Prediction of Magnetic Skyrmions in A New Kind of Twisted Bilayer CrI₃

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Abstract

Magnetic skyrmions are topologically protected spin swirling vertices, which are promising in device applications due to their particle-like nature and excellent controlability. Magnetic skyrmions have been extensively studied in a variety of materials, and were proposed to exist in the extreme two-dimensional limit, i.e., in twisted bilayer CrI₃ (TBCI). Unfortunately, the magnetic states of TBCIs with small twist angles are disorderly distributed ferromagnetic (FM) and antiferromagnetic (AFM) domains in previous experiments, and thus the method to get rid of disorders in TBCIs is highly desirable. Here we propose the functions of interlayer exchange interactions obtained using first-principles calculations and stored in symmetry-adapted artificial neural networks. Based on them, the subsequent Landau-Lifshitz-Gilbert equation calculations explain the disorderly distributed FM-AFM domains in TBCIs with small twist angles and predict the orderly distributed skyrmions in TBCIs with large twist angles, which can be used in both spintronics and fundamental research.

Since the successful construction of twisted bilayer graphene[1, 2], rich properties of twisted bilayer systems have been demonstrated, such as
correlation-driven insulator[3, 4], superconductivity[5, 6], ferromagnetism[7–9], moiré exciton[10–12], and topolotical state[13, 14]. Recently, there has emerged the twisted bilayer CrI$_3$ (TBCI) with intrinsic magnetism. The magnetic state of a CrI$_3$ bilayer can be tuned by electric field[15–18], external pressure[19–22], and charge doping[23–25]. Theoretical studies have predicted that it also depends on the stacking structure[26–28], and a twisting may bring periodic magnetization domains with complex spin texture[29–31]; additional Dzyaloshinskii-Moriya (DM) interactions may further stabilize various magnetic skyrmions[32–35]. Compared with magnetic skyrmions in alloys[36–38], the magnetic skyrmions in TBCIs are much thinner, they reach the two-dimensional limit, and open up the field of spintwistronics[39].

In previous experiments, TBCIs with small twist angles show the coexistence of interlayer ferromagnetic (FM) and antiferromagnetic (AFM) states, but their domains are disorderly distributed[40, 41]. Therefore, experimentalists turned to the study of twisted multilayer CrI$_3$, such as twisted double bilayer CrI$_3$[42, 43] and twisted double trilayer CrI$_3$[41], since stacking more layers of CrI$_3$ suppresses the disorder. Then, orderly distributed magnetic domains were detected in twisted double trilayer CrI$_3$[42]. However, the method to get orderly distributed magnetic skyrmions in the much simpler TBCI is still unknown. In this work, we find a solution by extensively studying the magnetic states of TBCIs with arbitrary twist angles. Two kinds of TBCI have been found to have the coexistence of FM-AFM states. The first kind of TBCI has small twist angle, and it has been well studied in literature; the other kind is new, whose twist angle is around 60°. These two kinds of TBCI have quite different interlayer exchange fields, which explain the disorderly distributed FM-AFM domains in experiments and suggest the existence of orderly distributed skyrmions in the new kind of TBCI.

1 The Magnetic Exchange Interaction in TBCI

The magnetic exchange interactions in a TBCI are composed of intralyer and interlayer interactions. The interlayer interaction determines the interlayer magnetic order, and thus is the key to simulating magnetic states of TBCIs. In literature, it is usually approximated by using the interlayer interactions in the non-twisted bilayer CrI$_3$, which prevents the study of TBCIs with large twist angles. In this work, we used the accurate interlayer magnetic exchange interaction instead of the non-twist approximation. The interlayer magnetic exchange interaction between two Cr atoms depends on their in-plane relative position ($x = r \cos \theta$, $y = r \sin \theta$), interlayer distance ($z$), and the twist angle ($\varphi$). Since the Cr atoms in a CrI$_3$ layer form a honeycomb lattice consisting of two sublattices (named as A and B), the function $J(r, \theta, z, \varphi)$ has four different kinds, that are $J_{AA}$, $J_{AB}$, $J_{BA}$, and $J_{BB}$. The first and second footnotes denote the sublattices of the two Cr atoms. For instance, $J_{AB}$ describes the magnetic interaction between a Cr atom in the sublattice-A of down layer and another Cr atom in the sublattice-B of up layer. These four
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Fig. 1 (a) The twisted bilayer CrI$_3$ cluster model with the central Cr atoms shown in large balls to highlight their positions; (b) the structure of a SANN; (c,d) the isosurfaces of $J_{AA}$ for $\phi=0$ and $\pi/6$, respectively; (e,f) that of $J_{AB}$. The isosurfaces for the red and blue colors are 0.41 meV and -0.41 meV, respectively, where the positive (negative) value signs the FM (AFM) exchange interaction. The beginning of $z$ axis at the bottom is 6.5 Å. The corresponding cross section views of $J_{AA}$ and $J_{AB}$ at $z=6.62$ Å are shown in panel (g-k).

$J$-functions are not independent, since $J_{BA}$ and $J_{BB}$ can be obtained from $J_{AB}$ and $J_{AA}$ by symmetry constrains: $J_{BB}(r, \theta, z, \varphi) = J_{AA}(r, -\theta, z, -\varphi)$ and $J_{BA}(r, \theta, z, \varphi) = J_{AB}(r, -\theta, z, -\varphi)$. Besides, the symmetry of TBCI provides the following additional constrains (more details in Supplementary S.1).

$$J_{AA(B)}(r, \theta, z, \varphi) = J_{AA(B)}(r, \theta + \frac{2\pi}{3}, z, \varphi)$$

$$J_{AA(B)}(r, \theta, z, \varphi) = J_{AA(B)}(r, \theta, z, \varphi + \frac{2\pi}{3})$$

(1)

These constrains reduce the calculation of $J$-functions to a smaller variable space. The values of $J_{AA}$ and $J_{AB}$ at $\varphi=0$ and $\pi/3$ can be obtained directly by performing density functional theory (DFT) calculations, due to the small unit cells of the corresponding CrI$_3$ bilayers. In contrast, for most of the remaining twist angles, the TBCI has a large Morié pattern with huge amounts of atoms, which prevents the direct use of DFT calculations.
To solve the large-cell problem, we designed a cluster model, as shown in Fig. 1(a), to calculate the interlayer magnetic exchange interactions for arbitrary twist angles and translations. The cluster model contains two parallel CrI$_3$ plates, each of them has 13 Cr, 48 I, and 9 Li atoms. The radius of the plate is as large as 11.9 Å. The Li atoms are used to balance the electron-losing at the plate edge. We have checked the reliability of the cluster models by testing different cluster diameters, choosing different edge structures, and comparing the interlayer exchange interactions with those of periodic models (Supplementary S.2 and S.3). The cluster models were set with large amount of different stacking structures, with a $10 \times 10$ translation mesh from 0 Å to 9 Å, eight twist angles in the range of $0 \sim 2\pi/3$, and seven interlayer distances from 6.55 Å to 7.53 Å. The total number of stacking structures is 11200 for $J_{AA}$ and $J_{AB}$.

After DFT convergence[44], the values of $J_{AA}$ and $J_{AB}$ were obtained by using magnetic force theory (MFT) method[45, 46]. We then further constructed and trained symmetry-adapted artificial neural networks (SANNs), as shown in Fig. 1b and Supplementary S.4, to predict $J$-functions for periodic TBCIs with any stacking structures. We have used two SANNs to predict $J_{AA}$ and $J_{AB}$ separately, since the two functions have different symmetries. The input of a SANN is $(x, y, z, \cos\phi, \sin\phi)$. The twist angle was not used directly; instead we have used $\cos\phi$ and $\sin\phi$ to naturally encode the periodic condition in neural networks. The input data was proliferated into 18 equivalent copies according to the symmetries of the $J$ function. Then the 18 copies were separately fed into 18 feed-forward neural networks (FFNNs). These FFNNs share the same structure and parameters. Each branch of FFNN has four hidden layers containing 40, 100, 100, and 40 neurons, respectively. At last, the predicted results from the 18 branches were added together to produce the final prediction. The predicted $J_{AA}$ and $J_{AB}$ for two typical twist angles are shown in Fig. 1c, where the $J$ values decrease dramatically with an increase of the interlayer distance, and may switch signs by changing twist angles and in-plain relative positions. These $J$-functions from SANNs have been tested by reproducing the interlayer exchange energies of typical CrI$_3$ bilayers (Supplementary S.5). Based on the trained SANNs, the interlayer interaction parameters for a large TBCI can be obtained in very limited calculation consumings.

Due to the strong spin-orbit coupling and the unique atomic structure, the intralayer magnetic exchange interactions of CrI$_3$ are also complex. We have used the four-state method[47–49] (Supplementary S.6) to obtain the full 3×3 exchange matrices $J$ up to the third nearest neighbors, as shown in Fig. 2a. Our calculation results (Supplementary S.7) show that the exchange matrices for the first nearest neighbors and the third nearest neighbors are symmetric, which agree with the mirror symmetries between the two Cr atoms. The exchange matrices for the second nearest neighbors, however, are non-symmetric due to the missing of mirror symmetry. Therefore, $J_2$ contains DM interaction, which may stabilize magnetic skyrmions.
The Magnetic Domains in TBCI

After obtaining the interlayer and intralayer magnetic parameters, we then constructed the full Hamiltonians of TBCIs, and calculated their ground magnetic states. The Hamiltonian of a TBCI can be written as:

$$H = \sum_{i>j>0} S^T_i J_{ij} S_j + \sum_{i<j<0} S^T_i J_{ij} S_j + \sum_{i>0,j<0} J_{ij} S_i \cdot S_j$$

(2)
where $S_i$, whose length is $3/2$, is the spin of the Cr atom at site-$i$. The single ion anisotropy is neglected, since it is much smaller than the anisotropic exchange in CrI$_3$[50]. We have signed all the Cr atoms in the up layer by positive indices and those in the down layer by negative indices. The first and second terms at the right side describes the intralayer exchange interactions for up and down layers, respectively. The third term describes the interlayer exchange interactions. The matrices $J_{ij}$ for the first, second and third nearest neighbors were obtained by DFT calculations[51], and the other intralayer magnetic exchange interactions were neglected. The matrices $J_{ij}$ are different in different coordinate systems. A TBCI with nonzero $\varphi$ contains a rotated CrI$_3$ layer, where the matrix $J'_{ij}$ should be transformed to the new coordinate system by the rotation matrix, $J'_{ij} = R^{-1}(\varphi)J_{ij}R(\varphi)$. After obtained the $J_{ij}$ parameters in the last term from the SANNs, we then calculated the magnetic properties of the system. The equation of motion describing such a classical spin model is Landau-Lifshitz-Gilbert (LLG) equation

$$
\frac{dS_i}{dt} = \gamma S_i \times \frac{\partial H}{\partial S_i} + \gamma \alpha S_i \times (S_i \times \frac{\partial H}{\partial S_i}),
$$

where $\gamma$ is the gyromagnetic ratio, and $\alpha$ is the Gilbert damping coefficient, whose positive value (0.5~1.0 in our calculations) ensures that the system converges to a steady magnetic state. The ground states are FM in most cases, except for the TBCIs with $\varphi$ around 0° or 60°, which show the co-existence of FM and AFM states. In order to identify the FM and AFM domains, we have investigated the angle between the magnetic moment of a Cr atom in the up layer and that of neighboring Cr atoms in the down layer. The minimum angle is always 0°, which shows that the FM domains always exist. The maximum angle as a function of the twist angle is shown in Fig. 2b, where the angle increases significantly when $|\varphi| < 5.1^\circ$ or $|\varphi - 60^\circ| < 7.3^\circ$, and reaches 180° at $|\varphi| \sim 1.8^\circ$ or $|\varphi - 60^\circ| \sim 2^\circ$. These two kinds of TBCIs are different in their interlayer exchange fields. The AFM exchange field centers in the TBCI with $\varphi$ around 0° form a kagome lattice (Fig. 2c), while those around 60° form a triagonal lattice (Fig. 2g). Thus, we name these TBCIs as K-TBCI and T-TBCI, respectively.

In order to compare the two kinds of TBCI in more detail, we studied a K-TBCI with $\varphi = 1.7^\circ$ and a T-TBCI with $\varphi = 61.7^\circ$. Their Morié lattices have the same period length, with 4564 magnetic atoms in each period. Their interlayer exchange fields are shown in Fig. 2c,g: there are three squeezed AFM islands in a period of panel (c), and only one round AFM island in a period of panel (g). The three AFM islands in panel (c) are isolated by narrow and shallow FM canals. The exchange fields along $A_1-A_2$ and $F_1-F_2$ are shown in Fig. 2d. We can see that the FM canal is only about 30 Å wide, and the barrier as shown by shadow is lower than 0.3 meV/Cr. Thus, two neighboring AFM islands have chances to connect to each other, and the randomly occurring connections may lead to the disorder of magnetic states. By contrast, the AFM islands in panel (g) are far away from each other. There are two typical paths (line $A_1-A_2$ and curve $A_1-A_3$) connecting two neighboring islands.
The exchange fields (Fig. 2h) along the paths show that there are very wide and deep barriers (the shaddows). The barrier widths are larger than 150 Å and the barrier peaks along $A_1 - A_3$ are about 1.0 meV/Cr, and those along $A_1 - A_2$ are up to 1.5 meV/Cr. Therefore, two neighboring AFM islands are unlikely to fill up the barriers to form a connected area, and as a result, the system tends to have ordered magnetic domains.

![Image of skyrmions with charge -2, -1, 0, 1, and 2 in a T-TBCI with $\phi = 61.7^\circ$](image)

**Fig. 3** (a) The skyrmions with $c = -2, -1, 0, 1,$ and $2$ in a T-TBCI with $\phi = 61.7^\circ$, the yellow and blue colors signs the magnetic moments along $-z$ and $z$ directions, respectively, and the red arrows at skyrmion edge show $m_x$ and $m_y$ components; (b) the $m_z$ component of Cr atoms as a function of the distance between the Cr atom and the skyrmion center, and the solid lines fitted in $m_0 \tanh((r - R)/w)$; (c) the maximum skyrmion charge and the associated skyrmion radius as functions of Morié period length; (d,e) the excitation energy and skyrmion edge width as functions of skyrmion charge.

We then simulated the steady magnetic states for the K-TBCI with $\phi = 1.7^\circ$ and the T-TBCI with $\phi = 61.7^\circ$. The simulations used $2 \times 2$ Morié periods and random initial magnetic configurations. The results show that the AFM and FM areas are randomly distributed in the K-TBCI, two typical magnetic states are illustrated in Fig. 2e and 2f. On the other hand, the steady states for the T-TBCI are always isolated AFM domains surrounded by a connected FM
domain as shown in Fig. 2i,j. The simulations with other twist angles also show the same patterns: K-TBCIs have disorderly magnetic states, which agree well with the previous experiments\cite{40–42}; in contrast, the T-TBCIs have isolated AFM domains, which are in favor of skyrmions. Due to the ordered AFM domain distributions in T-TBCIs, we will focus on them in what follows.

The detailed domain structure in T-TBCI varies for different twist angles as shown in Fig. 2k-m. For $3.2^\circ < |\varphi - 60^\circ| < 7.3^\circ$, both the up and down layers have distorted magnetic spins, all Cr atoms in the island have positive $m_z$ components, and the in-plane components in different layers are in opposite directions. But for $|\varphi - 60^\circ| < 3.2^\circ$, the two layers become unsymmetric. The up layer has a skyrmion with significant spin distortions while the down layer has undistorted spins. The skyrmion may also exist in the down layer, and instead, the up layer has undistorted spins.

3 Magnetic Skyrmions in T-TBCI

Our further study shows that a variety of skyrmions may exist in T-TBCIs with $|\varphi - 60^\circ| < 3.2^\circ$. By setting many different initial spin configurations, we got different skyrmions. Examples are shown in Fig. 3a, where the second one is a chiral Bloch type Skyrmion. The $m_z$ components of skyrmions with three different twist angles are plotted in Fig. 3b, which clearly show the spin flipping across the skyrmion edge. They have shapes of hyperbolic tangent functions, and thus the fitting lines are also plotted by using:

\[
m_z(r, \phi) = m_0 \tanh(r - R)/w),
\]

where $R$ and $w$ are radius and width of the skyrmion, and the $m_0 = 3 \mu_B$ is the saturated magnetic moment. Considering the direction of magnetic moment at skyrmion edge, the $m_x$ and $m_y$ components as functions of $r$ and polar angle $\phi$ can be written as:

\[
m_x(r, \phi) = m_0 \frac{\cos(\phi_0 - c\phi)}{\cosh((r - R)/w)} \quad \text{and} \quad m_y(r, \phi) = m_0 \frac{\sin(\phi_0 - c\phi)}{\cosh((r - R)/w)}
\]

where $\phi_0$ is a phase factor. The number $c$ is a topological integer called skyrmion charge defined by

\[
c = \frac{1}{4\pi} \int n \cdot (\partial_x n \times \partial_y n) \, dx \, dy,
\]

where $n$ is the normalized magnetic moment $n = m/|m|$. By using this equation, the calculated skyrmion charges for Fig. 3a are -2, -1, 0, 1, and 2, respectively. The skyrmions with positive $c$ are also known as anti-skyrmions.

The skyrmion radius $R$ increases as $|c|$ increases, as shown in Fig. 3e, and both $R$ and the range of $c$ are dependent on $\varphi$, as shown in Fig. 3b, a skyrmion with small value of $|\varphi - 60^\circ|$ tends to have larger radius. The T-TBCI
with $\varphi = 62.5^\circ$ supports skyrmions with $c = -3 \sim 3$, therefore the maximum skyrmion charge is $|c|_{\text{max}} = 3$. All the skyrmions with $|c| > |c|_{\text{max}}$ are not stable during the LLG calculations. As shown in Fig. 3c, the $|c|_{\text{max}}$ increases linearly with increasing of Morié period length $a_M$, as $\varphi$ approaches $60^\circ$.

The excitation energy, defined as the energy difference between the system with a skyrmion and the system’s ground state, is shown in Fig. 3d. For a constant $c$, the excitation energy decreases with $\varphi$ approaching $60^\circ$. For example, the excitation energy of skyrmion with $c = 1$ in the T-TBCI with $\varphi = 62.3^\circ$ is 115 meV, while that for $\varphi = 60.3^\circ$ is only 8 meV. Since the skyrmion with $\varphi = 60.3^\circ$ contains $\sim$18000 atoms, the 8 meV is a very small amount of energy. From Fig. 3d, we can see that the trivial skyrmion ($c = 0$) always has the lowest energy, and the excitation energy increases as $|c|$ increases.

![Fig. 4](image-url) (a) A large upward translation of skyrmion caused by a small left moving of a CrI$_3$ layer in a T-TBCI; (b) skyrmion compressed by changing twist angle, and the skyrmion collapse at the critical point; (c) the structure of a monochromatic spin wave generator based on T-TBCI, where the H on skyrmions shows the high skyrmion charges; (d,e) the kagome and honeycomb skyrmion lattices (bright bubbles) at the up layers and the triagonal skyrmion lattices (shadowy bubbles) at the down layers.

### 4 Potential Applications

Due to the orderly distributed AFM domains and the existence of a variety of skyrmions, the T-TBCI can be used in device applications and fundamental research. Here we provide a few conceptual designs. Firstly, the T-TBCI with $|\varphi - 60^\circ| < 3.2^\circ$ contains a skyrmion lattice and can be used as a memory device. The information can be coded into two channels, the skyrmion charges as well as the skyrmion positions (at up or down CrI$_3$ layer). The skyrmion with the stored information can be transported by the in-plain translation of one CrI$_3$ layer (Fig. 4a and Supplementary S.8), and the skyrmion radius can...
be tuned by the change of $\varphi$ (Fig. 4b and Supplementary S.8). When $\varphi$ deviates from 60°, the skyrmion shrinks and eventually collapses across the critical point ($c = |c|_{\text{max}}$). The collapse releases the redundant energy in many forms including spin waves. Based on that, the second application, a monochromatic spin wave generator was designed as shown in Fig. 4c, where the collapse of skyrmions in lattice $E$ emit spin waves and the lattice $G$ works as a spin wave grating.

In the foundamental research, the T-TBCI is a promising platform for the study of spin topology and dynamics. For example, a variety of skyrmion lattices can be designed in T-TBCIs. As shown in Fig. 4d,e, the skyrmions at the up layers form honeycomb and kagome lattices, respectively; and the down layers have sparse triagonal lattices. The rich skyrmion lattice types combined with the multiple choices of skyrmion charges make T-TBCI an intriguing platform for searching topologically nontrivial spin waves, especially when the system contains chiral Bloch type skyrmions.

## 5 Conclusion

This work has proposed the cluster TBCI models and SANNs, which enable us to calculate the functions of interlayer exchange interactions and efficiently predict these interactions in an arbitrarily stacked CrI$_3$ bilayer. Besides TBCI, these methods can also be used in the study of twisted CrI$_3$ multilayers and the stacking structures of other layered magnetic materials. The following LLG calculations predicted that T-TBCI has orderly distributed skyrmions, which can be used in novel device applications. Furthermore, a variety of skyrmion lattices can be designed in T-TBCI, where topologically nontrivial spin waves may be found. Due to these unique properties, we call on further theoretical and experimental studies on this new material.

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