending on the sign of the interlayer HOI.

V. SUMMARY AND CONCLUSION

We have studied Mn mono- and bilayers on the Ir(111) surface using first-principles calculations based on DFT. For the Mn monolayer, the magnetic ground state depends on the stacking sequence. The energy dispersion of spin spirals exhibits the lowest state for both fcc and hcp stacking at the $\bar{K}$ point of the BZ corresponding to the Néel state with 120° angles between nearest neighbor spin moments. However, for fcc stacking the 3Q state with tetrahedron angles between nearest neighbors is energetically even lower. The stabilization of the 3Q state is a result of the energy gain due to intralayer higher-order exchange interactions.

For the Mn bilayer, we demonstrate based on spin spiral calculations that the antiferromagnetic pairwise intra- and interlayer exchange interactions favor a rowwise antiferromagnetic (RW-AFM) state with a net antiferromagnetic alignment ($\rightleftharpoons$) between spin moments of the Mn layers. Unexpectedly, the energy gain of the 3Q state due to higher-order interactions is much larger than the energy splitting between the two rowwise antiferromagnetic states with opposite interlayer couplings. In the predicted ideal bilayer 3Q state of the Mn bilayer on Ir(111), the magnetic moments exhibit tetrahedron angles between all nearest neighbor sites, i.e., within a

magnetic layer and between the two magnetic layers. Thereby, the topological orbital moments of the two layers—which emerge due to the noncoplanarity of the spin structure—point into the same direction leading to a significant total orbital magnetization [27]. In magnetic bilayers exhibiting the ideal 3Q state, we further anticipate the occurrence of a large spontaneous topological Hall effect that arises due to the emergent magnetic field in noncoplanar spin textures and has already been observed in transport experiments on the bulk system $\text{Co}_{1/3}\text{TaS}_2$ [51,52].

In order to explain the surprising observations for the Mn bilayer from our DFT calculations, we have developed an atomistic spin model for magnetic bilayers including intra- and interlayer higher-order exchange interactions. We show that the higher-order interactions can be classified into even and odd terms with respect to flipping the spin moments in one of the magnetic layers. This allows to distinguish quite generally the effect of HOI terms on the energy difference between a multi-Q state and its corresponding 1Q state. Based on our spin model, we explain that the bilayer 3Q state in a Mn bilayer on Ir(111) is stabilized by interlayer higher-order exchange interactions. The large energy gain of the bilayer 3Q state with tetrahedron angles between all nearest neighbors obtained from DFT is explained by an additive effect of even and odd higher-order exchange contributions.

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DATA AVAILABILITY

The data are available from the authors upon reasonable request.

APPENDIX A: FITTING FUNCTIONS FOR ENERGY DISPERSIONS OF MAGNETIC MONO- AND BILAYERS

In this section, we briefly explain how to calculate the fitting functions for the parameters of the pairwise Heisenberg exchange on the atomic hexagonal lattice of both a magnetic mono- and bilayer. Since our investigation focuses on periodic magnetic structures, it is convenient to represent the localized

magnetic moments of the Heisenberg model by their discrete Fourier components [47]

$$\mathbf{M}_i = \sum_{\mathbf{q}} \mathbf{M}_{\mathbf{q}} e^{i\mathbf{q}\mathbf{R}_i}, \quad (\text{A1})$$

with the sum running over the reciprocal lattice vectors $\mathbf{q}$ and the real space coordinates $\mathbf{R}_i$. By substituting the localized spins with their Fourier components in the Hamiltonian for the pairwise Heisenberg exchange [see the first term of Eq. (1)] and transforming the resulting expression further, we obtain the Heisenberg exchange interaction on the reciprocal lattice in terms of the Fourier transforms of the exchange constants $J(\mathbf{q})$:

$$E_{\text{exc}}(\mathbf{q}) = -NM^2 J(\mathbf{q}) = -NM^2 \sum_{\sigma} J_{\sigma} \sum_{\mathbf{r} \in \sigma} e^{-i\mathbf{q}\mathbf{r}}. \quad (\text{A2})$$

Here, M is the magnetic moment per site, N gives the number of lattice sites, and the index σ denotes the number of shells taken into account for the summation of interacting magnetic moments (cf. Fig. 11). As the exchange interaction is isotropic, spins with the same distance from a reference atom on the hexagonal lattice interact with the same exchange constant and can hence be grouped into one shell which is illustrated in Fig. 11(a). The fundamental solutions of the classical Heisenberg model on a periodic Bravais lattice can be written as [47]

$$\mathbf{M}_i = M(\mathbf{e}_1 \cos(\mathbf{q} \cdot \mathbf{R}_i) + \mathbf{e}_2 \sin(\mathbf{q} \cdot \mathbf{R}_i)). \quad (\text{A3})$$

This expression denotes a spin spiral in which the orthogonal unit vectors $\mathbf{e}_1$ and $\mathbf{e}_2$ span the rotational plane. In the absence of spin-orbit coupling, the vectors can be assumed to span the xy plane without loss of generality. Consequently, the resulting magnetic texture describes a flat spin spiral state in which the magnetic moments rotate by a constant angle from lattice site to lattice site around the z axis along the propagation direction given by the spin spiral vector $\mathbf{q}$. Rewriting the dot product between two spins as it appears in the Hamiltonian of

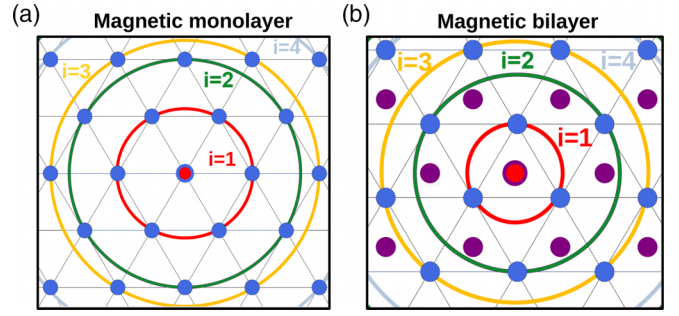


FIG. 11. Illustration of circular rings showing equidistant neighbors (shells) up to the third nearest neighbor for a reference atom on the hexagonal lattice in real space of (a) a magnetic monolayer and (b) a magnetic bilayer. Summing up the relative atomic positions of the σ th shell via Eq. (A4) and the first term of Eq. (1) yields an analytical expression for the pairwise (a) intralayer and (b) interlayer Heisenberg exchange which is further utilized to fit the respective DFT calculated energy dispersion.

the pairwise Heisenberg exchange [first term of Eq. (1)] via Eq. (A3), one obtains the following term:

$$\mathbf{M}_i \cdot \mathbf{M}_j = M^2 \cos(\mathbf{q} \cdot (\mathbf{R}_i - \mathbf{R}_j)). \quad (\text{A4})$$

This simplified expression can now be used to compute a fitting function including a desired number of nearest neighbor shells for the DFT calculated energy dispersion of a magnetic monolayer. With the two-dimensional spin spiral vector taking the form

$$\mathbf{q} = \frac{2\pi}{a} (q_x, q_y)^T, \quad (\text{A5})$$

the evaluation of the sum of the atomic positions of eight shells with respect to the position of a reference atom yields the analytical form of the pairwise intralayer Heisenberg exchange on the hexagonal lattice:

$$\begin{aligned} E_{\text{exc}} = & -J_1^{\parallel} [2 \cos(2\pi q_x) + 4 \cos(\pi q_x) \cos(\pi \sqrt{3} q_y)] - J_2^{\parallel} [2 \cos(2\pi \sqrt{3} q_y) + 4 \cos(3\pi q_x) \cos(\pi \sqrt{3} q_y)] \\ & - J_3^{\parallel} [2 \cos(4\pi q_x) + 4 \cos(2\pi q_x) \cos(2\pi \sqrt{3} q_y)] \\ & - J_4^{\parallel} [4 \cos(\pi q_x) \cos(3\pi \sqrt{3} q_y) + 4 \cos(4\pi q_x) \cos(2\pi \sqrt{3} q_y) + 4 \cos(5\pi q_x) \cos(\pi \sqrt{3} q_y)] \\ & - J_5^{\parallel} [2 \cos(6\pi q_x) + 4 \cos(3\pi q_x) \cos(3\pi \sqrt{3} q_y)] - J_6^{\parallel} [2 \cos(4\pi \sqrt{3} q_y) + 4 \cos(6\pi q_x) \cos(2\pi \sqrt{3} q_y)] \\ & - J_7^{\parallel} [4 \cos(2\pi q_x) \cos(4\pi \sqrt{3} q_y) + 4 \cos(5\pi q_x) \cos(3\pi \sqrt{3} q_y) + 4 \cos(7\pi q_x) \cos(\pi \sqrt{3} q_y)] \\ & - J_8^{\parallel} [2 \cos(8\pi q_x) + 4 \cos(4\pi q_x) \cos(4\pi \sqrt{3} q_y)]. \end{aligned} \quad (\text{A6})$$

Note that in principle any number of shells can be considered for the fitting function, however, since the influence of the pairwise exchange quickly decreases upon increasing the distance between two magnetic moments, we restrict ourselves to the inclusion of eight shells of neighboring atomic sites.

Applying the same approach to a magnetic bilayer, the fitting function for the pairwise interlayer Heisenberg exchange for one reference spin in the top magnetic layer interacting with neighboring spins up to the eighth order in the lower layer (the

same applies vice versa) is obtained as

$$\begin{aligned}
 E_{\text{exc}} = & -J_1^\perp \left[\cos\left(2\pi \frac{\sqrt{3}}{3} q_y\right) + 2 \cos(\pi q_x) \cos\left(\pi \frac{\sqrt{3}}{3} q_y\right) \right] - J_2^\perp \left[2 \cos(2\pi q_x) \cos\left(2\pi \frac{\sqrt{3}}{3} q_y\right) + \cos\left(4\pi \frac{\sqrt{3}}{3} q_y\right) \right] \\
 & - J_3^\perp \left[2 \cos(\pi q_x) \cos\left(5\pi \frac{\sqrt{3}}{3} q_y\right) + 2 \cos(3\pi q_x) \cos\left(\pi \frac{\sqrt{3}}{3} q_y\right) + 2 \cos(2\pi q_x) \cos\left(4\pi \frac{\sqrt{3}}{3} q_y\right) \right] \\
 & - J_4^\perp \left[2 \cos(3\pi q_x) \cos\left(5\pi \frac{\sqrt{3}}{3} q_y\right) + 2 \cos(4\pi q_x) \cos\left(2\pi \frac{\sqrt{3}}{3} q_y\right) + 2 \cos(\pi q_x) \cos\left(7\pi \frac{\sqrt{3}}{3} q_y\right) \right] \\
 & - J_5^\perp \left[\cos\left(8\pi \frac{\sqrt{3}}{3} q_y\right) + 2 \cos(4\pi q_x) \cos\left(4\pi \frac{\sqrt{3}}{3} q_y\right) \right] \\
 & - J_6^\perp \left[2 \cos(2\pi q_x) \cos\left(8\pi \frac{\sqrt{3}}{3} q_y\right) + 2 \cos(5\pi q_x) \cos\left(\pi \frac{\sqrt{3}}{3} q_y\right) + 2 \cos(3\pi q_x) \cos\left(7\pi \frac{\sqrt{3}}{3} q_y\right) \right] \\
 & - J_7^\perp \left[\cos\left(10\pi \frac{\sqrt{3}}{3} q_y\right) + 2 \cos(5\pi q_x) \cos\left(5\pi \frac{\sqrt{3}}{3} q_y\right) \right] \\
 & - J_8^\perp \left[2 \cos(4\pi q_x) \cos\left(8\pi \frac{\sqrt{3}}{3} q_y\right) + 2 \cos(2\pi q_x) \cos\left(10\pi \frac{\sqrt{3}}{3} q_y\right) + 2 \cos(6\pi q_x) \cos\left(2\pi \frac{\sqrt{3}}{3} q_y\right) \right]. \quad (\text{A7})
 \end{aligned}$$

Note that the interlayer exchange indeed requires a new analytical function since the shift of the two atomic sublattices of the magnetic bilayer results in other distances between interacting neighboring spins compared to the monolayer. As shown in Fig. 11(b), for a central reference atom located in one sublattice only three nearest and next-nearest neighbors in the other magnetic layer are left due to the lost rotational symmetry of the hexagonal lattice around the z axis.

APPENDIX B: EXCHANGE CONSTANTS FOR THE Mn ML ON Ir(111)

In the main part of the paper, a detailed discussion of the energy dispersion, the derived intralayer Heisenberg exchange parameters, the higher-order terms (HOI) as well as the influence of the Dzyaloshinskii-Moriya interaction (DMI) on the spin spiral states has been presented for both stackings of the Mn ML on Ir(111). For the sake of completeness, the full set of exchange constants obtained from DFT is listed in Tables VII–IX.

APPENDIX C: DMI CONTRIBUTION TO THE SPIN SPIRAL DISPERSION FOR THE Mn BL ON Ir(111)

In Fig. 13, we show the energy contributions of the DMI for the different types of spin spiral setups in the Mn BL on Ir(111) [cf. Figs. 7(b) and 8(b) for the respective scalar-relativistic energy dispersions]. Note that the maximum deviation for the DMI between the $\rightleftharpoons$ and the $\Rightarrow$ alignment of the magnetic layers in Fig. 13(b) amounts to only 4 meV/magnetic unit cell indicating the interlayer SOC effect to play a minor role for this ultrathin film system.

APPENDIX D: ANALYTICAL EXPRESSIONS FOR THE ENERGY OF 90° SPIN SPIRAL AND *uudd*-STATES IN BILAYERS

The main text focuses on the RW-AFM and the 3Q state, which arise from a spin spiral vector $\mathbf{q}$ at the $\overline{M}'$ -point of the BZ and the energy contributions of the Hamiltonian are discussed. In Eqs. (D1)–(D4), the energy contributions of the symmetrical bilayer Hamiltonian including HOIs $H = H_{\text{s-BL}} + H_{\text{HOI}}^\parallel + H_{\text{HOI}}^\perp$ to the 90°-spin spirals and *uudd* states are shown. $\rightleftharpoons$, $\Rightarrow$ denote the alignment of the bilayer magnetization according to Fig. 4.

$$E_{\frac{1}{2}\overline{\Gamma M'}, 1Q}^{\rightleftharpoons, \Rightarrow} = 2J_1^\parallel - 2B_1^\parallel + 0Y_1^\parallel - 12K_1^\parallel \mp 1J_1^\perp - 1B_1^\perp + 0Y_1^{\perp, \text{odd}} + 0Y_1^{\perp, \text{even}} \pm 2K_1^\perp, \quad (\text{D1})$$

$$E_{\frac{1}{2}\overline{\Gamma M'}, \text{uudd}}^{\rightleftharpoons, \Rightarrow} = 2J_1^\parallel - 6B_1^\parallel + 4Y_1^\parallel - 4K_1^\parallel \mp 1J_1^\perp - 3B_1^\perp \mp 2Y_1^{\perp, \text{odd}} + 2Y_1^{\perp, \text{even}} \pm 6K_1^\perp, \quad (\text{D2})$$

$$E_{\frac{3}{4}\overline{\Gamma K'}, 1Q}^{\rightleftharpoons, \Rightarrow} = -2J_1^\parallel - 2B_1^\parallel + 0Y_1^\parallel - 12K_1^\parallel \pm 1J_1^\perp - 1B_1^\perp + 0Y_1^{\perp, \text{odd}} + 0Y_1^{\perp, \text{even}} \pm 2K_1^\perp, \quad (\text{D3})$$

$$E_{\frac{3}{4}\overline{\Gamma K'}, \text{uudd}}^{\rightleftharpoons, \Rightarrow} = -2J_1^\parallel - 6B_1^\parallel - 4Y_1^\parallel - 4K_1^\parallel \pm 1J_1^\perp - 3B_1^\perp \pm 2Y_1^{\perp, \text{odd}} - 2Y_1^{\perp, \text{even}} \pm 6K_1^\perp. \quad (\text{D4})$$

TABLE VII. Full set of intralayer Heisenberg exchange constants ($J_i^{\parallel}$) calculated via DFT for fcc- and hcp-stacked Mn MLs on Ir(111). The values have been extracted from fitting the spin spiral energy dispersion $E(\mathbf{q})$ neglecting HOI shown in Fig. 6(a) of the main text. $J_i^{\parallel} > 0$ ($J_i^{\parallel} < 0$) denotes ferromagnetic (antiferromagnetic) coupling. All values are given in meV/Mn atom.

System	$J_1^{\parallel}$	$J_2^{\parallel}$	$J_3^{\parallel}$	$J_4^{\parallel}$	$J_5^{\parallel}$	$J_6^{\parallel}$	$J_7^{\parallel}$	$J_8^{\parallel}$
fcc-Mn/Ir(111)	-31.22	-4.29	-1.72	-0.30	0.00	-0.19	0.08	0.01
hcp-Mn/Ir(111)	-36.78	-4.47	-1.02	-0.18	0.01	0.01	0.08	-0.10

TABLE VIII. Intralayer Dzyaloshinskii-Moriya interaction constants ($D_i^{\parallel}$) and magnetocrystalline anisotropy energy constant (K_u) for fcc- and hcp-stacked Mn MLs on Ir(111) calculated via DFT. $D_i > 0$ ($D_i < 0$) denotes a preference of clockwise (counterclockwise) rotation of spin spirals and a positive value of K_u (obtained for the RW-AFM state) an in-plane easy magnetization axis. All values are given in meV/Mn atom.

System	$D_1^{\parallel}$	$D_2^{\parallel}$	$D_3^{\parallel}$	$D_4^{\parallel}$	$D_5^{\parallel}$	K_u
fcc-Mn/Ir(111)	2.77	0.26	0.27	-0.00	0.04	1.44
hcp-Mn/Ir(111)	2.82	0.44	0.28	-0.02	-0.04	1.72

TABLE IX. DFT calculated intralayer higher-order exchange interactions for both stackings of the Mn ML on Ir(111): four-spin four-site ($K_1^{\parallel}$), biquadratic ($B_1^{\parallel}$) and four-spin three-site interaction ($Y_1^{\parallel}$) constants calculated from Eqs. (3)–(5). ΔE terms the energy difference between the multi-Q and the corresponding 1Q spin spiral state, respectively, i.e., $\Delta E_{\frac{1}{2}\Gamma\bar{M}} = E_{\frac{1}{2}\Gamma\bar{M}}^{\text{uudd}} - E_{\frac{1}{2}\Gamma\bar{M}}^{\text{1Q}}$, $\Delta E_{\frac{3}{4}\Gamma\bar{K}} = E_{\frac{3}{4}\Gamma\bar{K}}^{\text{uudd}} - E_{\frac{3}{4}\Gamma\bar{K}}^{\text{1Q}}$, and $\Delta E_{\bar{M}} = E_{\bar{M}}^{\text{3Q}} - E_{\bar{M}}^{\text{1Q}}$. All values are given in meV/Mn atom.

System	$B_1^{\parallel}$	$Y_1^{\parallel}$	$K_1^{\parallel}$	$\Delta E_{\frac{1}{2}\Gamma\bar{M}}$	$\Delta E_{\frac{3}{4}\Gamma\bar{K}}$	$\Delta E_{\bar{M}}$
fcc-Mn/Ir(111)	-3.19	-3.02	-1.22	+15.05	-9.08	-13.95
hcp-Mn/Ir(111)	-3.19	-3.50	-0.89	+19.60	-8.36	-7.87

TABLE X. DFT calculated energy differences between multi- and single-Q states for a freestanding Mn bilayer and the hcp-Mn/hcp-Mn/Ir(111) film system. The values ΔE are calculated explicitly as follows: $\Delta E_{\frac{1}{2}\Gamma\bar{M}'}^{\rightleftharpoons, \rightarrow} = E_{\frac{1}{2}\Gamma\bar{M}'}^{\rightleftharpoons, \rightarrow} - E_{\frac{1}{2}\Gamma\bar{M}', \text{uudd}}^{\rightleftharpoons, \rightarrow} - E_{\frac{1}{2}\Gamma\bar{M}', \text{1Q}}^{\rightleftharpoons, \rightarrow}$, $\Delta E_{\frac{3}{4}\Gamma\bar{K}'}^{\rightleftharpoons, \rightarrow} = E_{\frac{3}{4}\Gamma\bar{K}'}^{\rightleftharpoons, \rightarrow} - E_{\frac{3}{4}\Gamma\bar{K}', \text{uudd}}^{\rightleftharpoons, \rightarrow} - E_{\frac{3}{4}\Gamma\bar{K}', \text{1Q}}^{\rightleftharpoons, \rightarrow}$, and $\Delta E_{3\text{Q}}^{\rightleftharpoons, \rightarrow} = E_{3\text{Q}}^{\rightleftharpoons, \rightarrow} - E_{\text{RW-AFM}}^{\rightleftharpoons, \rightarrow}$. All values are given in meV/per two Mn atoms in order to be able to directly compare them to the reference 1Q states. Note that $\rightleftharpoons, \rightarrow$ indicates the alignment of the Mn layers as described in Fig. 4.

System	$\Delta E_{3\text{Q}}^{\rightleftharpoons, \rightarrow}$	$\Delta E_{3\text{Q}}^{\rightarrow, \rightarrow}$	$\Delta E_{\frac{3}{4}\Gamma\bar{K}'}^{\rightleftharpoons, \rightarrow}$	$\Delta E_{\frac{3}{4}\Gamma\bar{K}'}^{\rightarrow, \rightarrow}$	$\Delta E_{\frac{1}{2}\Gamma\bar{M}'}^{\rightleftharpoons, \rightarrow}$	$\Delta E_{\frac{1}{2}\Gamma\bar{M}'}^{\rightarrow, \rightarrow}$
Mn/Mn	-39.20	65.70	82.72	26.12	50.10	-4.38
hcp-Mn/hcp-Mn/Ir(111)	-46.54	15.62	10.44	-25.72	17.34	-4.00

TABLE XI. Energy contributions from even and odd HOI terms to the energy difference of 1Q and multi-Q states obtained via DFT for the free-standing Mn bilayer through Eqs. (17) and (18). The three points in the BZ are considered (3Q and RW-AFM state, uudd and 90° spin spiral in $\Gamma\bar{M}'$ and $\Gamma\bar{K}'$ directions). The values are given in meV per two Mn atoms.

$\mathbf{q}$	States	$E_{\text{even}}^{\text{MQ}} - E_{\text{even}}^{\text{1Q}}$	$E_{\text{odd}}^{\text{MQ}} - E_{\text{odd}}^{\text{1Q}}$
$\bar{M}'$	3Q/RW-AFM	13.25	-52.45
$\frac{1}{2}\Gamma\bar{M}'$	uudd/90°-spiral	27.24	22.86
$\frac{3}{4}\Gamma\bar{K}'$	uudd/90°-spiral	54.42	28.30

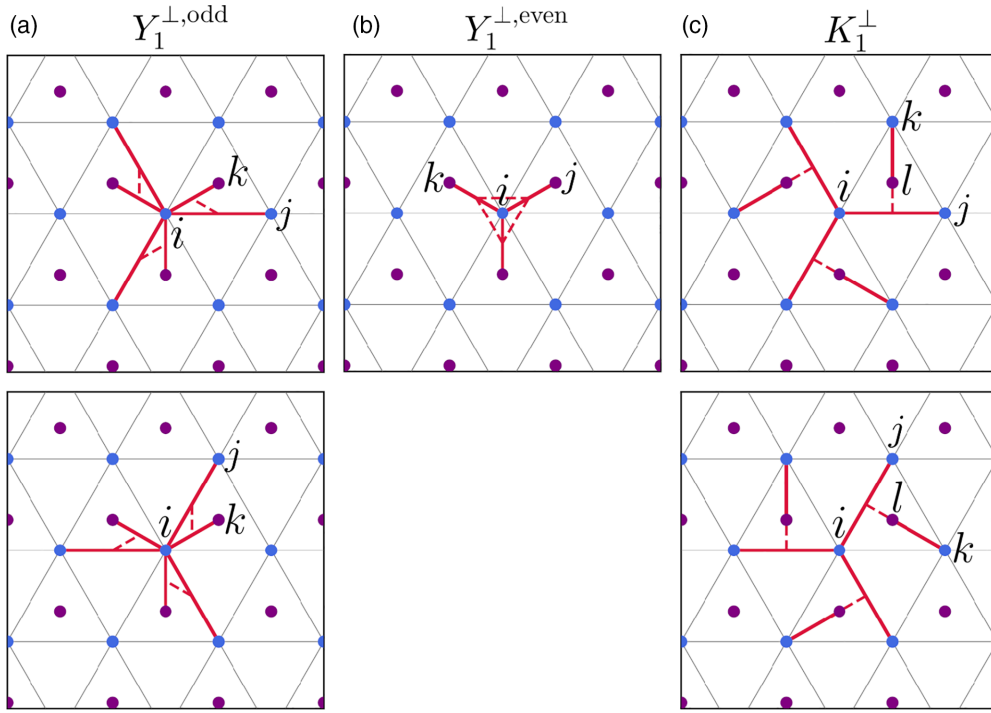


FIG. 12. Sketch of a hexagonal bilayer showing the pairs of atomic sites contributing to the interlayer higher-order interactions. (a) The odd interlayer four-spin three-site interaction, $Y_1^{\perp, \text{odd}}$. (b) The even interlayer four-spin three-site interaction, $Y_1^{\perp, \text{even}}$. (c) The odd interlayer four-spin four-site interaction, $K_1^{\perp}$. The atoms of the top layer (purple dots) lie in the hollow site of the bottom layer (blue dots). The four atomic sites participating in an interaction are connected by red lines, the scalar products (solid lines) are connected by a dotted line of the same color. Every panel displays the pairs given from the summation with a fixed atom i of one of the higher-order interactions. Every panel shows three pairs each. In (a) and (c), the pairs of the upper and lower panels are transferable through a C_3 -symmetry, adding up to six pairs for (a) and (c) and three for the $Y_1^{\perp, \text{even}}$ interaction (b). For the three-site terms (a) and (b), the indices i, j, k are shown for the three sites of one term and for the four-site term (c) the indices i, j, k, l are given for the four sites of one term.

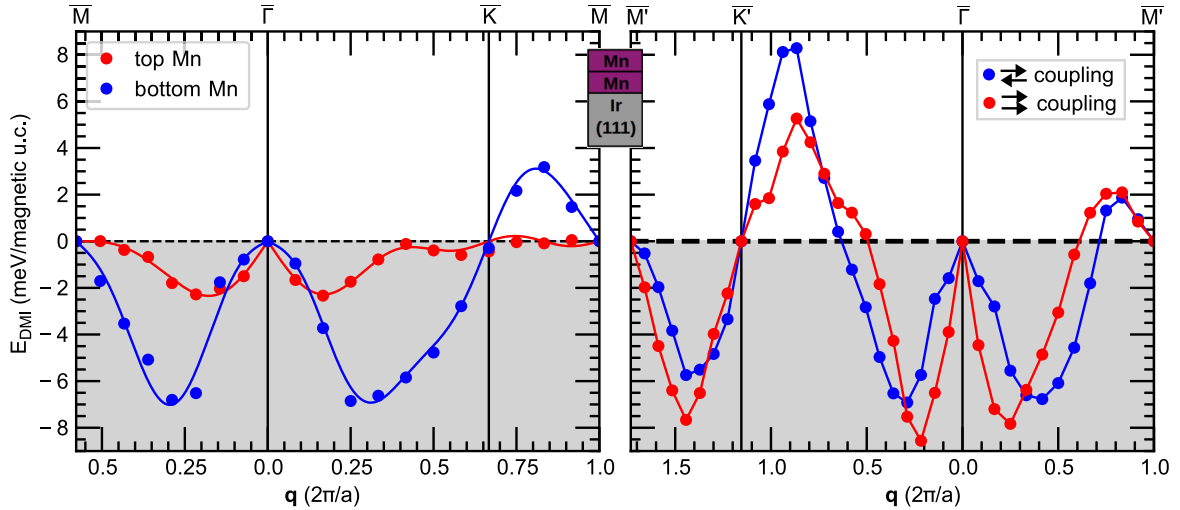


FIG. 13. Contributions of the Dzyaloshinskii-Moriya interaction (DMI) for cycloidal spin spiral states in the Mn BL on Ir(111). (a) Intralayer DMI obtained for $\mathbf{q}$ values of spin spirals in the 2D hexagonal BZ which propagate in only one magnetic layer, i.e., either the top or bottom Mn layer [the corresponding scalar-relativistically calculated energy dispersions are shown in Fig. 7(b)]. The solid lines running through the DFT values denote a fit to the intralayer DMI comprising five nearest neighbor shells. (b) DMI contributions for all $\mathbf{q}$ values of spin spirals propagating simultaneously in both magnetic layers [the respective scalar-relativistically calculated energy dispersion for a $\rightleftharpoons$ or $\Rightarrow$ alignment of the two layers at the $\bar{M}'$ point is displayed in Fig. 8(b)]. Here, the solid lines connecting the DFT data points serve as a guide to the eye. $E_{\text{DMI}} < 0 (> 0)$: preference of right-(left)-rotating cycloidal spin spirals. All values are given in meV/magnetic u.c., i.e., per two Mn atoms.

- [1] A. Fert, N. Reyren, and V. Cros, Magnetic skyrmions: advances in physics and potential applications, *Nat. Rev. Mater.* **2**, 17031 (2017).
- [2] C. Back, V. Cros, H. Ebert, K. Everschor-Sitte, A. Fert, M. Garst, T. Ma, S. Mankovsky, T. L. Monchesky, M. Mostovoy, N. Nagaosa, S. S. P. Parkin, C. Pfleiderer, N. Reyren, A. Rosch, Y. Taguchi, Y. Tokura, K. von Bergmann, and J. Zang, The 2020 skyrmionics roadmap, *J. Phys. D: Appl. Phys.* **53**, 363001 (2020).
- [3] B. Rimmler, B. Pal, and S. Parkin, Non-collinear antiferromagnetic spintronics, *Nat. Rev. Mater.* **10**, 109 (2024).
- [4] T. Moriya, New Mechanism of Anisotropic Superexchange Interaction, *Phys. Rev. Lett.* **4**, 228 (1960).
- [5] I. Dzyaloshinsky, A thermodynamic theory of “weak” ferromagnetism of antiferromagnetics, *Sov. Phys. JETP* **5**, 1259 (1957).
- [6] M. Takahashi, Half-filled Hubbard model at low temperature, *J. Phys. C* **10**, 1289 (1977).
- [7] A. H. MacDonald, S. M. Girvin, and D. Yoshioka, $\frac{1}{U}$ expansion for the Hubbard model, *Phys. Rev. B* **37**, 9753 (1988).
- [8] M. Hoffmann and S. Blügel, Systematic derivation of realistic spin models for beyond-Heisenberg solids, *Phys. Rev. B* **101**, 024418 (2020).
- [9] P. Kurz, G. Bihlmayer, K. Hirai, and S. Blügel, Three-Dimensional Spin Structure on a Two-Dimensional Lattice: Mn/Cu(111), *Phys. Rev. Lett.* **86**, 1106 (2001).
- [10] J. Spethmann, S. Meyer, K. von Bergmann, R. Wiesendanger, S. Heinze, and A. Kubetzka, Discovery of magnetic single- and triple-Q states in Mn/Re(0001), *Phys. Rev. Lett.* **124**, 227203 (2020).
- [11] S. Haldar, S. Meyer, A. Kubetzka, and S. Heinze, Distorted 3Q state driven by topological-chiral magnetic interactions, *Phys. Rev. B* **104**, L180404 (2021).
- [12] F. Nickel, A. Kubetzka, S. Haldar, R. Wiesendanger, S. Heinze, and K. von Bergmann, Coupling of the triple-q state to the atomic lattice by anisotropic symmetric exchange, *Phys. Rev. B* **108**, L180411 (2023).
- [13] Y. Yoshida, S. Schröder, P. Ferriani, D. Serrate, A. Kubetzka, K. von Bergmann, S. Heinze, and R. Wiesendanger, Conical spin-spiral state in an ultrathin film driven by higher-order spin interactions, *Phys. Rev. Lett.* **108**, 087205 (2012).
- [14] P. M. Weber, T. Drevelow, J. Qi, M. Bode, and S. Heinze, Evidence for a conical spin spiral state in the Mn triple layer on W(001): Spin-polarized scanning tunneling microscopy and first-principles calculations, *Phys. Rev. B* **108**, 134419 (2023).
- [15] A. Krönlein, M. Schmitt, M. Hoffmann, J. Kemmer, N. Seubert, M. Vogt, J. Küspert, M. Böhme, B. Alonazi, J. Kügel, H. A. Albrithen, M. Bode, G. Bihlmayer, and S. Blügel, Magnetic ground state stabilized by three-site interactions: Fe/Rh(111), *Phys. Rev. Lett.* **120**, 207202 (2018).
- [16] N. Romming, H. Pralow, A. Kubetzka, M. Hoffmann, S. von Malottki, S. Meyer, B. Dupé, R. Wiesendanger, K. von Bergmann, and S. Heinze, Competition of dzyaloshinskii-moriya and higher-order exchange interactions in Rh/Fe atomic bilayers on Ir(111), *Phys. Rev. Lett.* **120**, 207201 (2018).
- [17] S. Heinze, K. von Bergmann, M. Menzel, J. Brede, A. Kubetzka, R. Wiesendanger, G. Bihlmayer, and S. Blügel, Spontaneous atomic-scale magnetic skyrmion lattice in two dimensions, *Nat. Phys.* **7**, 713 (2011).
- [18] K. von Bergmann, M. Menzel, A. Kubetzka, and R. Wiesendanger, Influence of the local atom configuration on a hexagonal skyrmion lattice, *Nano Lett.* **15**, 3280 (2015).
- [19] M. Gutzeit, T. Drevelow, M. A. Goerzen, S. Haldar, and S. Heinze, Spontaneous square versus hexagonal nanoscale skyrmion lattices in Fe/Ir(111), *Phys. Rev. B* **108**, L060405 (2023).
- [20] F. Nickel, A. Kubetzka, M. Gutzeit, R. Wiesendanger, K. von Bergmann, and S. Heinze, Antiferromagnetic order of topological orbital moments in atomic-scale skyrmion lattices, *npj Spintronics* **3**, 7 (2025).
- [21] S. Paul, S. Haldar, S. Malottki, and S. Heinze, Role of higher-order exchange interactions for skyrmion stability, *Nat. Commun.* **11**, 4756 (2020).
- [22] M. Gutzeit, S. Haldar, S. Meyer, and S. Heinze, Trends of higher-order exchange interactions in transition metal trilayers, *Phys. Rev. B* **104**, 024420 (2021).
- [23] M. Gutzeit, A. Kubetzka, S. Haldar, H. Pralow, M. Goerzen, R. Wiesendanger, S. Heinze, and K. von Bergmann, Nano-scale collinear multi-q states driven by higher-order interactions, *Nat. Commun.* **13**, 5764 (2022).
- [24] A. Rodríguez-Sota, V. Saxena, J. Spethmann, R. Wiesendanger, R. Lo Conte, A. Kubetzka, and K. von Bergmann, Phase coexistence of Mn trimer clusters and antiferromagnetic Mn islands on Ir(111), *ACS Nano* **18**, 3699 (2024).
- [25] A. Aldarawsheh, M. Sallermann, M. Abusaa, and S. Lounis, Intrinsic Néel antiferromagnetic multimeric spin textures in ultrathin films, *J. Phys. Chem. Lett.* **14**, 8970 (2023).
- [26] J.-P. Hanke, F. Freimuth, A. K. Nandy, H. Zhang, S. Blügel, and Y. Mokrousov, Role of berry phase theory for describing orbital magnetism: From magnetic heterostructures to topological orbital ferromagnets, *Phys. Rev. B* **94**, 121114(R) (2016).
- [27] V. Saxena, M. Gutzeit, A. Rodríguez-Sota, S. Haldar, F. Zahner, R. Wiesendanger, A. Kubetzka, S. Heinze, and K. von Bergmann, Strain-driven domain wall network with chiral junctions in an antiferromagnet, *arXiv:2408.12580*.
- [28] B. Hardrat, A. Al-Zubi, P. Ferriani, S. Blügel, G. Bihlmayer, and S. Heinze, Complex magnetism of iron monolayers on hexagonal transition metal surfaces from first principles, *Phys. Rev. B* **79**, 094411 (2009).
- [29] S. Grytsiuk, J.-P. Hanke, M. Hoffmann, J. Bouaziz, O. Gomonay, G. Bihlmayer, S. Lounis, Y. Mokrousov, and S. Blügel, Topological-chiral magnetic interactions driven by emergent orbital magnetism, *Nat. Commun.* **11**, 511 (2020).
- [30] S. Brinker, M. dos Santos Dias, and S. Lounis, The chiral biquadratic pair interaction, *New J. Phys.* **21**, 083015 (2019).
- [31] A. Lászlóffy, L. Rózsa, K. Palotás, L. Udvardi, and L. Szunyogh, Magnetic structure of monatomic Fe chains on Re(0001): Emergence of chiral multispin interactions, *Phys. Rev. B* **99**, 184430 (2019).
- [32] S. Brinker, M. dos Santos Dias, and S. Lounis, Prospecting chiral multisite interactions in prototypical magnetic systems, *Phys. Rev. Res.* **2**, 033240 (2020).
- [33] Note that the distance along z does not matter in our setups since only spin spirals with $\mathbf{q} = (q_x, q_y, 0)^T$ are being investigated in this work.
- [34] <https://www.flapw.de>.
- [35] P. Kurz, F. Förster, L. Nordström, G. Bihlmayer, and S. Blügel, *Ab initio* treatment of noncollinear magnets with the

- full-potential linearized augmented plane wave method, *Phys. Rev. B* **69**, 024415 (2004).
- [36] P. E. Blöchl, Projector augmented-wave method, *Phys. Rev. B* **50**, 17953 (1994).
- [37] <https://www.vasp.at>.
- [38] G. Kresse and J. Furthmüller, Efficient iterative schemes for ab initio total-energy calculations using a plane-wave basis set, *Phys. Rev. B* **54**, 11169 (1996).
- [39] G. Kresse and D. Joubert, From ultrasoft pseudopotentials to the projector augmented-wave method, *Phys. Rev. B* **59**, 1758 (1999).
- [40] B. Dupé, M. Hoffmann, C. Paillard, and S. Heinze, Tailoring magnetic skyrmions in ultra-thin transition metal films, *Nat. Commun.* **5**, 4030 (2014).
- [41] K. von Bergmann, S. Heinze, M. Bode, E. Y. Vedmedenko, G. Bihlmayer, S. Blügel, and R. Wiesendanger, Observation of a complex nanoscale magnetic structure in a hexagonal Fe monolayer, *Phys. Rev. Lett.* **96**, 167203 (2006).
- [42] R. P. Feynman, Forces in molecules, *Phys. Rev.* **56**, 340 (1939).
- [43] S. H. Vosko, L. Wilk, and M. Nusair, Accurate spin-dependent electron liquid correlation energies for local spin density calculations: a critical analysis, *Can. J. Phys.* **58**, 1200 (1980).
- [44] P. Ferriani, K. von Bergmann, E. Y. Vedmedenko, S. Heinze, M. Bode, M. Heide, G. Bihlmayer, S. Blügel, and R. Wiesendanger, Atomic-scale spin spiral with a unique rotational sense: Mn monolayer on W(001), *Phys. Rev. Lett.* **101**, 027201 (2008).
- [45] M. Heide, G. Bihlmayer, and S. Blügel, Describing Dzyaloshinskii–Moriya spirals from first principles, *Phys. B: Condens. Matter* **404**, 2678 (2009).
- [46] C. Li, A. J. Freeman, H. J. F. Jansen, and C. L. Fu, Magnetic anisotropy in low-dimensional ferromagnetic systems: Fe monolayers on Ag(001), Au(001), and Pd(001) substrates, *Phys. Rev. B* **42**, 5433 (1990).
- [47] P. Kurz, Non-collinear magnetism at surfaces and in ultrathin films, Ph.D. thesis, RWTH Aachen, Jülich (2001), record converted from VDB: 12.11.2012; Aachen, Techn. Hoch., Diss., 2000.
- [48] S. Schröder, First-Principles Study of Non-Collinear Magnetism and Spin-Orbit Driven Physics in Nanostructures at Surfaces, Ph.D. thesis, Christian-Albrechts-Universität zu Kiel, Kiel, 2013.
- [49] In principle, both intra- and interlayer pairwise Heisenberg exchange constants could be determined simultaneously from a fit to the energy dispersion of $\rightleftharpoons$ or $\Rightarrow$ coupled magnetic layers. However, this only works for a symmetric bilayer with the same atom type in both layers and without substrate since the fitting routine cannot clearly assign the intralayer exchange to the corresponding layer.
- [50] Note that due to inversion symmetry no DMI contributions for the freestanding Mn bilayer are possible.
- [51] P. Park, W. Cho, C. Kim, Y. An, Y.-G. Kang, M. Avdeev, R. Sibille, K. Iida, R. Kajimoto, K. H. Lee *et al.*, Tetrahedral triple-Q magnetic ordering and large spontaneous Hall conductivity in the metallic triangular antiferromagnet $\text{Co}_{1/3}\text{TaS}_2$, *Nat. Commun.* **14**, 8346 (2023).
- [52] H. Takagi, R. Takagi, S. Minami, T. Nomoto, K. Ohishi, M.-T. Suzuki, Y. Yanagi, M. Hirayama, N. Khanh, K. Karube *et al.*, Spontaneous topological Hall effect induced by non-coplanar antiferromagnetic order in intercalated van der Waals materials, *Nat. Phys.* **19**, 961 (2023).
- [53] <https://www.nhr-verein.de/unsere-partner>.