Magnetic domain engineering in bilayer CrI$_3$ by applying external electric fields

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Motivated by manipulating the magnetic order of bilayer CrI$_3$, we carry out microscopic calculations to find the magnetic order and various magnetic domains of the system in the presence of an electric field. Making use of density functional simulations, a spin model Hamiltonian is introduced consisting of isotropic exchange couplings, Dzyaloshinskii-Moriya (DM) interaction, and on-site magnetic anisotropy. The spin dynamics of two well-known states of bilayer CrI$_3$, low temperature (LT) and high temperature (HT) phases, are obtained by solving the Landau-Lifshitz-Gilbert equation. We show the magnetic texture is stacking-dependent in bilayer CrI$_3$ and stable magnetic domains can appear in the HT stack which are tunable by external electric and magnetic fields. Therefore, we suggest that the HT phase represents a promising candidate for data storage in the modern generation of spintronic devices which work based on magnetic domain engineering.

I. INTRODUCTION

Two dimensional (2D) CrI$_3$ structures as a flagship of synthesized 2D magnets [1–6] have been explored by theoretical and experimental researchers to investigate their attractive properties. Not long ago, it has been reported that CrI$_3$, in monolayer and few layers forms, can represent a promising candidate as the modern generation of memory devices owing to their sensitive to the electric field. The Ising Hamiltonian represents a standard model to explain spin canting physics. A Heisenberg Hamiltonian with Kitaev or DM interactions has been used to describe the spin wave in a CrI$_3$ [25] structure and a small amount of the DM interactions in 2D CrI$_3$ is responsible for a topological gap [24].

A vital question here is that can we manipulate and tune magnetic domains of 2D magnets to make them applicable in technology and spintronics? To resolve this specific question, we focus on the bilayer CrI$_3$ in the presence of an external electric field [10, 13, 16, 19, 26].

Here, we use a spin model Hamiltonian; containing isotropic exchange and DM interactions, in the presence of external fields. We invoke density functional theory (DFT) based calculations to extract a suitable spin model Hamiltonian of bilayer CrI$_3$ that reproduces accurate and viable magnetic properties of the system. It should be worth mentioning that the isotropic exchange coupling parameters of bilayer CrI$_3$ have been explored in the presence of an external electric field, but Dzyaloshinskii-Moria (DM) interaction has been unexplored so far. Our calculations show the DM interaction is strongly related to the applied electric field in the HT bilayer CrI$_3$ contrary to that we obtained for a monolayer CrI$_3$ [23].

The spin dynamics of the system are computed by making use of the Landau-Lifshitz-Gilbert formalism [27] and the DFT-obtained exchange parameters as the initial inputs. We furthermore use Monte Carlo Metropolis algorithm with $10^6$ time steps for a $50 \times 50$ nm$^2$ sheet to obtain temperature-dependent magnetization.

Our results show the weak strength of the isotropic intralayer exchange coupling denotes the magnetic order of the layers and can be tuned by the external electrical field. The results also show the electric field changes the magnetic texture of the system as same as a magnetic field. Moreover, the magnetic order temperature of the HT bilayer is increased by the number of layers [1]. The Néel temperature of 45 K is reported for the HT bilayer [19] and it is expected that the LT bilayer maintains a larger Curie temperature due to the strong FM ordering between the layers [20].

The Ising Hamiltonian represents a standard model to explain the magnetic behavior of 2D CrI$_3$, but it cannot provide a reasonable explanation of the observed spin wave in experiments [21]. Subsequently, an extended Heisenberg Hamiltonian with Kitaev [22], biquadratic anisotropy exchange term [8], or Dzyaloshinskii-Moria (DM) [23, 24] interaction terms were proposed to explain spin canting physics. A Heisenberg Hamiltonian with Kitaev or DM interactions has been used to describe the spin wave in a CrI$_3$ [25] structure and a small amount of the DM interactions in 2D CrI$_3$ is responsible for a topological gap [24].

A vital question here is that can we manipulate and tune magnetic domains of 2D magnets to make them applicable in technology and spintronics? To resolve this specific question, we focus on the bilayer CrI$_3$ in the presence of an external electric field [10, 13, 16, 19, 26].

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the intralayer exchange coupling to stabilize it in the system, therefore, the obtained magnetic domain is unstable and disappears rapidly. This paper is organized as follows. We commence with a description of our theoretical formalism in Sec. II, followed by the details of the DFT simulations, the spin model Hamiltonian and spin dynamics. Numerical results for the spin-spin interaction parameters in the presence of an electric field, the exchange coefficients in both the HT and LT phases and spin dynamics in two cases are reported in Sec. III. We summarize our main findings in Sec. IV.

II. THEORY AND MODELS

A. DFT calculations

We use Quantum Espresso package [28] to simulate density-functional based calculations. The generalized gradient exchange-correlation within Perdew-Burke-Ernzerhof [29] functional is considered to expose the electronic and magnetic ground states of two bilayer CrI$_3$ structures. An energy cut-off of 50 Ry for wave vector and an $8 \times 8 \times 1$ grid mesh of $k$-points within the first Brillouin zone are defined as the converged input parameters. To avoid repeats effects, we consider a vacuum of 20 Å along the z-direction. The van der Waals Grimme-D2 [30] correction is used to consider the interaction between layers. DFT+$U$ calculations [31] are also needed to find correct magnetic ground states of CrI$_3$ bilayers. At this point, we consider $U = 3$ eV as the on-site Hubbard parameter. To obtain a reliable total ground-state energy, we maintain a high degree of accuracy of $10^{-10}$ eV. The bilayers are relaxed until the maximum force on each atom is 0.01 eV/Å even under the electric field conditions. It should be noted that the consideration of the spin-orbit coupling (SOC) and noncollinear spin-polarization are essential to obtain the DM interaction and magnetic anisotropic energy (MAE).

B. Spin-model Hamiltonian

To provide spin-spin interactions in a magnetic 2D CrI$_3$ bilayer system, a model spin Hamiltonian for a 2D magnetic hexagonal lattice in each layer is used:

$$\mathcal{H} = \sum_{i,j} \left[ \frac{1}{2} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \gamma_i |S_{iz}|^2 + \frac{1}{2} D_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j) + \sum_i \mu_B \mathbf{B}_{ext} \cdot \mathbf{S}_i \right],$$  \hspace{1cm} (1)

where $J_{ij}$ is the isotropic exchange coupling parameter between $i$ and $j$ atoms, $\gamma_i$ denotes a magnetic anisotropy coefficient, $D_{ij}$ is the DM interaction vector and the last term shows the effect of an external magnetic field, $\mathbf{B}_{ext}$, on the Hamiltonian of the system and $\mu_B$ is the magnetic Bohr. The magnetic moment of $i^{th}$ Cr atom is indicated by $\mathbf{S}_i$ in Eq. 1. It is intriguing to compare the strength of the interlayer and intralayer exchange coefficient parameters which are respectively referred by $\nu$ and $t$ indices in this paper.

C. Spin Dynamics

Having identified the coefficients of the spin Hamiltonian, the Landau-Lifshitz-Gilbert (LLG) equation [27] is used to atomistic simulate the spin dynamics of the system. We follow the method introduced by Evans et al. [32] implemented in VAMPIRE code. The time evolution of the spin direction is given by LLG equation according to

$$\frac{\partial \mathbf{S}_i}{\partial t} = -\frac{\gamma}{1 + \lambda^2} [\mathbf{S}_i \times \mathbf{H}_{eff}^i + \lambda \mathbf{S}_i \times (\mathbf{S}_i \times \mathbf{H}_{eff}^i)],$$  \hspace{1cm} (2)

where $\gamma$ is the gyromagnetic ration and $\lambda$ is microscopic damping equal to unity for finding the equilibration states quickly. The effective field applied on each spin is defined by $\mathbf{H}_{eff}^i = -\partial \mathcal{H}/\partial \mathbf{S}_i + \mathbf{H}_{th}^i$ and the effective thermal field, $\mathbf{H}_{th}^i$, is counted by Langevin dynamic approach [33] to consider thermal spin fluctuations.

In order to obtain the dynamical spin configuration of the bilayer, a $100 \times 100$ nm$^2$ sheet is considered and the long-range demagnetization field is also considered in the calculations. The magnetic dipole-dipole interaction induces local spin-flip in the magnetic materials and leading to the creation of complicated spin texture even in the materials with a FM ground state [34, 35]. We discretize the sheet to macrocells with 1 nm$^3$ volume to avoid expenses of the demagnetization field computing at the atomistic level. The magnetic moments

![FIG. 1: (Color online) The top view (a) and (c) and side view (b) and (d) of the HT and LT bilayer CrI$_3$ structures, respectively. The black lozenges show the unit cell of bilayers. The blue (pink) circles represent chromium (iodine) atoms. In (b) and (d), chromium atoms are labeled by 1-4 in the unit cell. In (b) the bonding angles between the atoms of each layer are shown by $\alpha$, $\beta$ and $\theta$. The crystal parameters are given in Table I.](image-url)
of macromolecules, \( m_{nc} \), are defined by the sum of atomic magnetic moments within the macromolecule and the demagnetization field of \( i^{th} \) macromolecule is given by

\[
H^{mci}_d = \frac{\mu_0}{4\pi} \sum_{ij} M_{ij} \cdot m_{nc}^j - \frac{\mu_0}{3} \frac{m_{nc}^i}{V_{ic}^{mc}}
\]

where \( V_{ic}^{mc} \) is the volume of \( i^{th} \) macromolecule, \( \mu_0 = 4\pi \times 10^{-7} \) is the primitivity of the free space and \( M_{ij} \) is the dipole-dipole interaction matrix between \( i \) and \( j \) macromolecules defined as [32].

\[
M_{ij} = \begin{bmatrix}
3x_i - \frac{1}{3} & 3x_i y_j & 3x_i z_j
3x_j y_i & 3y_j - \frac{1}{3} & 3y_j z_i
3x_j z_i & 3y_j z_i & 3z_j - \frac{1}{3}
\end{bmatrix},
\]

where the positions of macromolecules are obtained by the magnetic center of the mass relation and the vector between the position of \( i \) and \( j \) macromolecules is shown by \( r_{ij} = r_{ij}(x_i + y_j + z_j) \). We consider that the sheet is linearly cooled in 2 ns from an equilibrium temperature, upper the Curie temperature, to zero Kelvin. To obtain temperature dependent magnetization, we use Monte Carlo Metropolis algorithm with \( 10^6 \) time steps for a 50×50 nm\(^2\) sheet.

III. NUMERICAL RESULTS AND DISCUSSIONS

In this section, we initially present the magnetic ground state, the symmetric and asymmetric exchange coefficients, and temperature-dependent magnetization of bilayer CrI\(_3\) in both the HT and LT phases in the ground state. Afterward, the changes of the coefficients by an external electric field are investigated. Ultimately, we discuss the spin dynamics of both phases in the presence of the perpendicular electric field.

The HT phase of bilayer CrI\(_3\) is in the form of the monoclinic structure, while the LT bilayer possesses a rhombohedral crystal structure displayed in Figs. 1(a)-(d). The different crystal structures lead to the unique magnetic behaviors. Both the HT and LT phases are relaxed by spin-dependent DFT calculations and their optimized lattice constant, bonding lengths and angles are reported in Table I. The results show the bonding angles and lengths of both layers are as the same and the geometry of the bilayer does not change significantly by the electric field which is in good agreement with those results reported in Ref. [10]. To obtain a formation of the energy, we use the straight formula of \( E_v = E_v(\text{bilayer}) - n_{Cr}E(Cr) - n_I E(I) \) where \( E_v(\text{bilayer}) \) denotes the total energy of the bilayer CrI\(_3\), \( n_{Cr} \) is the number of the chromium (iodine) atoms in the unit cell, and \( E(Cr) \) and \( E(I) \) are the energy of free Cr and I atoms, respectively. The formation energies of the HT and LT bilayers are given in Table I indicating that both phases are energetically favored to occur.

A. Exchange coefficients in the HT phase

In the HT phase of bilayer CrI\(_3\), there are four Cr atoms in the unit cell; two in each layer. At zero external magnetic field, if the nearest neighbor interlayer and intralayer interactions are considered, the extended first two terms of the model Hamiltonian (Eq. 1) in the unit cell can be written as

\[
\mathcal{H} = \frac{1}{2}(\eta_v J_{iv}[\langle S_1 \cdot S_2 \rangle + \langle S_2 \cdot S_1 \rangle + \langle S_3 \cdot S_4 \rangle + \langle S_4 \cdot S_3 \rangle] + \eta_{1v} J_{1v}[\langle S_1 \cdot S_3 \rangle + \langle S_3 \cdot S_1 \rangle + \langle S_2 \cdot S_4 \rangle + \langle S_4 \cdot S_2 \rangle] + \eta_{2v} J_{2v}[\langle S_1 \cdot S_4 \rangle + \langle S_4 \cdot S_1 \rangle + \langle S_2 \cdot S_3 \rangle + \langle S_3 \cdot S_2 \rangle]) + \eta_{12v} S_1 S_2
\]

where \( \eta_v = 3 \) is the number of the nearest neighbors in each layer and \( \eta_{1v} = \eta_{2v} = 1 \) are the number of type-1 (between atoms numbered by \( 1 \) (2) and \( 3 \) (4) in Fig. 1 (b)) and type-2 (between atoms numbered by \( 1 \) (2) and \( 4 \) (3) in Fig. 1 (b)) neighbors in the interlayer. \( \eta = 4 \) is the number of Cr atoms in the unit cell. The exchange coupling coefficients are obtained by mapping the model Hamiltonian onto the DFT-based calculations which are computed for various magnetic configurations. For calculation of the isotropic exchange coefficient, 4-distinct spin configurations are considered as: FM \((S_1 = S k, S_2 = S k, S_3 = S k, S_4 = S k)\), AFM1 \((S_1 = S k, S_2 = S k, S_3 = -S k, S_4 = -S k)\), AFM2 \((S_1 = S k, S_2 = -S k, S_3 = -S k, S_4 = S k)\) and AFM3 \((S_1 = S k, S_2 = -S k, S_3 = S k, S_4 = S k)\) in which all the magnetic moments of Cr atoms are aligned in the \( z \)-direction and \( S = 3/2 \). In these configurations, the DM terms of Eq. 1 are vanished owing to the parallel magnetic moments of Cr atoms, and their total energies are obtained by

\[
E_{FM} = 2S^2(\eta_{1v} J_{1v} + \eta_{1v} J_{1v} + \eta_{2v} J_{2v}) + \eta_{12v} S^2,
\]

\[
E_{AFM1} = 2S^2(\eta_{1v} J_{1v} - \eta_{1v} J_{1v} - \eta_{2v} J_{2v}) + \eta_{12v} S^2,
\]

\[
E_{AFM2} = 2S^2(-\eta_{1v} J_{1v} - \eta_{1v} J_{1v} + \eta_{2v} J_{2v}) + \eta_{12v} S^2,
\]

\[
E_{AFM3} = 2S^2(-\eta_{1v} J_{1v} + \eta_{1v} J_{1v} - \eta_{2v} J_{2v}) + \eta_{12v} S^2.
\]

We carry out spin-dependent DFT calculations to find the total energies of the considered magnetic configurations and as a consequence the exchange coupling parameters of the HT bilayer are given by

\[
J_1 = \frac{(E_{FM} + E_{AFM1}) - (E_{AFM2} + E_{AFM3})}{8S^2\eta_v},
\]

\[
J_v = \frac{E_{FM} - E_{AFM1}}{4S^2\eta_v},
\]

where \( J_v = J_{1v} + J_{2v} \) is the interlayer exchange coupling and \( \eta_v = \eta_{1v} = \eta_{2v} \). The intralayer exchange coupling is \( J_1 = -7.00 \text{ meV} \) which presents the strong ferromagnetic coupling of Cr atoms in the layers. \( J_v = +0.08 \text{ meV} \) indicates the layers are coupled in a form of anti-ferromagnetic. Furthermore, the weak strength of the isotropic intralayer exchange coupling denotes the magnetic order of the layers can be tuned by the external electrical field [10]. We observe the variation of the isotropic interlayer and intralayer exchange
coupling by DFT-based calculations, shown in Fig. 2. It is observed that the sign of the interlayer exchange coupling is changed between 2.5 V/\text{nm}, hence a magnetic transition from the AFM to the FM appears between the layers while the strength of the intralayer exchange coefficient is no longer altered by the electric field. It means the ferromagnetic coupling of the \text{Cr} atoms in the layer is extremely strong. The extended Hamiltonian for the DM term is given by

\begin{equation}
\mathcal{H}_{DM} = \frac{1}{2} [D_{\text{112}} \cdot (S_1 \times S_2) + D_{\text{112'}} \cdot (S_1 \times S_2') + D_{\text{112''}} \cdot (S_1 \times S_2'') + \ldots ] + \ldots,
\end{equation}

where $S_i'$ and $S_i''$ are equal to the magnetic moment of $i^{th}$ \text{Cr} atom, and $S_i' = S_i'' = S_i$. On the other hand, the DM vector between two atoms can be obtained by $D_{ij} = D_{ij} \mathbf{u}_{ij}$, where $D_{ij}$ is a scalar and $\mathbf{u}_{ij}$ is a unit vector in the direction of the line connecting $i$ and $j$ atoms [36]. Therefore, $D_{ij} = -D_{ji}$. $D_{112'} = R_y(120)D_{112}$, $D_{112''} = R_y(240)D_{112}$ and $R_y(\theta)$ is a $\theta$ degree rotation around the $z$-axis. Three first terms of Eq. 9 can be written as

\begin{equation}
\mathcal{H}_{DM12} = (D_{\text{112}} + R_y(120)D_{\text{112}} + R_y(240)D_{\text{112}}) \cdot (S_1 \times S_2).
\end{equation}

If $D_{112} = (D_{112}^x \hat{x} + D_{112}^y \hat{y} + D_{112}^z \hat{z})$, then $(D_{112} + R_y(120)D_{\text{112}} + R_y(240)D_{\text{112}}) = 3D_{112}^z \hat{k}$, hence only the $z$-component of the DM vector between two \text{Cr} atoms in the layers of the bilayer \text{CrI}_3 can be entered in the model Hamiltonian of Eq. 1. In the similar way, we can write $(D_{34} + R_y(120)D_{34} + R_y(240)D_{34}) = 3D_{34}^z \hat{k}$ for the DM interaction between \text{Cr} atoms in the top layer. Due to the symmetries between the layers, it is possible to consider $D_{112}^z = D_{34}^z = D_t^z$. On the other hand, $D_{121}$ and $D_{13} = -D_{131}$, hence for the first five lines of Eq. 9, we have the interlayer terms of the DM interaction as,

\begin{equation}
\mathcal{H}_{DMt} = \frac{1}{2} [6D_{t}^z \hat{k} \cdot (S_1 \times S_2) + 6D_{t}^z \hat{k} \cdot (S_3 \times S_4)].
\end{equation}

Therefore, the symmetries of the HT phase dictate that just the $z$-component of the intralayer DM interaction vector, $D_{t}^z$, has a non-zero value.

Moreover, the geometry of the bilayer crystal indicates that $u_{13} = u_{24}$ and $u_{32} = R_y(180)u_{14}$, then we can assume $D_{13} = D_{24}$ and $D_{32} = R_y(180)D_{41}$. To obtain the interlayer contribution of the DM vector, we have to focus on the last four lines of Eq. 9 which can be summarized in the following form,

\begin{equation}
\mathcal{H}_{DMl} = (D_{13} \cdot (S_1 \times S_3) + D_{13} \cdot (S_2 \times S_4) + D_{14} \cdot (S_1 \times S_4) + R_y(180)D_{41} \cdot (S_3 \times S_2),
\end{equation}

where $D_{13} = (D_{v1}^x, D_{v1}^y, D_{v1}^z), D_{14} = (D_{v2}^x, D_{v2}^y, D_{v2}^z)$ and $R_y(180)D_{41} = (-D_{v2}^x, D_{v2}^y, -D_{v2}^z)$. Eight different spin configurations are needed to obtain enough information about the interlayer and intralayer DM vectors given in Table II.

The DM interactions are calculated by mapping the considered spin configurations on the model Hamiltonian and comparing the spin-dependent DFT-obtained total energies. At this place, we want to focus on the DM terms described in

| Phase | $a$(\AA) | $h_{Cr-Cr}$(\AA) | $h_{I-Cr}$(\AA) | $d_{Cr-Cr}$(\AA) | $d_{Cr-I}$(\AA) | $\beta_{I-Cr-I}$($^\circ$) | $\alpha_{I-Cr-I}$($^\circ$) | $\theta_{Cr-Cr-Cr}$($^\circ$) | $E_f$ (eV/\text{per Cr}) |
|-------|----------|----------------|---------------|----------------|----------------|-----------------|----------------|-----------------|----------------|
| HT    | 6.977    | 7.24          | 3.99          | 2.81           | 4.03           | 177.4           | 89.9           | 91.6            | -8.81          |
| LT    | 6.979    | 6.59          | 3.35          | 2.81           | 4.03           | 177.6           | 89.9           | 91.7            | -8.86          |
of the HT bilayer is achieved by DFT-obtained total energies. In a similar way, we can find the DM terms of model Hamiltonian.

Eqs. 11 and 12.

\[
H_{DM1-2} = 6S[D_i^2 \cdot (\sin(\pi/6)\hat{k})],
\]

\[
H_{DM2-4} = -6S[D_i^2 \cdot (\sin(\pi/6)\hat{k})],
\]

\[
H_{DM6-1} = H_{DM6-2} = 0.
\]

By imposing the above relations into Eq. 1, the total energy equations are defined and the z-component of the intralayer DM vector is given by

\[
D_{i\perp} = \frac{E_{1z} - E_{2z}}{12S^2\sin(\pi/6)}.
\]

In a similar way, we can find the DM terms of model Hamiltonian for the other configurations and the interlayer DM vector of the HT bilayer is achieved by DFT-obtained total energies.

We obtain \(D_{i\perp} = 1.1 \mu eV\), \(D_{i\parallel} = 3.5 \mu eV\), \(D_{i\parallel}^0 = -38.8 \mu eV\) and \(D_{\perp}^0 = 3.1 \mu eV\) for the HT bilayer. Accordingly, the intralayer DM interaction is ignorable in comparison with the interlayer one. The results show the strongest component of the interlayer DM vector is in the \(y\)-direction; this means the magnetic moments of Cr atoms in the layers tend to rotate along the \(x\)-\(z\) plane, as long as they align in the \(z\)-direction because of the high MAE of the bilayer.

The interlayer and intralayer DM interactions can be tuned by an external electric field, very similar to the isotropic exchange coupling. In Fig. 2 (b), it is observed that the intralayer exchange coupling is changed significantly by the electric field. Although the \(z\) and \(x\) components of the interlayer DM interactions are independent of the electric field, the sign and the value of \(D_{i\parallel}^0\) vary. The sign of the \(y\)-component of the DM changes from negative to positive values between 2-2.5 V/nm where a transition from the AFM to FM is achieved for the HT bilayer. In fact, a phase transition needs a spin-canting between layers in the \(x\)-\(z\) plane satisfied by the non-zero \(D_{i\parallel}^0\) which is in the same order of the \(J_o\) in the HT bilayer.

To calculate the magnetic order temperature, Curie \(T_C\), or Néel \(T_N\) temperature of the FM or AFM bilayer, a sample of \(50 \times 50\) nm\(^2\) is considered and the metropolis Monte Carlo algorithms are used for the calculation of temperature-dependent magnetization (see Fig. 3 (a)). According to Fig. 3 (b), the magnetic order temperature of the HT bilayer is increased by applying the electric field that arises from the increasing of the intralayer isotropic exchange coefficient.

The variation of \(MAE = (E_{\perp} - E_{\parallel})/\eta S^2\) as a function of the electric field is shown in Fig. 3 (b). It shows that the magnetic moments of the Cr atoms are in out-of-plane direction, and it is unchangeable by the external electric field.
B. Exchange coefficients in the LT phase

Now we turn our attention to another phase of the bilayer system. Four Cr atoms in a unit cell of the LT phase bilayer CrI₃ prefer to determine parallel spin-orientation in which the minimized total energy belongs to FM magnetic configuration [10]. Here, similar to the HT phase, we are interested in obtaining the symmetric and antisymmetric exchange coefficients of the LT bilayer. We should notice that the intralayer nearest neighbors of the LT is equivalent with that in the HT phase, while there is only one interlayer nearest neighbor bond between Cr atoms numbered by 2 and 3 in Fig. 1 (d). In the LT bilayer CrI₃ case, the first two terms of Eq. 1 are extended to be as

\[ \mathcal{H} = \frac{1}{2}(\eta_l J_l [(\mathbf{S}_1 \cdot \mathbf{S}_2) + (\mathbf{S}_2 \cdot \mathbf{S}_1)] + \eta_v J_v [(\mathbf{S}_2 \cdot \mathbf{S}_3) + (\mathbf{S}_3 \cdot \mathbf{S}_2)]) + \eta_l^2 (\mathbf{S}_2 - \mathbf{S}_3)^2, \]

where \( \eta_l = 3 \) and \( \eta_v = 1 \) are the number of the intralayer and interlayer nearest neighbors. We consider four different magnetic configurations of Cr atoms, as a FM (\( \mathbf{S}_1 = \mathbf{S}_k \), \( \mathbf{S}_2 = \mathbf{S}_k \), \( \mathbf{S}_3 = \mathbf{S}_k \), \( \mathbf{S}_4 = \mathbf{S}_k \)), AFM1 (\( \mathbf{S}_1 = \mathbf{S}_k \), \( \mathbf{S}_2 = -\mathbf{S}_k \), \( \mathbf{S}_3 = -\mathbf{S}_k \), \( \mathbf{S}_4 = \mathbf{S}_k \)), AFM2 (\( \mathbf{S}_1 = \mathbf{S}_k \), \( \mathbf{S}_2 = -\mathbf{S}_k \), \( \mathbf{S}_3 = \mathbf{S}_k \), \( \mathbf{S}_4 = \mathbf{S}_k \)) and AFM3 (\( \mathbf{S}_1 = \mathbf{S}_k \), \( \mathbf{S}_2 = -\mathbf{S}_k \), \( \mathbf{S}_3 = -\mathbf{S}_k \), \( \mathbf{S}_4 = \mathbf{S}_k \)) and thus able to find interlayer and intralayer symmetric exchange coefficients. By applying the spin vectors on Eq. 16, the relations for the total energies of the magnetic configurations are given by,

\[
\begin{align*}
E_{FM} &= E_0 + 2S^2 \eta_l J_l + S^2 \eta_v J_v + \eta_l^2 S^2, \\
E_{AFM1} &= E_0 + 2S^2 \eta_l J_l - S^2 \eta_v J_v + \eta_l^2 S^2, \\
E_{AFM2} &= E_0 - 2S^2 \eta_l J_l + S^2 \eta_v J_v + \eta_l^2 S^2, \\
E_{AFM3} &= E_0 - 2S^2 \eta_l J_l - S^2 \eta_v J_v + \eta_l^2 S^2.
\end{align*}
\]

The DFT-obtained total energies give us the symmetric exchange coefficients as

\[
\begin{align*}
J_l &= \frac{(E_{FM} + E_{AFM1}) - (E_{AFM2} + E_{AFM3})}{8S^2 \eta_l}, \\
J_v &= \frac{(E_{FM} - E_{AFM1})}{2S^2 \eta_v}.
\end{align*}
\]

The results displayed in Fig. 4 (a) show the interlayer and intralayer symmetric exchange couplings possess negative and consistent values with the FM ground state of the LT bilayers. The sign of \( J_l \) and \( J_v \) remain negative by applying the electric field, while their amounts are slightly increased. In fact, the electric field develops the ferromagnetic coupling of layers increase in the LT bilayer.

The third term of the Hamiltonian in Eq. 1 can be written for the LT bilayer as,

\[
\mathcal{H}_{DM} = \frac{1}{2} \left[ \mathbf{D}_{112} \cdot (\mathbf{S}_1 \times \mathbf{S}_2) + \mathbf{D}_{122} \cdot (\mathbf{S}_1 \times \mathbf{S}_2^\prime) + \mathbf{D}_{112}^\prime \cdot (\mathbf{S}_1 \times \mathbf{S}_2^\prime) + \mathbf{D}_{122}^\prime \cdot (\mathbf{S}_1 \times \mathbf{S}_2) + \mathbf{D}_{222} \cdot (\mathbf{S}_2 \times \mathbf{S}_2^\prime) + \mathbf{D}_{222} \cdot (\mathbf{S}_2 \times \mathbf{S}_2^\prime) + \mathbf{D}_{121} \cdot (\mathbf{S}_1 \times \mathbf{S}_1^\prime) + \mathbf{D}_{121} \cdot (\mathbf{S}_1 \times \mathbf{S}_1^\prime) + \mathbf{D}_{121}^\prime \cdot (\mathbf{S}_1 \times \mathbf{S}_1^\prime) + \mathbf{D}_{121}^\prime \cdot (\mathbf{S}_1 \times \mathbf{S}_1^\prime) + \mathbf{D}_{123} \cdot (\mathbf{S}_1 \times \mathbf{S}_3) + \mathbf{D}_{123} \cdot (\mathbf{S}_1 \times \mathbf{S}_3) + \mathbf{D}_{123} \cdot (\mathbf{S}_1 \times \mathbf{S}_3) + \mathbf{D}_{123} \cdot (\mathbf{S}_1 \times \mathbf{S}_3) + \mathbf{D}_{133} \cdot (\mathbf{S}_3 \times \mathbf{S}_3) + \mathbf{D}_{133} \cdot (\mathbf{S}_3 \times \mathbf{S}_3) + \mathbf{D}_{133} \cdot (\mathbf{S}_3 \times \mathbf{S}_3) + \mathbf{D}_{133} \cdot (\mathbf{S}_3 \times \mathbf{S}_3) + \mathbf{D}_{323} \cdot (\mathbf{S}_2 \times \mathbf{S}_3) + \mathbf{D}_{323} \cdot (\mathbf{S}_2 \times \mathbf{S}_3) + \mathbf{D}_{323} \cdot (\mathbf{S}_2 \times \mathbf{S}_3) + \mathbf{D}_{323} \cdot (\mathbf{S}_2 \times \mathbf{S}_3) \right].
\]

Similar to the HT phase, the symmetric of atomic positions dictates to have only the \( z \)-component of the interlayer DM interaction in the Hamiltonian which is given by Eq. 11. For the interlayer DM interaction of the LT phase

\[
\mathcal{H}_{DMv} = \mathbf{D}_{v23} \cdot (\mathbf{S}_2 \times \mathbf{S}_3).
\]

It should be noticed that there is a \( C_3 \) rotation axis on the bonding length between Cr atoms numbered by 2 and 3 in the unit cell of the LT bilayers (see Fig. 1 (d)). According to Moriya symmetry rules [37], the DM interaction between atoms 2 and 3 is parallel to the \( C_3 \) axis which is along the \( z \)-direction. Therefore, we need to find the \( z \)-component of the interlayer DM interaction which can be obtained by two spin configurations (see Table III). Finally, the \( z \)-component of intralayer and interlayer DM interactions are respectively given by

\[
\begin{align*}
D_{\ell}^z &= \frac{(E_{1st} - E_{2nd}) + (E_{3rd} - E_{4th})}{12S^2 \sin(\pi/6)}, \\
D_{v}^z &= \frac{E_{3rd} - E_{4th}}{2S^2 \sin(\pi/6)}.
\end{align*}
\]

The ignorable values of \( D_{\ell}^z \) = \(-0.4 \) \( \mu \)eV and \( D_{v}^z \) = \(-0.3 \) \( \mu \)eV are obtained using our spin-dependent DFT-based calculations. It should be noted that there is an inversion symmetry.
TABLE III: Four spin configurations for calculation of the interlayer and intralayer DM vectors of the LT bilayer CrI$_3$.

| Config. | 1$^\text{st}$ | 2$^\text{nd}$ | 3$^\text{rd}$ | 4$^\text{th}$ |
|---------|-------------|-------------|-------------|-------------|
| S$_1$   | $Si$        | $Si$        | $Si$        | $Si$        |
| S$_2$   | $S\cos(\frac{\pi}{6})+\sin(\frac{\pi}{6})l$ | $S\cos(\frac{\pi}{6})i−\sin(\frac{\pi}{6})j$ | $Si$        | $Si$        |
| S$_3$   | $Si$        | $Si$        | $S\cos(\frac{\pi}{6})i+\sin(\frac{\pi}{6})j$ | $S\cos(\frac{\pi}{6})i−\sin(\frac{\pi}{6})j$ |
| S$_4$   | $S\cos(\frac{\pi}{6})+\sin(\frac{\pi}{6})l$ | $S\cos(\frac{\pi}{6})i−\sin(\frac{\pi}{6})j$ | $S\cos(\frac{\pi}{6})i+\sin(\frac{\pi}{6})j$ | $S\cos(\frac{\pi}{6})i−\sin(\frac{\pi}{6})j$ |

in the center of 2 and 3 atoms bond in the LT bilayer, so $D^z_{\text{v}}$ should be zero, and therefore, there is no any DM interaction between the layers. We investigate the effect of the external electric field on the interlayer and intralayer DM interactions of the LT phase, and the results are announced in Fig. 4 (b). It is observed that the interlayer and intralayer DM interactions of the LT phase are increased by developing the electric field. It is interesting that the order of the DM interaction variation is similar for the both LT and HT phases, but their behaviors are different. In the presence of an electric field, just the $z$-component of the DM interaction of the LT bilayer increases; this means that the spin-canting are in the plane of Cr atoms. On the other hand, the interlayer symmetric exchange coupling, $J_{\text{v}}$, is much stronger than $D^z_{\text{v}}$. Subsequently, the FM coupling survives in a competition between the FM coupling of layers and in-plane canting of spins.

In the HT bilayer, the amount of the interlayer DM vector and interlayer symmetric exchange coupling are in the same order. In fact, they are in a close competition with each other and that it remains in the external electric field. It is intriguing that the both of them leads to a transition from the AFM to FM in the HT phase; the interlayer DM interaction is dominant in the $y$-direction which leads to the spin canting in the $x$-$z$ plane and, as a consequence, helps to the rotation of the spin-direction from the AFM to FM and also the sign of $J_{\text{v}}$ is changed by applying the electric field.

The temperature dependence of the magnetic moments of Cr atoms, $M(T)$ in the LT bilayer is illustrated in Fig. 5 (a). It is indicated that $M(T)$ is nearly electric field independent. The variation of the Currie temperature, $T_C$, by the electric field arises directly from the variation of $J_{\text{v}}$ and it is increased gradually in an intenser electric field (see Fig. 5 (b)). The MAE of the LT phase as a function of the electric field is shown in Fig. 5 (b). The MAE remains negative by varying the electric field so the easy axis of the LT bilayer is in the $z$-direction and its value does not change significantly.

It should be noted that the formation energy is $10^3$ times greater than one in the exchange couplings of bilayer CrI$_3$, therefore, it is possible to consider that the spin dynamics can be changed in the constant atomic positions.

C. Spin dynamics of the HT phase

In the HT bilayer, the AFM coupling between layers is dictated the spin dynamics of the system. The spin dynamics of the top and bottom layers are plotted in Fig. 6. In $T = 0.3$ K, it is observed that the bilayer is divided into two areas in which layers are coupled in a form of an anti-ferromagnetic. While the bottom layer has a magnetic moment in the $\pm z$-direction, the magnetic moment of the top layer is in the $-z$-direction and vice versa; this means that the AFM order is constant between the layers. This regularity is also established on the domain walls, where the perpendicular components of the magnetic moments, $M_{\perp}$, of two layers are in the opposite directions. The results of simulations show that the created magnetic texture is stable in size, but $M_{\perp}$ is slowly changed to be perpendicular to the domain wall by the time evolution shown in Figs. 6 (a)-(c). Thus, an ultrathin Neel-type domain wall appears on both the top and bottom layers. It is clear that the zero magnetic moments are obtained for the HT bilayer, but there is an interesting microscopic magnetic pattern on each layer.

We obtain the symmetric and asymmetric exchange coupling coefficients of the spin Hamiltonian depending on the external electric field, therefore, we explore the variation of the spin dynamics of the bilayer by the electric field shown in Figs. 7 (a)-(c). In $E_l = 1$ V/nm, the magnetic order of the HT bilayer is the AFM and $J_{\perp}$ and $J_{\parallel}$ are near to their values in the absence of the electric field case, while the value of $D^z_{\text{v}}$ is increased to 25.5 $\mu$eV, therefore the new spin texture is created (see Fig. 7 (b)). If we focus on one of the layers, we see that the wider magnetic domains in $E_l = 0$ case (Fig. 7 (a)) changes to two smaller quasi-circle magnetic domains.
FIG. 6: (Color online) The magnetic texture of the top and bottom layers of the HT bilayer in $E_l = 0 \text{ V/nm}$, $T = 0.3 \text{ K}$, $B = 0$ in (a) time $= 2 \text{ ns}$, (b) time $= 2.2 \text{ ns}$ and (c) time $= 3 \text{ ns}$. The blue (red) areas show where the magnetic moments of Cr atoms are in the -z(+z)-direction. $M_\parallel$ is represented by black arrows on the domain walls. The created magnetic texture is stable in size, but $M_\perp$ is slowly changed to be perpendicular to the domain wall by the time evolution.

with finite diameters (nearly 25 nm and 10 nm). Skyrmion patterns occur around the magnetic moments, while their chirality is different for the top and bottom layers. The places and sizes of magnetic domains are the same in the top and bottom layers, however, the direction of the magnetic moments are exactly opposite for inside and outside of the domain walls. We obtain these stable quasi-circle magnetic domains under the time evolution.

For the HT bilayer, the amount of the intralayer DM interaction reaches to $41 \mu \text{eV}$ by increasing the electric field to $E_l = 2 \text{ V/nm}$, and the interlayer isotropic exchange coupling is equal to $-20 \mu \text{eV}$. Although the value of $J_v$ is reduced, the negative sign shows the bilayer phase is still in the AFM. These changes lead to a new spin texture in the bilayer. In Fig. 7 (c), 30 nm-diameter quasi-circle magnetic domains are created in the both top and bottom layers with opposite spin orientation, so the AFM configuration of the bilayer is conserved. Most importantly, the diameter, position, number of the magnetic domain and the domain wall chirality are related to the external electric field. Furthermore, the magnetic texture of the HT bilayer can be manipulated by an external electric field owing to the variation of the DM and isotropic exchange coupling interactions. Therefore, stable magnetic domains are constituted in the HT bilayer that possess fascinating microscopic information, while the total magnetism is zero in the macroscopic level.

In $E_l = 3 \text{ V/nm}$, the layers are coupled in a form of a ferromagnetic. This phase transition is obviously observed in the spin dynamics simulation which both layers have the same spin orientation (see Fig. 8) and originates from the positive sign of $J_v$. On the other hand, the perpendicular-component of the magnetic moments of the top and bottom layers are similar on the domain walls. The spin dynamics simulations show that the obtained magnetic domain in the FM bilayer is still stable under the time evolution.

According to Figs. 9 (a)-(c), our simulation results show that the external magnetic field changes the spin dynamic of the bilayer. In the absence of an external electric field, Fig.9 (b) shows that there is an area with FM configurations in the AFM background of the bilayer. Indeed, the external magnetic field is in competition with the interlayer exchange cou-
FIG. 8: (Color online) The magnetic texture of the top and bottom layers of HT bilayer in $E_l = 3$ V/nm, $T = 0.3$ K, $B = 0$ in (a) time 2 ns. Notice that the magnetic domain in the FM bilayer is stable under the time evolution.

FIG. 9: (Color online) The spin dynamic results of the HT bilayer in $E_l = 0$ V/nm, (a) $B = 0$, $T = 0.3$ K and time = 2 ns, (b) $B = 0.5$ T, $T = 0$ K and time = 2 ns and (c) $B = 0.5$ T, $T = 0$ K and time = 3 ns. The external magnetic field is in competition with the interlayer exchange coupling and desire to bring the spin of the Cr atoms parallel to the external magnetic field.

Another example is the system when imposed under an external electric field, $E_l = 2$ V/nm. In $B_{ext} = 0.01$ T, the AFM pattern is completely disappeared and the magnetic moments of the Cr atoms will be parallel. In competition between exchange coupling coefficients and the Zeeman energy, the Zeeman energy of the strong magnetic field has conquered. In the presence of 0.01 T external magnetic field, the simulation results (Figs. 10(a)-(c)) show that the quasi-circle magnetic domain converts to a quasi-ellipse form, but the AFM configuration between the top and bottom layers are conserved because the applied magnetic field is not large enough to create a phase transition in the magnetic order of the bilayer. Interestingly, the obtained magnetic domain and almost domain wall does not change by the time evolution after cooling time (2 ns). Accordingly, a stable magnetic texture can be obtained and tuned by a weak external magnetic field, while the strong magnetic field leads to the magnetic phase transition in the bilayer.
D. Spin dynamic of the LT phase

In the LT bilayer, a strong negative interlayer exchange coupling leads to the creation of the FM magnetic configuration. The spin dynamic simulation, in Fig. 11 (a) shows two wide areas with opposite spin directions in each layer while the magnetic moments of the layers are actually analogous. In the boundary of the two areas, the perpendicular magnetic moments of the Cr atoms are dominated and their chirality is the same in both layers. In fact, a 1D spin-wave is created at the boundary and the results show that the chirality of the spin-wave is changed smoothly in time (compare Fig. 11 (a) and (b)). On the other hand, the created areas sizes are stable by the time evolution, so we can consider that the 1D in-plane spin-wave is metastable (due to the change of chirality). In the absence of an electric field, the ignorable DM interaction cannot create a magnetic domain, and the observed spin-wave is due to the demagnetization field. For clarity, we report the results of the spin dynamic simulation in the absence of a demagnetization field in Fig. 11 (c). It is obviously observed that the magnetic domain is disappeared and all the Cr atoms of both layers have parallel magnetic moments due to strong interlayer and intralayer isotropic exchange couplings of the LT bilayer CrI$_3$.

The interlayer and intralayer DM interactions are increased by applying the external electric field, while the sign of the exchange coupling and their coefficients values remain practically constant in the LT bilayer CrI$_3$. The spin dynamic simulations show the existence of a similar quasi-circle magnetic domain in the top and bottom layers in the presence of the electric field. Hence, the meta-stable magnetic domain is disturbed by increasing the DM interaction in $E_l = 1$ V/nm and $E_l = 2$ V/nm, see Figs. 12 (a)-(c). The results show that this quasi-circle magnetic domains are disappeared by cooling the system and the magnetic moments of the Cr atoms gained parallelized, see Fig. 13 for the LT bilayer under $E_l = 1$ V/nm. We can assume that the DM interactions leads to the creation of a quasi-circle domain in the layers, but its value is extremely smaller than the exchange coupling to stabilize it in the system, therefore, the obtained magnetic domain is unstable and disappears rapidly in the LT bilayer.

IV. CONCLUSION

The interlayer and intralayer isotropic exchange coupling coefficients and DM interactions are obtained by DFT-based calculations for bilayer CrI$_3$. We have shown that the interlayer DM interactions has an indispensable impact on the magnetic phase transition of the HT bilayer in the presence of an external electric field. The effects of the external electric field on the parameters of the spin-model Hamiltonian and temperature-dependent magnetic moments of bilayers are explored. The spin dynamics simulations show that the HT bilayer CrI$_3$ can represent a promising candidate for spintronic and logic memory applications. Most importantly, a stable magnetic domain in the HT bilayer CrI$_3$ which can be manipulated by the electric and magnetic fields can be constituted owing to the tunable interlayer and intralayer exchange coupling and DM interactions. On the other hand, the interlayer exchange coupling changes very slightly by the electric field, subsequently there is no stable magnetic domain in the LT bilayer CrI$_3$ under the external electric field. These finding can be explore by current experiments. Our results can be extended in a system when disorder and magnetic impurity are considered.

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FIG. 12: (Color online) The magnetic texture of the top and bottom layers of the LT bilayer in (a) $E_l = 0$, (b) $E_l = 1 \text{ V/nm}$ and (c) $E_l = 2 \text{ V/nm}$, $T = 0.3 \text{ K}$, $B = 0$ and time $= 2 \text{ ns}$. The existence of a similar quasi-circle magnetic domain in the top and bottom layers in the presence of the electric field are seen and the meta-stable magnetic domain is disturbed by increasing the DM interaction.

FIG. 13: (Color online) The time evolution of the top layer of the LT phase under $E_l = 1 \text{ V/nm}$, $T = 0 \text{ K}$ and $B = 0$. The bottom layer has a similar spin dynamic results due to the FM coupling of the layers.

[1] B. Huang, G. Clark, E. Navarro-Moratalla, D. R. Klein, R. Cheng, K. L. Seyler, D. Zhong, E. Schmidgall, M. A. McGuire, D. H. Cobden, et al., “Layer-dependent ferromagnetism in a van der Waals crystal down to the monolayer limit,” Nature 546, 270 (2017).

[2] Y. Deng, Y. Yu, Y. Song, J. Zhang, N. Z. Wang, Z. Sun, Y. Yi, Y. Z. Wu, S. Wu, J. Zhu, et al., “Gate-tunable room-temperature ferromagnetism in two-dimensional Fe 3 GeTe 2,” Nature 563, 94 (2018).

[3] C. Gong, L. Li, Z. Li, H. Ji, A. Stern, Y. Xia, T. Cao, W. Bao,
C. Wang, Y. Wang, et al., “Discovery of intrinsic ferromagnetism in two-dimensional van der waals crystals,” Nature 546, 265 (2017).

[4] M. Gibertini, M. Koperski, A. F. Morpurgo, and K. S. Novoselov, “Magnetic 2d materials and heterostructures,” Nature nanotechnology 14, 408 (2019).

[5] C. Gong and X. Zhang, “Two-dimensional magnetic crystals and emergent heterostructure devices,” Science 363 (2019), 10.1126/science.aav4450.

[6] J. Meseguer-Sánchez, C. Popescu, J. L. García-Muñoz, H. Luethens, G. Tanashvili, E. Navarro-Morallata, Z. Guguchia, and E. J. Santos, “Coexistence of structural and magnetic phases in van der Waals magnet CrI₃,” Nature communications 12, 1 (2021).

[7] A. Hubert and R. Schäfer, Magnetic domains: the analysis of magnetic microstructures (Springer Science & Business Media, 2008).

[8] D. A. Wahab, M. Augustin, S. M. Valero, W. Kuang, S. Jenkins, E. Coronado, I. V. Grigorieva, I. J. Vera-Marun, E. Navarro-Morallata, R. F. Evans, et al., “Quantum Rescaling, Domain Metastability, and Hybrid Domain-Walls in 2D CrI₃ Magnets,” Adv. Mater. 33, 2004138 (2021).

[9] L. Chen, J.-H. Chung, M. B. Stone, A. I. Kolesnikov, B. Winn, V. O. Garlea, D. L. Abernathy, B. Gao, M. Augustin, E. J. G. Santos, and P. Dai, “Magnetic field effect on topological spin excitations in CrI₃,” Phys. Rev. X 11, 031047 (2021).

[10] E. S. Morell, A. León, R. H. Miwa, and P. Vargas, “Control of magnetism in bilayer CrI₃ by an external electric field,” 2D Mater. 6, 025020 (2019).

[11] P. Jiang, C. Wang, D. Chen, Z. Zhong, Z. Yuan, Z.-Y. Lu, and W. Ji, “Stacking tunable interlayer magnetism in bilayer CrI₃,” Phys. Rev. B 99, 144401 (2019).

[12] H. H. Kim, B. Yang, S. Li, S. Jiang, C. Jin, Z. Tao, G. Nichols, F. Sfigakis, S. Zhong, C. Li, et al., “Evolution of interlayer and intralayer magnetism in three atomically thin chromium trihalides,” Proceedings of the National Academy of Sciences 116, 11131 (2019).

[13] C. Lei, B. L. Chittari, K. Nomura, N. Banerjee, J. Jung, and A. H. MacDonald, “Magnetoelectric Response of Antiferromagnetic CrI₃ Bilayers,” Nano Lett. 21, 1948 (2021).

[14] T. Song, Z. Fei, M. Yankowitz, Z. Lin, Q. Jiang, K. Hwangbo, Q. Zhang, B. Sun, T. Taniguchi, K. Watanabe, et al., “Switching 2D magnetic states via pressure tuning of layer stacking,” Nat. Mater. 18, 1298 (2019).

[15] T. Li, S. Jiang, N. Sivadas, Z. Wang, Y. Xu, D. Weber, J. E. Goldberger, K. Watanabe, T. Taniguchi, C. J. Fennie, et al., “Pressure-controlled interlayer magnetism in atomically thin CrI₃,” Nat. Mater. 18, 1303 (2019).

[16] S. Jiang, J. Shan, and K. F. Mak, “Electric-field switching of two-dimensional van der Waals magnets,” Nat. Mater. 17, 406 (2018).

[17] S. Jiang, L. Li, Z. Wang, K. F. Mak, and J. Shan, “Controlling magnetism in 2d CrI₃ by electrostatic doping,” Nature nanotechnology 13, 549 (2018).

[18] I. Kashin, V. Mazurenko, M. Katsnelson, and A. Rudenko, “Orbitally-resolved ferromagnetism of monolayer CrI₃,” 2D Mater. 7, 025036 (2020).

[19] B. Huang, G. Clark, D. R. Klein, D. MacNeill, E. Navarro-Morallata, K. L. Seyler, N. Wilson, M. A. McGuire, D. H. Cobden, D. Xiao, et al., “Electrical control of 2D magnetism in bilayer CrI₃,” Nat. nanotechnol. 13, 544 (2018).

[20] D. Wang and B. Sanyal, “Systematic study of monolayer to trilayer CrI₃: Stacking sequence dependence of electronic structure and magnetism,” J. Phys. Chem. C 125, 18467 (2021).

[21] L. Chen, J.-H. Chung, B. Gao, T. Chen, M. B. Stone, A. I. Kolesnikov, Q. Huang, and P. Dai, “Topological Spin Excitations in Honeycomb Ferromagnet CrI₃,” Phys. Rev. X 8, 041028 (2018).

[22] I. Lee, F. G. Utermohlen, D. Weber, K. Hwang, C. Zhang, J. van Tol, J. E. Goldberger, N. Trivedi, and P. C. Hammel, “Fundamental spin interactions underlying the magnetic anisotropy in the Kitaev ferromagnet CrI₃,” Phys. Rev. Lett. 124, 017201 (2020).

[23] S. I. Vishayki, Z. Torbatian, A. Qaimzadeh, and R. Asgari, “Strain and electric-field control of spin-spin interactions in monolayer CrI₃,” Phys. Rev. Mater. 4, 094004 (2020).

[24] R. Jaeschke-Ubieiro, E. S. Morell, and A. S. Nunes, “Theory of magnetism in the van der Waals magnet CrI₃,” Phys. Rev. B 103, 174410 (2021).

[25] L. Chen, J.-H. Chung, T. Chen, C. Duan, A. Schneidewind, I. Radelnytsky, D. J. Voneshen, R. A. Ewings, M. B. Stone, A. I. Kolesnikov, et al., “Magnetic anisotropy in ferromagnetic CrI₃,” Phys. Rev. B 101, 134418 (2020).

[26] S. Jiang, L. Li, Z. Wang, K. F. Mak, and J. Shan, “Controlling magnetism in 2D CrI₃ by electrostatic doping,” Nat. Nanotechnol. 13, 549 (2018).

[27] M. O. Ellis, R. F. Evans, T. A. Ostler, J. Barker, U. Axtioli, O. Chubykalo-Fesenko, and R. W. Chantrell, “The Landau–Lifshitz equation in atomistic models,” Low Temperature Physics 41, 705 (2015).

[28] P. Giannozzi, S. Baroni, N. Bonini, M. Calandra, R. Car, C. Cavazzoni, D. Ceresoli, G. L. Chiarotti, M. Cococcioni, I. Dabo, et al., “QUANTUM ESPRESSO: a modular and open-source software project for quantum simulations of materials,” J. Phys. Condens. Matter. 21, 395502 (2009).

[29] J. P. Perdew, K. Burke, and M. Ernzerhof, “Generalized gradient approximation made simple,” Phys. Rev. Lett. 77, 3865 (1996).

[30] S. Grimes, “Semimempirical GGA-type density functional constructed with a long-range dispersion correction,” J. Comput. Chem. 27, 1787 (2006).

[31] M. Cococcioni and S. De Gironcoli, “Linear response approach to the calculation of the effective interaction parameters in the LDA+U method,” Phys. Rev. B 71, 035105 (2005).

[32] R. F. Evans, W. J. Fan, P. Chureemart, T. A. Ostler, M. O. Ellis, and R. W. Chantrell, “Atomistic spin model simulations of magnetic nanomaterials,” J. Phys. Condens. Matter. 26, 103202 (2014).

[33] W. Brown, “Thermal fluctuation of fine ferromagnetic particles,” IEEE Trans. Magn. 15, 1196 (1979).

[34] Y. Kawaguchi, H. Saito, and M. Ueda, “Can spinor dipolar effects be observed in Bose-Einstein condensates?” Phys. Rev. Lett. 98, 110406 (2007).

[35] M. Ezawa, “Giant skyrmions stabilized by dipole-dipole interactions in thin ferromagnetic films,” Phys. Rev. Lett. 105, 197202 (2010).

[36] H. Yang, A. Thiaviile, S. Rohart, A. Fert, and M. Chshiev, “Anatomy of dzyaloshinskii-moriya interaction at Co/Pt interfaces,” Phys. Rev. Lett. 115, 267210 (2015).

[37] T. Moriya, “Anisotropic superexchange interaction and weak ferromagnetism,” Phys. Rev. 120, 91 (1960).