Moiré-driven multiferroic order in twisted CrCl$_3$, CrBr$_3$ and CrI$_3$ bilayers

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Layered van der Waals materials have risen as a powerful platform to engineer artificial competing states of matter. Here we show the emergence of multiferroic order in twisted chromium trihalide bilayers, an order fully driven by the moiré pattern and absent in aligned multilayers. Using a combination of spin models and ab initio calculations, we show that a spin texture is generated in the moiré supercell of the twisted system as a consequence of the competition between stacking-dependent interlayer magnetic exchange and magnetic anisotropy. An electric polarization arises associated with such a non-collinear magnetic state due to the spin-orbit coupling, leading to the emergence of a local ferroelectric order following the moiré. Among the stochiometric trihalides, our results show that twisted CrBr$_3$ bilayers give rise to the strongest multiferroic order. We further show the emergence of a strong magnetoelectric coupling, which allows the electric generation and control of magnetic skyrmions. Our results put forward twisted chromium trihalide bilayers, and in particular CrBr$_3$ bilayers, as a powerful platform to engineer artificial multiferroic materials and electrically tunable topological magnetic textures.

Multiferroic materials display more than one ferroic order at the same time [1–3], and in particular, they can simultaneously host magnetic and ferroelectric orders. The existence of multiple symmetry-breaking orders allows having a coupling between electric and magnetic degrees of freedom [4]. Over the last two decades, a variety of multiferroic bulk compounds has been demonstrated [5–8], providing alternative strategies for multifunctional devices [7, 9]. However, focusing on the realm of two-dimensional (2D) materials, purely 2D multiferroics have remained elusive until the recent demonstration of multiferroic order in NiI$_2$ [10, 11]. Beyond isolating individual multiferroic monolayers, a potential alternative strategy to realize multiferroic order in van der Waals materials relies on artificially engineering it from originally non-multiferroic monolayers [12]. The electric control of magnetism provided by van der Waals multiferroics would open radically new ways of controlling artificial van der Waals matter, including Chern insulators [13–16], unconventional and topological superconductors [17–22] and correlated quantum magnets [23–25].

The weak bonding between layers in van der Waals materials allows combining monolayer layers in twisted heterostructures. Monolayer 2D materials can display different symmetry-breaking orders [11, 26–28], constituting a family of minimal building blocks that can be used to artificially engineer other emergent orders. This strategy has been widely exploited to engineer moiré correlated and topological states using 2D materials [13, 16, 17, 29–33]. Recently, this strategy has been extended to 2D magnetic materials including chromium trihalides, leading to a variety of twist-induced magnetic orders [34–40]. However, using twist engineering to realize a multiferroic order has so far remained unexplored.

Here we demonstrate the emergence of a multiferroic state in the family of twisted chromium trihalide CrX$_3$ (X=Cl, Br and I) bilayers by combining first principles and effective spin Hamiltonians. We first show the emergence of a non-collinear spin texture due to the modulation of the interlayer exchange coupling in the moiré unit cell. Associated with the spin texture an electric polarization emerges as a consequence of spin-orbit coupling (SOC) and the local magnetic non-collinearity in the moiré domains. Using ab initio calculations we extract the value of the electric polarization driven by non-collinear magnetic texture, and demonstrate its dependence on the halide of CrX$_3$. Furthermore, we analyze the emergent magnetoelectric coupling, demonstrating how it allows to directly electrically drive transitions between different spin textures employing an interlayer bias.

We start our analysis by describing the magnetic order that emerges in twisted CrX$_3$ bilayers. The magnetic behavior of CrX$_3$ monolayers can be described by a spin Hamiltonian in a honeycomb lattice

$$\mathcal{H} = -\frac{J}{2} \sum_{<i,j>} \mathbf{S}_i \cdot \mathbf{S}_j - \frac{A_v}{2} \sum_{<i,j>} S^z_i S^z_j - A_u \sum_i (S^z_i)^2 + \mathcal{V},$$

(1)

where $J$ is the first neighbor intralayer ferromagnetic exchange, taking a value on the order of 2-3 meV [41–43]. $A_v$ is the anisotropic magnetic exchange. $A_u$ is the single-ion anisotropy, which is dominated by $A_v$ in CrI$_3$, CrBr$_3$, and competing with $A_v$ in CrCl$_3$ [44]. Our analysis will focus on the anisotropy regime leading to out-of-plane easy axis, that corresponds to CrI$_3$, CrBr$_3$ and slightly strained CrCl$_3$ [45] [46]. The term $\mathcal{V}$ contains additional contributions that do not qualitatively affect our analysis, including Kitaev exchange [47], biquadratic exchange [48], Dzyaloshinskii-Moriya interaction [49], and dipolar coupling [50].

Considering now two layers of CrX$_3$ stacked together an interlayer magnetic exchange $J_M$ will arise. A moiré pattern like the one shown in Fig. 1a emerges for twist angles lower than 3°. Depending on the stacking between
layers two different regions can be distinguished, monoclinic and rhombohedral. Associated with these different regions the sign of $J_M$ will change, i.e., $J_M$ is ferromagnetic (positive) in the rhombohedral stacking and antiferromagnetic (negative) in the monoclinic one\cite{51–53}. This leads to a modulation of $J_M$ in the moiré supercell like the one shown in Fig. 1b\cite{34–37, 54}. Therefore, to model the twisted CrX$_3$ bilayer we add the interlayer interaction to the Hamiltonian of Eq. (1) as

$$\mathcal{H}_{\text{Inter}} = -\frac{1}{2} \sum_{i,j} J_M(\mathbf{r}_i, \mathbf{r}_j) \mathbf{S}_i \cdot \mathbf{S}_j, \quad (2)$$

where $J_M(\mathbf{r}_i, \mathbf{r}_j)$ is the site-dependent interlayer exchange\cite{55}. Since CrX$_3$ is composed of Cr$^{3+}$ with a spin state $S=3/2$, we can solve in a classical way the spin Hamiltonian for the twisted system. The ground state magnetic order is depicted in Fig. 2a. We can see that a non-collinear magnetic texture emerges between ferromagnetic and antiferromagnetic regions. In the presence of spin-orbit coupling, non-collinear magnetism leads to the emergence of an electric polarization $\mathbf{P}_{ij}$ between neighboring spins separated by a distance $\mathbf{r}_{ij}$ of the form\cite{56, 57}

$$\mathbf{P}_{ij} = \lambda_{\text{SOC}} (\mathbf{r}_{ij} \times (\mathbf{S}_i \times \mathbf{S}_j)), \quad (3)$$

where $\lambda_{\text{SOC}}$ is a coefficient that controls the strength of the spin-orbit coupling. From Eq. (3) we can clearly see the requirement of non-collinearity and the presence of SOC to produce an electric polarization. For the emerging ground state spin texture of the twisted system (Fig. 2a), the associated electric polarization when SOC is introduced is shown in Fig. 2b, with $P_z$ the dominant component. From Figs. 2ab, we can observe that an electric polarization emerges in both layers in the areas where the non-collinear spin texture occurs. The polarization emerges locally, and for the ground state spin texture the net electric polarization is zero. Therefore, to analyze the strength of the multiferroic order as a function of the parameters that appear in the spin Hamiltonian, we will consider an average of the electric polarization moduli $\overline{P}$. The moduli of the electric polarization in the moiré system associated with the ground state spin texture can be seen in Fig. 1c. We can observe there, that the ground state breaks the $C_6$ symmetry of the original system leading to a $C_2$ symmetry. Figure 2c shows the average polarization as a function of the maximum interlayer exchange $J_M^{\max}$. At low values, the twisted system behaves like two independent layers, remaining ferromagnetic, no spin texture emerges and consequently, there is no electric polarization. By increasing the interlayer coupling, non-collinearity appears and with it an associated electric polarization in virtue of Eq. (3). Our calculations are performed for values of $J_M^{\max}/J = 0.1$, typical order of magnitude for the family of CrX$_3$\cite{34, 58}. Higher values such as those obtained by applying uniaxial pressure\cite{59–61} would produce a stronger multiferroic order. Figure 2d shows the average polarization as a function of the anisotropic exchange $A_v$. At high values, the twisted system tends to align the magnetic moments out of the plane. Therefore, no spin texture occurs and thus there is no electric polarization. This situation happens for $A_v/J > 0.1$, so CrI$_3$ would be on the verge of displaying a multiferroic behavior due to its strong uniaxial anisotropy.

We now demonstrate and quantitatively quantify using first-principles density functional theory calculations the emergence of an electric polarization due to the non-collinear order at the domain wall. Performing first-
principles calculations in a full twisted CrX₃ bilayer with spin-orbit coupling and non-collinear magnetism is well beyond the current computational capabilities. However, since the electric polarization arises locally, the ab initio analysis can be performed in a system like the one shown in Fig. 3a by imposing a magnetic texture in the CrX₃ layer like the one shown in Fig. 3b. We set the same out-of-plane spin texture to the three compounds to systematically extract the effect of the halide atom [62]. The associated polarization of a non-collinear texture given in eq. (3) can be rewritten in terms of an spin spiral propagation vector \( \mathbf{q} \) and the spin rotation axis \( \mathbf{e} = (0, -1, 0) \), leading to an electric polarization of the spin texture\[56, 57]\)

\[
\mathbf{P} = \lambda_{SOC}(\mathbf{q} \times \mathbf{e}).
\]  

To demonstrate the emergence of an electric polarization in the magnetic texture shown in Fig. 3b, we now perform calculations in two equivalent configurations (Fig. 3b) with the same spin rotation vector \( \mathbf{e} \), but with opposite spin propagation vector \( \mathbf{q} \). Therefore, an opposite electric polarization will emerge in each of the configurations. The emergence of the electric polarization in the spin texture is directly obtained by taking the difference between the two equivalent configurations. This procedure provides a direct methodology to extract electric polarization stemming from non-collinear magnetic order. \textit{Ab initio} density functional theory calculations\[63\] were carried out with the all-electron full-potential linearized augmented-plane-wave method, using a fully non-collinear formalism with spin-orbit coupling (SOC) as implemented in Elk\[64\]. We have used the local density approximation for the exchange-correlation functional\[65\]. The results presented are converged concerning all the parameters\[66\].

The emergence of an electric polarization is accompanied by a reconstruction of the electronic density \( \rho \). Therefore we can analyze the difference in the electronic density \( \delta \rho \) between both configurations. This is shown in Fig. 3c for the three chromium halides. We observe that the electronic reconstruction increases by taking a heavier halide. Thus, for the same spin texture, we can see that CrI₃ will produce the strongest ferroelectric polarization. This result is a consequence of the increase of the spin-orbit coupling when one goes down in the halide group, thus demonstrating the effective equations (3) and (4) governed by the SOC prefactor \( \lambda_{SOC} \).

The reconstruction of the electronic density will lead to the appearance of a ferroelectric force in the atoms in the spin texture in the direction of the emergent electric polarization\[10\]. Therefore, we can compute the force difference between both configurations in Fig. 3b to provide a direct quantification of the electric polarization in CrX₃. Figure 3d shows the force difference between both configurations. We can see that the forces emerge only in the Cr atoms in which the non-collinear magnetism is present. Moreover, we can see that the forces emerge on the z-direction, indicating the direction of the electric dipole, and as expected from the schematic in Fig. 3b.

We can quantify the dependence on the halide by taking the average of the force module among Cr-atoms for each of the compounds, as shown in 3d. Taking a heavier halide produces an increase in the ferroelectric force, as expected from the increase of the spin-orbit coupling. As a reference, NiI₂, a 2D multiferroic governed by this same mechanism of spin-orbit coupling and non-collinear magnetism, leads to ferroelectric forces of \( \approx 20 \text{ meV/Å} \)\[10\]. Therefore, this confirms that spin textures produced in
FIG. 3. (a) Unit cell used in the ab initio calculations, Cr atoms depicted in green, halide atoms omitted for clarity. (b) Equivalent non-collinear magnetic configurations with opposite spin propagation vector $\mathbf{q}$ and same helicity $\mathbf{e}$, and hence opposite electric polarization. (c) Electronic density difference $\delta \rho$ between both equivalent configurations in panel (b) for the three trihalides in the Cr-atoms plane. (d) Force difference between the configurations of the panel (b). A net ferroelectric force emerges in the z-direction in the Cr atoms where the magnetism is non-collinear. An average of the ferroelectric force for each trihalide shows the dependence of the electric polarization with the ligand.

twisted CrX₃ bilayers lead to the emergence of an electric polarization with strong magnetoelectric coupling.

We now elaborate on some conclusions on the multiferroic order in twisted CrX₃ bilayers considering together the results obtained from the low energy models and the ab initio calculations. On the one hand, ab initio density functional theory methods show that as the halide becomes heavier, the ferroelectric polarization becomes stronger. On the other hand, the low energy
model shows that the anisotropic exchange has a detrimental impact on the formation of the spin texture and hence on the multiferroic order. In particular, in CrI$_3$ the strong anisotropic exchange might partially quench the formation of a sizable non-collinear magnetic texture. In CrCl$_3$, the small SOC would yield a comparably weak ferroelectric polarization despite the formation of the non-collinear texture. Therefore, the optimal twisted bilayer yielding the strongest ferroelectric polarization would be CrBr$_3$, or ultimately, a bilayer of an intermediate stoichiometry between CrBr$_3$ and CrI$_3$.

Finally, we analyze the magnetoelectric coupling present in twisted CrX$_3$ bilayers. To do so, we now include in the low energy Hamiltonian a coupling to an external electric $E = (0, 0, E_z)$ field perpendicular to the twisted system in the $z$-direction:

$$\mathcal{H}_E = \frac{1}{2} \sum_{ij} \mathbf{E} \cdot \mathbf{P}_{ij}. \tag{5}$$

We now show how this interlayer bias allows controlling the magnetic order due to the emergent multiferroicity[67]. Figures 4a and 4b show the spin texture and the associated electric polarization at $E_z/J = 3$. We can observe that the ground state spin texture shown in Fig. 2a gets significantly modified due to the strong magnetoelectric coupling, leading to the formation of a magnetic skyrmion around the AA rhombohedral stacking in one of the layers[39, 68–74]. Interestingly, such a magnetic state features a non-zero total electric polarization in the $z$-direction. The evolution of the total electric polarization in the moiré supercell in the $z$-direction as a function of the external electric field is shown in Fig. 4c. The different bumps in $P_z$ as a function of the electric field indicate transitions to different non-trivial spin textures which are shown as insets in Fig. 4c. Starting at $E_z/J = 0$ in the ground state spin texture, at $E_z/J = \pm 3$ the magnetic skyrmion shown in Fig. 4ab arises. At $E_z/J = \pm 5$ a second pair of magnetic skyrmions emerge in the other layer around the AB/BA stacking regions. As a reference, $J \approx 3$ meV and $\lambda_{SOC} \approx 10^{-4}e$ in twisted CrBr$_3$ bilayers, implying that such magnetic transitions can be experimentally driven via gating at voltages of $1 \sim 10$ V[67]. This brings twisted CrX$_3$ bilayers as a promising platform for the electric control of non-trivial magnetic textures, and ultimately as a platform for magnetoelectric skyrmionics.

To summarize, we have demonstrated that twisted CrX$_3$ bilayers develop a multiferroic order due to the interplay between the moiré structure, non-collinear magnetic order, and spin-orbit coupling. Furthermore, we have shown that the multiferroic order is accompanied by a strong magnetoelectric coupling that allows to electrically control of non-trivial magnetic textures using an interlayer bias. Our results put forward a strategy to design a new family of artificial multiferroics with a strong magnetoelectric coupling based on twisted magnetic van der Waals materials.

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