

Stacking theory for bilayer two-dimensional magnets

Jun-Xi Du¹,^{*} Sike Zeng¹,^{*} and Yu-Jun Zhao¹,[†]

Department of Physics, South China University of Technology, Guangzhou 510640, China



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Two-dimensional unconventional magnetism has recently attracted growing interest due to its intriguing physical properties and promising applications in spintronics. However, existing studies on stacking-induced unconventional magnetism mainly focus on specific materials and stacking configurations. Here, we develop a general symmetry-based stacking theory for two-dimensional magnets. We first introduce spin layer groups as the fundamental symmetry framework, providing the essential magnetic symmetry information for the stacking theory. Based on this framework, we construct the complete set of 448 collinear spin layer groups for describing two-dimensional collinear magnets. Subsequently, we develop a general magnetic stacking theory applicable to arbitrary magnetic systems and derive its general solutions. Using CrF_3 as an illustrative example, we show how this theory enables designs of two-dimensional unconventional magnetism, as validated by first-principles calculations. We realize two-dimensional fully compensated ferrimagnetism through our stacking theory. Our work provides a general symmetry-guided platform for discovering and designing stacking-induced unconventional magnetism.

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I. INTRODUCTION

Magnetism, as one of the most fundamental and dynamic fields in condensed matter physics, has long attracted intense research interest owing to its rich underlying physics and its pivotal role in modern applications. Recently, unconventional magnetic materials that integrate the advantages of both ferromagnetism and antiferromagnetism have attracted considerable attention, as they hold strong promise as a foundation for next-generation spintronic devices [1]. Meanwhile, the experimental realization of monolayer CrI_3 [2] and bilayer $\text{Cr}_2\text{Ge}_2\text{Te}_6$ [3], together with the successful observation of long-range magnetic order therein, has stimulated increasing interest in two-dimensional (2D) magnetism [4]. From the perspective of applications, 2D magnetic materials, in contrast to bulk systems, possess high tunability and can be readily integrated into van der Waals (vdW) heterostructures, thereby offering versatile platforms for next-generation spintronic devices [5]. In this context, the extension of unconventional magnetism into the 2D limit has become a rapidly growing research direction. Several classes of two-dimensional unconventional magnetism, including 2D altermagnets [6,7], 2D fully compensated ferrimagnets [8], and type-IV 2D collinear magnets [9], have been proposed recently and have attracted considerable interest. These systems exhibit time-reversal symmetry-breaking responses even in the absence of a macroscopic magnetization. From a symmetry perspective, such magnetic states can be identified using spin-group symmetries. So far, research on 2D unconventional magnetism is still at an early stage, and corresponding experimental realizations remain largely unexplored [10].

Compared with bulk materials, 2D systems offer a unique “stacking” degree of freedom, enabling efficient manipulation of crystalline symmetries [11]. Consequently, stacking bilayer or multilayer structures has become a powerful route to realize 2D unconventional magnetism [12–18], with stacking-induced 2D altermagnetism receiving the most intensive attention. Nevertheless, previous investigations remain largely restricted to specific materials, and the theory developed for A-type altermagnets [14,15] fails to capture other altermagnets as well as other categories of 2D unconventional magnetism. These limitations naturally raise a fundamental question: Is there a general theory of magnetic stacking to guide the realization of 2D unconventional magnetism? Such a theory must resolve two central problems: first, given a monolayer and a specific stacking operation, predicting the resulting magnetic properties of the bilayer; second, given a monolayer and a target magnetic state, identifying the requisite stacking operations. This general theory becomes possible owing to recent developments in spin-group theory [19–24]. In contrast to magnetic groups, spin groups capture finer details of magnetic geometry, offering a powerful foundation for the theoretical design of magnetic materials.

Moreover, recent works have emphasized that spin-space-group symmetry provides a more complete description than magnetic-group symmetry for magnets with weak spin-orbit coupling (SOC), where it can distinguish electronic structures and quasiparticle features that are invisible at the magnetic-group level [19,20]. These developments further motivate the use of spin-layer-group symmetry as the natural language for stacking-engineered 2D magnetism.

In this work, we establish a general stacking theory for bilayer two-dimensional magnets by integrating spin-group symmetry with the stacking degree of freedom. We begin by introducing the spin layer group and constructing the complete set of 448 collinear spin layer groups. Subsequently,

^{*}These authors contributed equally to this work.

[†]Contact author: zhaoyj@scut.edu.cn

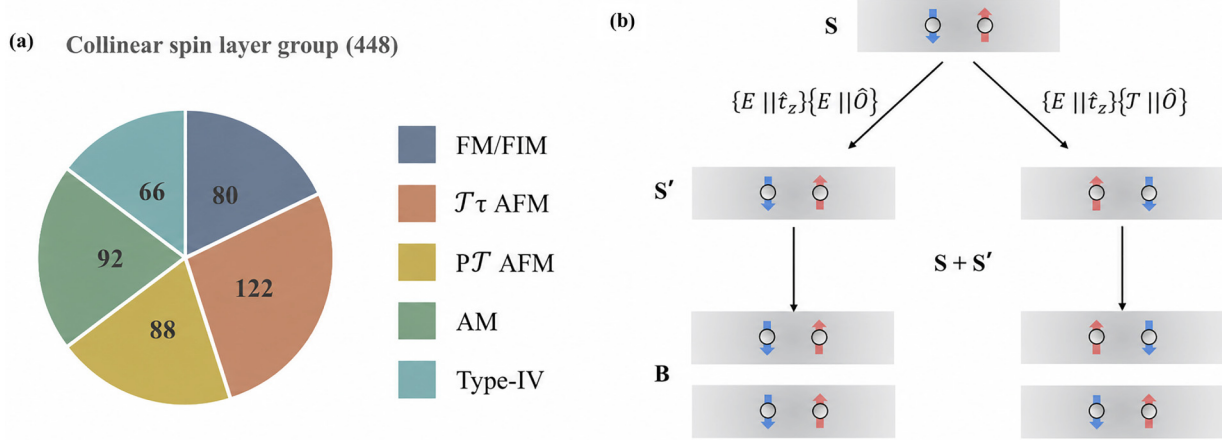


FIG. 1. Classification of collinear spin layer groups and schematic illustration of bilayer stacking in magnetic materials. (a) The 448 collinear spin layer groups (cSLGs) are classified into five categories according to the magnetic states they describe, including 80 ferromagnetic or ferrimagnetic (FM/FIM), 122 $\mathcal{T}\tau$ antiferromagnetic ($\mathcal{T}\tau$ AFM), 88 $\mathcal{PT}$ antiferromagnetic ($\mathcal{PT}$ AFM), 92 altermagnetic (AM), and 66 type-IV 2D collinear magnetic (Type-IV) cSLGs. (b) Starting from a monolayer S , a stacking operation $\{E \parallel \hat{t}_z\}\{c \parallel \hat{O}\}$ is applied to generate S' , which is then stacked onto S to form a bilayer B .

we develop the stacking theory for 2D magnets together with its analytical solutions. Using CrF_3 as a prototype, we demonstrate how this theory enables predictive design of unconventional magnetic states, with first-principles calculations confirming the theoretical predictions. Moreover, through symmetry analysis, we reveal that monolayer CrF_3 is a type-IV two-dimensional collinear magnet. This symmetry-based framework provides unified guidelines for engineering magnetism in 2D systems, opening avenues for stacking-induced unconventional magnetism.

II. SPIN LAYER GROUPS

As the foundation of the magnetic stacking theory, spin layer groups (SLGs) are introduced first. A symmetry operation of a SLG can be expressed as $\{R_i \parallel \hat{R}_j\}$, where R_i acts in spin space and $\hat{R}_j$ is a layer-group operation in real space. In this article, a caret is used to distinguish space-group symmetries from point-group symmetries. A spin layer group can be decomposed as $G = G_{\text{SO}} \times G_{\text{NSL}}$, where G_{SO} is the spin-only group containing only pure spin operations $\{R_i \parallel E\}$, and G_{NSL} denotes the nontrivial spin layer group [25]. For collinear spin layer groups (cSLGs) describing 2D collinear magnets, the spin-only group is the internal semidirect product of $\text{SO}(2)$ and $\mathbb{Z}_2^K$, $G_{\text{SO}} = \text{SO}(2) \rtimes \mathbb{Z}_2^K$, where $\text{SO}(2)$ describes continuous spin rotations about the collinear axis and $\mathbb{Z}_2^K \equiv \{E, U_n(\pi)\mathcal{T}\}$ [21]. Here, $U_n(\pi)$ denotes a 180° spin rotation about an axis perpendicular to the spin, and $\mathcal{T}$ is the time-reversal operator. For coplanar SLGs, the spin-only group is given by $G_{\text{SO}} = \{E, U_n(\pi)\mathcal{T}\}$, where $\mathbf{n}$ denotes the direction perpendicular to the plane of the magnetic structure. For noncoplanar SLGs, the spin-only group is only the identity, i.e., $G_{\text{SO}} = \{E\}$.

We then provide the complete set of 448 cSLGs that characterize 2D collinear magnets. The nontrivial part of a cSLG is in one-to-one correspondence with a magnetic layer group (MLG), owing to their similarity in mathematical structure [26], as listed in Table S1 [27]. Since a collinear

spin arrangement allows only two opposite spin orientations, the spin-space part of the nontrivial SLG is restricted to the identity E and time reversal $\mathcal{T}$. For collinear ferromagnets, which possess a single magnetic sublattice, the nontrivial cSLG takes the form $\{E \parallel G\}$, corresponding to type-I MLGs. For collinear antiferromagnets, which possess two magnetic sublattices, the nontrivial cSLG takes the form $\{E \parallel H\} + \{\mathcal{T} \parallel AH\}$, corresponding to type-III and type-IV MLGs. Here, H denotes an index-2 normal subgroup of G , and A is a coset representative. Based on this correspondence with MLGs [28], we enumerate all nontrivial parts of cSLGs, totaling 448, as listed in Tables S3–S7 [27].

We further classify these cSLGs according to the magnetism they describe, as illustrated in Fig. 1(a). First, the 80 cSLGs of the form $\{E \parallel G\}$ describe ferromagnets, as well as ferrimagnets in which multiple magnetic sublattices exist but are not related by symmetry. Two-dimensional fully compensated ferrimagnets are also described by these 80 cSLGs. As discussed above, the nontrivial spin layer group describing collinear antiferromagnets takes the form $\{E \parallel H\} + \{\mathcal{T} \parallel AH\}$. When A is a pure fractional translation τ , these cSLGs describe $\mathcal{T}\tau$ antiferromagnets, totaling 122. There are 88 cSLGs that possess $\mathcal{PT}$ symmetry, where $\mathcal{P}$ denotes spatial inversion, but lack $\mathcal{T}\tau$ symmetry. We classify these as $\mathcal{PT}$ antiferromagnets. When A is neither in $\{\tau, \mathcal{P}\}$ but belongs to $\{C_{2z}, M_z\}$, the corresponding cSLGs describe type-IV 2D collinear magnets, totaling 66. Finally, when A is not in $\{\tau, \mathcal{P}, C_{2z}, M_z\}$, these cSLGs correspond to 2D altermagnets, totaling 92. Altermagnets exhibit spin-splitting bands in the nonrelativistic limit, whereas the other antiferromagnets have spin-degenerate bands.

III. MAGNETIC STACKING THEORY

Subsequently, we present the magnetic stacking theory, which integrates spin layer groups with the freedom of stacking for 2D materials [11]. Since adjacent layers in van der Waals (vdW) materials are coupled primarily through

weak interlayer van der Waals interactions rather than strong chemical bonds [29], the interlayer coupling is generally much weaker than the intralayer interactions that stabilize the magnetic order within each layer. Therefore, we assume that stacking does not alter the intralayer magnetic order, which remains identical to that of the corresponding monolayer [3]. As illustrated in Fig. 1(b), a magnetic monolayer S with spin layer group $G_S = \{s \parallel \hat{R}_S\}$ generates a second layer S' through the stacking operation $\{E \parallel \hat{\tau}_z\} \{c \parallel \hat{O}\}$. Here, $\{s \parallel \hat{R}_S\}$ represents a symmetry operation of the monolayer, with s and $\hat{R}_S$ acting in spin space and real space, respectively. The operation $\{E \parallel \hat{\tau}_z\}$ is a pure translation along the z axis and does not affect the symmetry of the bilayer. In contrast, $\{c \parallel \hat{O}\} \equiv \{c \parallel O \mid \tau_O\}$ represents the nontrivial part of the stacking operation, where c denotes the spin-space part, and O and τ_O are the point-group operation and the translation in real space, respectively. The resulting bilayer B , characterized by the spin layer group $G_B = \{b \parallel \hat{R}_B\}$, is composed of these two layers. Here, b denotes the spin-space part of the spin-group symmetry of the bilayer. Based on the above analysis, we can formulate a magnetic bilayer stacking model:

$$\begin{aligned} S &= {}^s\hat{R}_S S, \quad \forall {}^s\hat{R}_S \in G_S \\ B &= {}^b\hat{R}_B B, \quad \forall {}^b\hat{R}_B \in G_B \\ B &= S + S' \\ S' &= {}^1\hat{\tau}_z^c \hat{O} S, \end{aligned} \quad (1)$$

where we denote the spin-group operations in a more compact form for simplicity, such as ${}^s\hat{R}_S \equiv \{s \parallel \hat{R}_S\}$. Note that the SLGs G_S and G_B include not only the nontrivial part but also spin-only group elements.

We now turn to the first problem: Given a monolayer and a specified stacking operation, how can one predict the SLG of the resulting bilayer system? Depending on whether the z coordinate is reversed, the spin-group symmetries of the bilayer can be divided into two classes, ${}^b\hat{R}_B^+$ and ${}^b\hat{R}_B^-$, which preserve and reverse the z coordinate, respectively. From a physical perspective, ${}^b\hat{R}_B^-$ interchanges S and S' , while ${}^b\hat{R}_B^+$ does not. For example, the spin-group operation $\{E \parallel P \mid \mathbf{0}\}$ interchanges the two monolayers and is thus classified as ${}^b\hat{R}_B^-$. The SLG of a bilayer is composed of two sets, $\{{}^b\hat{R}_B^+\}$ and $\{{}^b\hat{R}_B^-\}$, i.e., $G_B = \{{}^b\hat{R}_B^+\} \cup \{{}^b\hat{R}_B^-\}$. Using Eq. (1), we can obtain

$$\{{}^b\hat{R}_B^+\} = G_S \cap {}^c\hat{O} G_S {}^{c^{-1}}\hat{O}^{-1} \quad (2)$$

and

$$\{{}^b\hat{R}_B^-\} = {}^c\hat{O} G_S \cap G_S {}^{c^{-1}}\hat{O}^{-1}. \quad (3)$$

Here, ${}^{c^{-1}}\hat{O}^{-1}$ denotes the inverse of ${}^c\hat{O}$. The detailed derivation is presented in Sec. II of Supplemental Material [27]. The spin-only group of G_S and the stacking operation determine the spin-only group of G_B , which implies whether the bilayer magnetic configuration is collinear, coplanar, or noncoplanar. Therefore, if the class of the magnetic configuration in the bilayer is known, the spin-only group of G_S can be neglected. Equations (2) and (3) indicate that the SLG of the bilayer is determined not only by the stacking configuration ($\hat{O}$), but also by the interlayer magnetic order (c).

We then turn to the second problem: determining the stacking operations that yield a target bilayer magnetic state for a given monolayer. In the following, we still discuss ${}^b\hat{R}_B^-$ and ${}^b\hat{R}_B^+$ separately, and the detailed derivations are provided in Sec. II of Supplemental Material [27]. The spin-group symmetry ${}^b\hat{R}_B^-$ is allowed in the bilayer only if $({}^b\hat{R}_B^-)^2 \in G_S$. This condition is a prerequisite for a given monolayer to be stacked into a bilayer with a specific symmetry ${}^b\hat{R}_B^-$. Once the above condition is satisfied, ${}^b\hat{R}_B^-$ can be generated or preserved in B through a stacking operation ${}^c\hat{O} \in {}^b\hat{R}_B^- G_S$. In other words, selecting a stacking operation ${}^c\hat{O} \notin {}^b\hat{R}_B^- G_S$ can lead to the breaking of ${}^b\hat{R}_B^-$ in B . The ${}^b\hat{R}_B^-$ symmetry in a bilayer can in general be modulated through three independent channels: the real-space point-group operation, the interlayer sliding, and the interlayer spin configuration. Among them, the translational part of the stacking operation generally breaks the ${}^b\hat{R}_B^-$ symmetry; the only exception occurs for $R_B^- = P$.

For ${}^b\hat{R}_B^+$, a necessary condition for its presence in the bilayer is that the monolayer already possesses the same symmetry, i.e., ${}^b\hat{R}_B^+ \in G_S$. Consequently, stacking cannot generate ${}^b\hat{R}_B^+$; it can only preserve or break this symmetry, consistent with Eq. (2). Different ${}^b\hat{R}_B^+$ symmetries exhibit different dependencies on the stacking operation. For ${}^b\hat{C}_{nz}$, where C_{nz} denotes a rotation about the z axis, its preservation in B is determined by the spin part (c) and the translational part (τ_O), but not by the rotational part (O). By contrast, ${}^b\hat{E}$ (e.g., $\mathcal{T}\tau$) is determined by c and O , but not by τ_O . The symmetry is broken when the translational direction of ${}^b\hat{E}$ in the second layer does not align with the direction of any spin spiral in the first layer. For ${}^b\hat{M}_\beta$, where M_β represents a mirror plane perpendicular to the material plane, all three components, c , O , and τ_O , play a role. All operations in the spin-only group belong to ${}^b\hat{R}_B^+$ and thus can only be preserved or broken. Their preservation is determined solely by the spin-space part c of the stacking operation. Crucially, this operation also determines whether the magnetic configuration in the bilayer is collinear, coplanar, or noncoplanar.

These results demonstrate that stacking offers versatile means to manipulate symmetry, including interlayer magnetic order (c), sliding (τ_O), and relative rotational or mirror alignments between monolayers (O). It is worth noting that ${}^c\hat{O}$ uniquely labels an equivalence class of stacking operations, represented by the coset ${}^c\hat{O} G_S$. All operations within the coset ${}^c\hat{O} G_S$ generate the same bilayer.

We next consider several special cases. For the collinear case, where both the monolayer and the stacked bilayer exhibit collinear magnetic configurations, the spin part c is restricted to $\{E, \mathcal{T}\}$. In this case, the realization of ${}^b\hat{R}_B^+$ becomes independent of c . Consequently, ${}^b\hat{C}_{nz}$ can only be broken by sliding, whereas ${}^b\hat{E}$ can only be broken by proper or improper rotations. In the collinear case, the target bilayer symmetry ${}^b\hat{R}_B^-$ can still be classified into three cases. For case I, where ${}^b\hat{R}_B^- \in G_{S0}$, the stacking operation is given by $O = E$, $(E + R_B^-)\tau_O = \tau_{S0}$, and $c = 1$. For case II, where $R_B^- \in \tilde{G}_{S0}$ but ${}^b\hat{R}_B^- \notin G_{S0}$, one obtains $O = E$, $(E + R_B^-)\tau_O = \tau_{S0}$, and $c = \mathcal{T}$. For case III, where $R_B^- \notin \tilde{G}_{S0}$, the stacking operation must satisfy $O = R_B^-$, $(E + R_B^-)\tau_O = \tau_S^+ + \tau_{S0}$, and $c = b$. Here, G_{S0} denotes the spin-point-group part of the monolayer spin layer group G_S , while $\tilde{G}_{S0}$ represents the real-space part

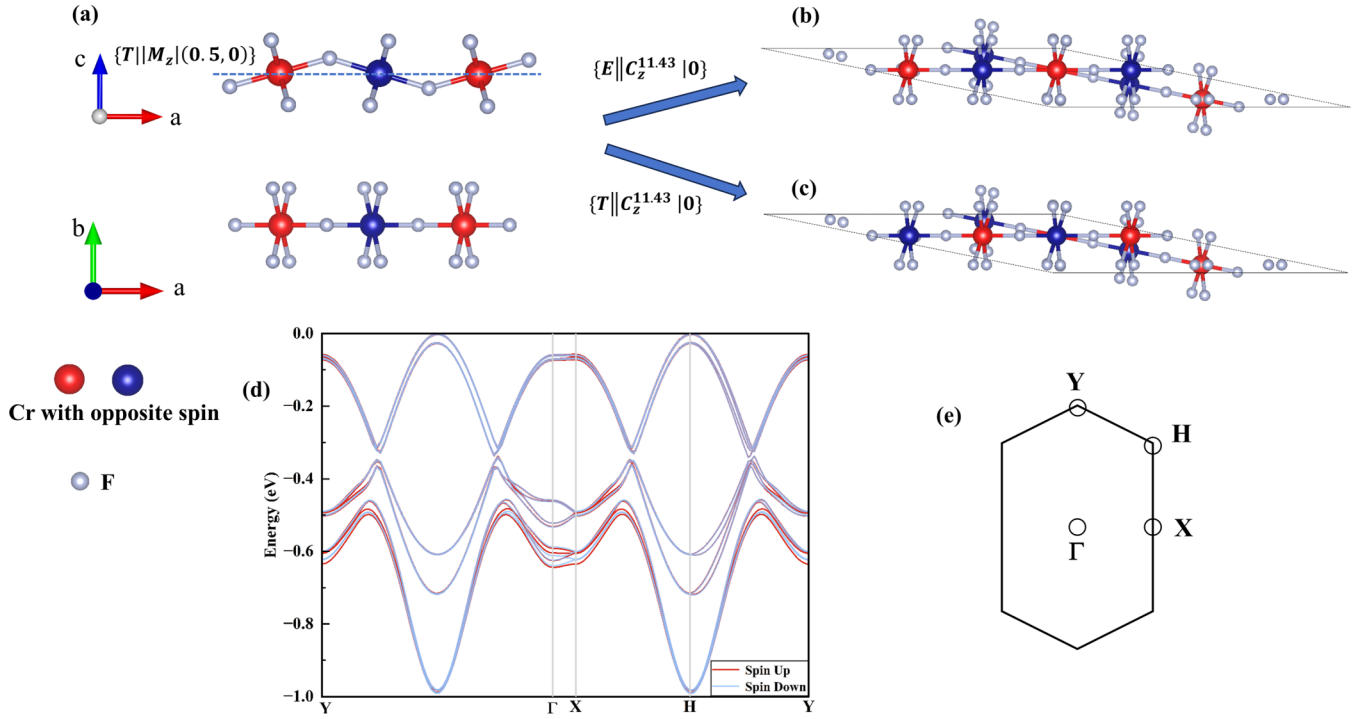


FIG. 2. Crystal structures of monolayer CrF_3 and twisted bilayer CrF_3 with a twist angle of 11.43° , together with the band structure of bilayer CrF_3 . (a) Crystal structure of monolayer CrF_3 . A monolayer CrF_3 is stacked into a bilayer via stacking operations $\{c \parallel C_z^{11.43} | \mathbf{0}\}$, where $C_z^{11.43}$ denotes a rotation of 11.43° about the z axis. For $c = E$, the resulting bilayer CrF_3 in (b) hosts fully compensated ferrimagnetism, whereas for $c = T$, the bilayer CrF_3 in (c) exhibits altermagnetism. (d) Band structure of the twisted bilayer CrF_3 shown in (b), with the high-symmetry points in reciprocal space shown in (e).

of G_{S0} and is also the real-space point-group part of G_S ; τ_{S0} represents an integer combination of the lattice translation vectors of the monolayer magnetic unit cell [27]. In other cases, tuning the interlayer magnetic order provides an effective means to modulate ${}^b\hat{R}_B^+$. When only the breaking of ${}^b\hat{R}_B^-$ is of interest, such as the breaking of $\mathcal{PT}$ symmetry, a simple strategy is to choose ${}^cO \notin {}^b\hat{R}_B^- G_{S0}$. Here, ${}^b\hat{R}_B^- ({}^cO)$ denotes the spin-point-group part of ${}^b\hat{R}_B^- ({}^c\hat{O})$, and G_{S0} is the spin point group associated with G_S .

IV. UNCONVENTIONAL MAGNETISM IN CrF_3

Recently, an antiferromagnet CrF_3 with a rectangular crystal structure has been proposed [30], as illustrated in Fig. 2(a), which is found to be energetically more stable than the widely assumed hexagonal phase. Through symmetry analysis, we find that monolayer CrF_3 is a type-IV 2D collinear magnet with the SLG $P^{-1}m^1m^{-1}a$, whose symmetry operations are listed in Table S6 [27]. With the easy axis along the out-of-plane direction [30], the system belongs to the magnetic group $Pmm'a'$ that permits a finite net magnetization along the a direction. First-principles calculations including spin-orbit coupling (SOC) further reveal a small canting of the magnetic moments toward the a direction. From the symmetry viewpoint, symmetry $\mathcal{M}_z\mathcal{T}$ enforces the integral of out-of-plane Berry-curvature component $\Omega_z(k_x, k_y)$ over the Brillouin zone to vanish, thereby forbidding the experimentally relevant Hall response σ_{xy} . For a detailed discussion, see Part E of Sec. II in SI [27]. Consequently, monolayer CrF_3 does not exhibit an observable anomalous Hall effect.

We then investigate stacking bilayer CrF_3 as a platform for realizing other types of 2D unconventional magnetism. First, we discuss altermagnetism, which has recently attracted considerable attention. To realize altermagnetism, the symmetries $\{T \parallel \mathcal{A}\}$ must be broken, where $\mathcal{A} \in \{\mathcal{P}, \mathcal{M}_z, \mathcal{C}_{2z}, \tau\}$. Since the translational components associated with $\mathcal{P}$ and $\mathcal{M}_z$ are irrelevant in the present context, this requirement can be satisfied by choosing cO such that it belongs neither to $\{T \parallel \mathcal{P}\}G_{S0}$ nor to $\{T \parallel \mathcal{M}_z\}G_{S0}$, thereby ensuring the absence of these two symmetry operations in the bilayer. Because $\{E \parallel \mathcal{P}\} \in G_{S0}$ and $\{T \parallel \mathcal{M}_z\} \in G_{S0}$, the corresponding cosets satisfy $\{T \parallel \mathcal{P}\}G_{S0} = \mathcal{T}G_{S0}$ and $\{T \parallel \mathcal{M}_z\}G_{S0} = G_{S0}$. If one further requires the bilayer to have the same magnetic unit-cell size as the monolayer, the point-group part of the stacking operation must be chosen from the symmetries of the rectangular magnetic lattice. Since the point group of this lattice is mmm , which coincides with the real-space part of G_{S0} , and since $c \in \{E, T\}$, all such stacking operations necessarily preserve either $\mathcal{PT}$ or $\mathcal{M}_z\mathcal{T}$. Stacking operations cannot remove all spin-degeneracy-enforcing symmetries; therefore, they cannot realize altermagnetism. Because monolayer CrF_3 does not exhibit the $\mathcal{T}\tau$ symmetry, the bilayer cannot exhibit this symmetry. The $\mathcal{C}_{2z}$ symmetry can be broken by sliding. To satisfy the symmetry requirement of altermagnetism, different magnetic sublattices must be connected by a symmetry operation. We consider an in-plane twofold rotation $\mathcal{C}_{2\alpha}$, with α taken along a generic in-plane direction, excluding the high-symmetry x - and y axes. Clearly, $(\{T \parallel \mathcal{C}_{2\alpha} | \mathbf{0}\})^2 \in G_S$. We choose ${}^c\hat{O} = \{T \parallel \mathcal{C}_{2\alpha} | \mathbf{0}\}\{E \parallel \mathcal{C}_{2y} | \mathbf{0}\} = \{T \parallel \mathcal{C}_z^\beta | \mathbf{0}\}$, where $\mathcal{C}_z^\beta$ denotes a rotation by an angle β about the z axis. Figure 2(c)

shows the crystal structure for $\beta = 11.43^\circ$, which exhibits altermagnetism. In this sense, there are indeed many possible stacking operations that can be chosen. In our twisted-bilayer calculations, we considered both lattice matching and the size of the magnetic unit cell. As a representative example, we selected the twist angle $\beta = 11.43^\circ$, which keeps a relatively small supercell with a lattice mismatch of 1.9% and a volume ratio of 2. Furthermore, we find that in this bilayer system the symmetry operation connecting different magnetic sublattices is only an in-plane twofold rotation. Choosing $^c\hat{O} = \{E \| C_z^{11.43} | \mathbf{0}\}$ can break this symmetry, leading to a fully compensated ferrimagnetic state, as illustrated in Fig. 2(b). A more detailed discussion is provided in Supplemental Material [27]. First-principles calculations show that the fully compensated ferrimagnetic state is energetically more favorable than the altermagnetic state. Therefore, we further calculate the band structure of the bilayer with the stacking operation $\{E \| C_z^{11.43} | \mathbf{0}\}$ without SOC, as shown in Fig. 2(d), using the high-symmetry path indicated in Fig. 2(e), and confirm a vanishing net magnetization. These results verify the fully compensated ferrimagnetism.

V. CONCLUSION

In summary, we propose a general magnetic stacking theory for two-dimensional magnets with all possible magnetic

configurations and systematically enumerate all 448 collinear spin layer groups. Using CrF_3 as an example, we demonstrate how magnetic stacking theory can be used to design and manipulate unconventional magnetism. Beyond explaining previously reported stacking-induced magnetic phenomena, our theory offers a unified and predictive guideline for discovering and engineering unconventional magnetism in 2D magnetic materials. Our work highlights stacking as a vital degree of freedom for magnetic symmetry in 2D magnets, thereby enriching the theoretical understanding of magnetism and symmetry breaking in two dimensions.

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DATA AVAILABILITY

There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

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