Magnetic Skyrmion Lattices in a Novel 2D-Twisted Bilayer Magnet

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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 are extensively studied in a variety of materials and proposed to exist in the extreme 2D limit, i.e., in twisted bilayer CrI$_3$ (TBCI). Unfortunately, the magnetic states of TBCIs with small twist angles are disorderly distributed ferromagnetic and antiferromagnetic (AFM) domains in recent experiments, and thus the method to get rid of disorders in TBCIs is highly desirable. Here, intralayer exchange interactions up to the third nearest neighbors without empirical parameters and very accurate interlayer exchange interactions are used to study the magnetic states of TBCIs. The functions of interlayer exchange interactions obtained using first-principles calculations and stored in symmetry-adapted artificial neural networks are proposed. Based on these, 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. This novel twisted 2D bilayer magnet can be used to design memory devices, monochromatic spin wave generators and many kinds of skyrmion lattices.

1. Introduction

Since the successful construction of twisted bilayer graphene,[1,2] rich properties of twisted bilayer systems have been demonstrated, such as correlation-driven insulators,[3,4] superconductivity,[5,6] ferromagnetism,[7–9] moiré excitons,[10–12] and topological states.[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,[26–29] and a twisting may bring periodic magnetization domains with complex spin texture.[30–32] Additional Dzyaloshinskii–Moriya (DM) interactions may further stabilize various magnetic skyrmions.[33–36] Rich phase diagrams with noncollinear spin configurations were obtained.[17,38] Compared with magnetic skyrmions in alloys,[39–42] the magnetic skyrmions in TBCIs are much thinner, which reach the 2D limit and open up the field of spin-twistrronics.

In previous experiments, TBCIs with small twist angles show the coexistence of interlayer ferromagnetic (FM) and antiferromagnetic (AFM) states, but their magnetic domains are disorderly distributed.[43,44] Therefore, experimentalists turned to the study of twisted multilayer CrI$_3$, such as twisted double bilayer CrI$_3$[45–47] and twisted double trilayer CrI$_3$[46] since stacking more layers of CrI$_3$ suppresses the disorder. Then, orderly distributed magnetic domains were detected in twisted double trilayer CrI$_3$.[45] However, the method for getting 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 were found having the coexistence of FM-AFM states, but their magnetic domains are disorderly distributed.[43,44] Therefore, experimentalists turned to the study of twisted multilayer CrI$_3$, such as twisted double bilayer CrI$_3$[45–47] and twisted double trilayer CrI$_3$[46] since stacking more layers of CrI$_3$ suppresses the disorder. Then, orderly distributed magnetic domains were detected in twisted double trilayer CrI$_3$.[45]

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2. Results and Discussion

2.1. The Magnetic Exchange Interaction in TBCI

The magnetic exchange interactions in a TBCI are composed of intralayer and interlayer interactions. The interlayer
interaction determines the interlayer magnetic order, and thus is the key to simulating magnetic states of TBCIs. In literature, the interaction is usually approximated by using the interlayer interactions in the non-twisted bilayer CrI$_3$, which are only applicable to the study of TBCIs with small 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 (\(\phi\)). Since the Cr atoms in a CrI$_3$ layer form a honeycomb lattice consisting of two sublattices (named as A and B), the interlayer magnetic exchange interaction \(J(r, \theta, z, \phi)\) 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 lower layer and another Cr atom in the sublattice-B of upper layer. These four \(J\)-functions are not independent, since \(J_{BA}\) and \(J_{BB}\) can be obtained from \(J_{AB}\) and \(J_{AA}\) by symmetry constraints:

\[
J_{BA}(r, \theta, z, \phi) = J_{AA}(r, -\theta, z, -\phi) \\
J_{BB}(r, \theta, z, \phi) = J_{AB}(r, \theta - \varphi, z, \phi) \\
J_{AA}(r, \theta, z, \phi) = J_{AA}(r, \theta \pm \frac{2\pi}{3}, z, \phi) \\
J_{AB}(r, \theta, z, \phi) = J_{AB}(r, \theta \pm \frac{2\pi}{3}, z, \phi)
\]

These constraints reduce the calculation of \(J\)-functions to a smaller variable space. The unit cells for \(\varphi = 0\) and \(\pi/3\) are very small, thus the values of \(J_{AA}\) and \(J_{AB}\) can be obtained directly by performing density functional theory (DFT) calculations. In contrast, for most of the remaining twist angles, the TBCIs have large Möbius patterns with huge amounts of atoms, which prevent the direct use of DFT calculations.

To solve the large-cell problem, we designed a cluster model, as shown in Figure 1a, 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

**Figure 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 \(\varphi = 0\) and \(\pi/6\), respectively; e,f) that of \(J_{AB}\). The isosurfaces for the red and blue colors are 0.41 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 panels (g–k).
interactions with those of periodic models (see the Supporting Information for further details on model testing). The cluster models were set with large amount of different stacking structures, with a $10 \times 10$ translation mesh from 0 to 9 Å, eight twist angles in the range of $0 - 2\pi/3$, and seven interlayer distances (between the two Cr-plane) from 6.55 to 753 Å. The total number of stacking structures is 11200 for $J_{AA}$ and $J_{AB}$.

After DFT convergence, the values of $J_{AA}$ and $J_{AB}$ were obtained by using a magnetic force method (MFT) method. We then further constructed and trained symmetry-adapted artificial neural networks (SANNs), as shown in Figure 1b, 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 were 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 Figure 1c, where the $J$ values decrease dramatically with an increase of the interlayer distance, and may switch signs by changing twist angles and in-plane relative positions. These $J$-functions from SANNs have been tested by reproducing the interlayer exchange energies of typical CrI$_3$ bilayers (see the Supporting Information for further details on interlayer exchange energies). 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 to obtain the full 3x3 exchange matrices $\mathcal{J}$ up to the third nearest neighbors, as shown in Figure 2a. The exchange interactions for the fourth nearest neighbors were neglected, since the corresponding values of $\mathcal{J}$ were estimated to be smaller than 0.05 meV in MFT calculations. Our calculation results show that the exchange matrices for the first nearest neighbors and the third nearest neighbors are symmetric, which agree with the inverson symmetries between the two Cr atoms. The exchange matrices for the second nearest neighbors, however, are non-symmetric due to the missing of inversion symmetry. Therefore, $\mathcal{J}_2$ contains DM interaction, which may stabilize magnetic skyrmions.

### 2.2. The Magnetic Domains in TBCI

After obtaining the interlayer parameters from the SANNs and intralayer parameters from DFT calculations, we then constructed the full Hamiltonians of TBCIs. The Hamiltonian of a TBCI can be written as:

$$H = \sum_{i \neq j} S_i^+ \mathcal{J}_{ij} S_j + \sum_{i < j} S_i^+ \mathcal{J}^\prime_{ij} S_j + \sum_{i < j} J_{ij} S_i S_j,$$

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$_3$. We have signed all the Cr atoms in the upper layer by positive indices and those in the lower layer by negative indices. The first and second terms at the right side describes the intralayer exchange interactions for upper and lower layers, respectively. The matrices $\mathcal{J}_{ij}$ for the first, second and third nearest neighbors were obtained by DFT calculations, and the other intralayer magnetic exchange interactions were neglected. The matrices $\mathcal{J}_2$ are different in different coordinate systems. A TBCI with nonzero twist angle $\phi$ contains a rotated CrI$_3$ layer, where the matrix $\mathcal{J}_2$ were transformed to the new coordinate system by the rotation matrix, $\mathcal{J}_2'' = R^{-1}(\phi) \mathcal{J}_2 R(\phi)$. The third term describes the interlayer exchange interactions, whose value was predicted by the SANNs. It is well known that the local interlayer distance varies depending on the position in the Morié pattern and changes the strength of interlayer exchange interactions. We adopted a scheme to approximately estimate the interlayer distances by using a dipole-force based interpolation, and predicted more realistic interlayer exchange interactions. More details can be found in the Supporting Information.

We then calculated ground magnetic states by performing the Landau–Lifshitz–Gilbert (LLG) equation calculations. The calculation results show that the ground states are FM in most cases, except for the TBCIs with $\phi = 0^\circ$ or $60^\circ$, 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 upper layer and that of neighboring Cr atoms in the lower layer. The minimum angle is always $0^\circ$, which shows that the FM domains always exist. The maximum angle as a function of the twist angle is shown in Figure 2b, where the angle increases significantly when $|\phi| < 5.1^\circ$ or $|\phi - 60^\circ| < 7.3^\circ$, and reaches $180^\circ$ at $|\phi| \approx 1.8^\circ$ or $|\phi - 60^\circ| \approx 2^\circ$. These two kinds of TBCIs are different in their interlayer exchange fields (defined as $E_i = -\frac{9}{4} \sum_j J_{ij}$).

The AFM exchange field centers in the TBCI with $\phi = 0^\circ$ form a kagome lattice (Figure 2c), while those $\approx 60^\circ$ form a triagonal lattice (Figure 2g). Thus, we name these TBCIs as K-TBCI and T-TBCI, respectively. Then, the phase diagram of TBCI is composed of uniform FM phase, T-phase, and K-phase. In order to compare the two kinds of TBCIs in more detail, we studied a K-TBCI with $\phi = 1.7^\circ$ and a T-TBCI with $\phi = 61.7^\circ$. Their Morié lattices have the same period length, with 4564 magnetic atoms in each period. Their interlayer exchange fields are shown in Figure 2c.g; there are three squeezed AFM islands in a period of panel (c), where the maximum AFM (FM) exchange field is 2.55 (+1.26) meV; and only one disc-shaped AFM island in a period of panel (g), where the maximum AFM (FM) exchange field is 1.55 (+1.04) meV. 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 Figure 2d. We can see that the FM canal is only >30 Å wide, and the barrier as shown by shaddow is <0.3 meV Cr$^{-1}$. 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 (Figure 2h) along the paths show that there are very wide and deep barriers (the shadows). The barrier widths are $>150$ Å and the barrier peaks along $A_1 - A_2$ are $\approx 1.0$ meV Cr$^{-1}$, and those along $A_1 - A_3$ are up to $1.5$ meV Cr$^{-1}$. 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.

We then simulated the steady magnetic states for the K-TBCI with $\varphi = 1.7^\circ$ and the T-TBCI with $\varphi = 61.7^\circ$. 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 Figure 2e,f. 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 Figure 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{43-45} in contrast, the T-TBCIs have isolated AFM domains, which are in favor of skyrmions. Thus, even thou both K-phase and T-phase have co-existence of FM and AFM domains, their distributions are quite different. 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 Figure 2k–m. For $3.2^\circ < |\varphi - 60^\circ| < 7.3^\circ$, both the upper and lower 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 upper layer has a skyrmion while the lower layer has undistorted FM-order. The skyrmion may also exist in the lower layer, and instead, the upper layer has undistorted FM-order.

2.3. 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 Figure 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 Figure 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 $m_0$ 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

$$ m_x(r, \phi) = m_0 \frac{\cos(\phi_0 - c\phi)}{\cosh((r - R)/w)} $$

$$ m_y(r, \phi) = m_0 \frac{\sin(\phi_0 - c\phi)}{\cosh((r - R)/w)} $$

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

$$ c = \frac{1}{4\pi} \int n \cdot (\partial_n n \times \partial_n n) \, dx \, dy $$

where $n$ is the normalized magnetic moment $n = m/|m|$. By using this equation, the calculated skyrmion charges for Figure 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 Figure 3e, and both $R$ and the range of $c$ are dependent on $\varphi$. 

Figure 3. a) The skyrmions with $c = -2, -1, 0, 1, \text{ and } 2$ in a T-TBCI with $\varphi = 61.7^\circ$, the yellow and blue colors sign 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 with $m_0 \tanh((r - R)/w)$; c) the maximum skyrmion charge and the associated skyrmion radius as functions of Moré period length; d,e) the excitation energy and skyrmion edge width as functions of skyrmion charge.
As shown in Figure 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 $c = -3$, 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 Figure 3c, the $|c_{\text{max}}|$ increases linearly with increasing of Morié period length $a_M$, as $\varphi$ approaches 60°.

The excitation energy, defined as the energy difference between the system with a skyrmion and the system’s ground state, is shown in Figure 3d. For a constant $c$, the excitation energy decreases with $\varphi$ approaching 60°. 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 ~18000 atoms, the 8 meV is a very small amount of energy. From Figure 3d, we can see that the trivial skyrmion ($c = 0$) always has the lowest energy, and the excitation energy increases as $|c|$ increases.

### 2.4. 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. First, 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 charge is an integer number with many options. It is topologically protected, thus suitable to store information. Besides that, the skyrmion position can also be used to store information. As we have shown that a skyrmion in T-TBCI can stay either at the upper layer or at the lower layer and these two states are degenerate. Therefore, the position of a skyrmion can store 1 bit information. It is well known that the skyrmions in magnetic alloy films are free to move, and their positions can be controlled by an electric current. By contrast, the skyrmions in T-TBCI are pinned at the AFM domain. Thus, in T-TBCI, the skyrmions with the stored information can not be transported by an electric current. Fortunately, the in-plane of one CrI$_3$ layer can be used to transport the skyrmions. As shown in Figure 4a, a small left move of the upper CrI$_3$ layer would lead to a large up move of the skyrmion. Actually, the motion of the skyrmion is always perpendicular to and much faster than that of the CrI$_3$ layer. The theoretical analysis can be found in the Supporting Information.

Besides translation, the rotation can also be used to tune the skyrmion in T-TBCI. The rotation of one CrI$_3$ layer changes the twist angle $\varphi$, which alters the size of the Morié pattern. Then, the size of AFM domain, i.e., the size of skyrmion, is altered accordingly. As shown in Figure 4b, the clockwise rotation of the upper CrI$_3$ layer makes $\varphi$ deviate from 60°, and compresses the skyrmion. A skyrmion with smaller radius has a smaller maximum skyrmion charge $|c_{\text{max}}|$; therefore, the skyrmion charge $c$ may exceed $|c_{\text{max}}|$ during the compressing. Then, the skyrmion would collapse to another one with a lower $c$. The skyrmion would collapse completely if $|\varphi - 60^\circ| > 3.2^\circ$, where the TBCI would have a uniform FM state. 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 Figure 4c. It is composed of one large lower CrI$_3$ layer, and two smaller upper CrI$_3$ layers. The upper CrI$_3$ layer at emitter (E) region rotates slowly away from 60°. Then the skyrmions with high skyrmion charges at the lower CrI$_3$ layer will shrink their radiuses. The following collapses emit spin waves with wide range of frequencies along all in-plane directions. After that, these spin waves will be filtered by the skyrmion lattices at the grating (G) region. The twist angle of the G region is kept fixed, then the skyrmions at the lower layer form a steady lattice, which only allows spin wave with certain frequencies to pass through.

**Figure 4.** 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 upper layers and the triagonal skyrmion lattices (shadowy bubbles) at the lower layers.
In the fundamental research, the T-TBCI is a promising platform for the study of spin topology and dynamics. For example, by using the degenerate upper and lower positions of skyrmions, a variety of skyrmion lattices can be designed in T-TBCIs. As shown in Figure 4d,e, the skyrmions at the upper layers form honeycomb and kagome lattices respectively; and the both lower 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.

3. Conclusion

In summary, we have 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 CrI3 bilayer. Besides TBCI, these methods can also be used in the study of twisted CrI3 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.

4. Experimental Section

Artificial Neural Network: Two SANNs were constructed in Matlab to predict $J$ for any $r$, $\theta$, $z$, and $\varphi$ values. As it was discussed, $J_{AB}$ and $J_{AA}$ could be directly obtained from $J_{AA}$ and $J_{AB}$, therefore there was only needed to predict the values of $J_{AA}$ and $J_{AB}$. Due to their different symmetry properties, two SANNs were used for $J_{AA}$ and $J_{AB}$ separately. The two SANNs have the same structure as shown in Figure 1b. There were 18 weight shared feed forward neural networks (FFNNs) in a SANN. Each FFNN contained four hidden layers with $20 \times 60 \times 60 \times 60 \times 80 \times 80 \times 80 \times 40$ neurons, and 40 neurons showed the best prediction accuracy.

There were five input variables: $x = r \cos \theta$, $y = r \sin \theta$, $z$, $\cos \varphi$, and $\sin \varphi$. The use of $\cos \varphi$ and $\sin \varphi$ rather than $\varphi$ naturally assured the periodicity of $J$ on $\varphi$. Data points of 5600 DFT+MFT were used for each SANN. The number of data points were extended to 100,800 according to the symmetry operations. And 80% of them were randomly chosen to be the training dataset, and the remaining 20% data points to be the testing dataset. After training $1 \times 10^3$ scaled conjugate gradient (SCG) cycles, the neural networks could predict $J$ values in high accuracy. The regression accuracies for $J_{AA}$ and $J_{AB}$ were 99.98% and 99.95% with squared mean error 0.007 and 0.011 meV, respectively. In the real applications, the same settings were adopted, and all the DFT+MFT data point were used as the training dataset.

Magnetic Structure Calculation: After obtained the interlayer parameters from the SANNs and intralayer parameters from DFT calculations, magnetic properties of the system were then calculated.

The equation of motion describing such a classical spin model is Landau–Lifshitz–Gilbert (LLG) equation

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

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 $-\gamma S_i$ in the last term works as an effective magnetic field.

The steady magnetic structures for K-TBCI and T-TBCI were obtained by random initial magnetic configurations. In order to exhaust all different skyrmions, variety of vortex-shaped initial magnetic configurations were also used.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements

The author would like to thank Dr. Chen Si, Associate Professor of Beihang University, for the discussion on the neural networks; Dr. Jize Zhao, Professor of Lanzhou University, for the discussion on spin lattice models of CrI3 layer; and Dr. Yue Ji, Assistant Professor of Beijing Institute of Technology, for language editing. This work was financially supported by National Natural Science Foundation of China (Grants Nos. 12022415 and 11974056).

Conflict of Interest

The author declares no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available in the supplementary material of this article.

Keywords

CrI3, magnetic skyrmions, moiré patterns, twisted bilayers

Received: June 17, 2022
Revised: October 13, 2022
Published online: November 28, 2022

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