Twisted double bilayers of an antiferromagnet show tunable moiré magnetism

Stacking a bilayer of chromium triiodide, a layered antiferromagnet, onto another with a twist angle gives rise to a moiré magnet with rich magnetic phases, including ferromagnetic and antiferromagnetic orders. The magnetic orders can be controlled through the twist angle, temperature and electrical gating, with the system also showing voltage-assisted magnetic switching.

This is a summary of:
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The mission

Moiré superlattices[1] are created by stacking one layer of a van der Waals (vdW) material onto another with either a lattice mismatch or a small twist angle. These large superperiodic patterns define a new crystal structure and can drastically alter the electronic properties of the material. Various correlated and topological phenomena have been observed in moiré materials such as twisted bilayer graphene and twisted transition metal dichalcogenides[1]. These varied phases (ranging from superconducting to insulating) can be controlled by the twist angle as well as by electric and magnetic fields. It is desirable to expand moiré physics to additional material types and degrees of freedom (that is, beyond charge).

The discovery of magnetism down to the two-dimensional (2D) limit in vdW magnets[2] has provided opportunities to explore exotic magnetic states and manipulate spin–charge coupling, giving rise to one of the most active fields in condensed-matter physics. However, the experimental engineering and control of the spin degree of freedom by moiré superlattices is still in its infancy[3] compared with the control of the charge degree of freedom. We aimed to realize and control magnetic states resulting from moiré superlattices in twisted vdW magnets, particularly for bilayer chromium triiodide (CrI_3).

The discovery

In bilayer CrI_3, the two layers are antiferromagnetically coupled with opposite magnetizations and thus have zero net magnetization near zero magnetic field[2]. However, the interlayer magnetic exchange between the two CrI_3 layers depends on the structural stacking order[4]. Twisting one CrI_3 layer relative to the other can create moiré superlattices with alternating structural domains. In twisted CrI_3, the stacking-dependent interlayer magnetic exchange can therefore give rise to non-collinear spin textures and non-trivial magnetic phases[4,5].

To demonstrate such moiré magnetism, we fabricated twisted double bilayers (tDBs) of CrI_3 – that is, one CrI_3 bilayer on top of another with a twist angle between them – using the tear-and-stack technique (Fig. 1a), which is commonly used to build twisted bilayers of graphene or transition metal dichalcogenides[1]. We then employed magneto-optical Kerr effect (MOKE) microscopy to characterize the magnetic behaviour.

Although the constituent bilayers of CrI_3 are each a layered antiferromagnet, after stacking two bilayers into a tDB, we observed a ferromagnetic state with non-zero magnetization. This result suggests the coexistence of antiferromagnetic and ferromagnetic orders, which is the hallmark of moiré magnetism (Fig. 1b). Furthermore, the emergent ferromagnetic state extends over a wide range of twist angles and exhibits non-monotonic temperature dependence. We also demonstrated voltage-assisted magnetic switching, whereby states with different magnetizations can be accessed and controlled by varying the gate voltage.

Our experimental and theoretical investigations revealed that tDB CrI_3 has a rich phase diagram with non-collinear magnetic phases that exhibit coexisting ferromagnetic and antiferromagnetic domains. The observed non-trivial magnetic states and control of these states through the twist angle, temperature and electrical voltage are consistent with the phase diagram, in which the energy balance between magnetic domains and domain-wall formation can lead to different phases being favoured.

Future directions

Our findings extend moiré physics to the spin degree of freedom and deepen the understanding of the non-trivial magnetic orders in twisted magnets, including layered antiferromagnets. The electrical tunability of the non-trivial magnetism could be of practical interest for the development of memory and spin-logic devices. Moreover, moiré magnets might provide a platform for the exploration of spintronics and magnetoelectronics.

Nevertheless, our polar MOKE measurements were only sensitive to out-of-plane magnetization. To demonstrate the in-plane component of the magnetization of the non-collinear phases, other probes sensitive to in-plane magnetization are needed, such as longitudinal MOKE measurements or spatially sensitive spin probes. Furthermore, spatially resolved measurements are required to study the topological skyrmion lattices and magnon networks potentially hosted in the non-trivial magnetic phases.

Twisting is becoming a powerful knob to 'create' materials that do not exist in nature. As this approach is applicable to a wide range of homo- and heterobilayers of vdW magnets, including ferromagnets, antiferromagnets, multiferroics and even spin liquids, it could enable the exploration of new types of magnetism and applications ranging from spintronics to quantum information science.

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EXPERT OPINION

“The authors study the magnetic properties of twisted double bilayer CrI_3 as a function of the external magnetic field and applied voltage using MOKE. This topic is of great interest to the community, and the conclusions are striking.” **Onur Erten, Arizona State University, Tempe, AZ, USA.**

FIGURE

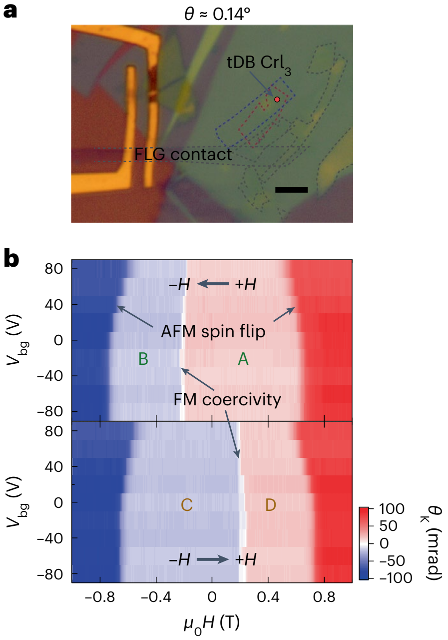

Fig. 1 | A tDB CrI_3 device and the electrical control of moiré magnetism. **a**, Optical micrograph of a tDB CrI_3 device used for demonstrating the electrical control of the magnetic behaviour (where θ is the twist angle). A few-layer graphene (FLG) flake is used as the contact to the stack. Scale bar, 5 µm. **b**, MOKE signal as a function of the magnetic field (μ_0H) and back-gate voltage (V_{bg}). Four regions labelled A–D represent magnetic states with different magnetizations that can be accessed and controlled by varying the gate voltage. The top and bottom panels correspond to opposite sweep directions of the magnetic field. Ferromagnetic (FM) coercivity and antiferromagnetic (AFM) spin-flip transitions across different moiré magnetic states are marked by arrows. θ_K, Kerr rotation angle. © 2023, Cheng, G. et al.

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BEHIND THE PAPER

We started exploring stacked 2D magnets in 2019. Back then, moiré physics was mainly studied in twisted graphene and semiconducting transition metal dichalcogenides. These studies revealed strongly correlated and topological phenomena that do not exist in the materials themselves, such as superconductivity, correlated insulating states and orbital magnetism. In principle, the twisting technique can be applied to any vdW material, enabling new physics to be explored within the diverse physical properties of vdW materials.

We first worked on heterostructures of layered antiferromagnets, CrI_3 and $CrCl_3$, in which we found emergent interfacial ferromagnetism, and gained experience in 2D magnetic heterostructures. Exploring twisted CrI_3 (with a focus on stacking bilayers, which are also layered antiferromagnets) was thus a natural progression and led us to reveal that tDB CrI_3 has a rich magnetic phase diagram (as calculated by our theory collaborators, Pramey Upadhyaya and colleagues). **Y.P.C.**

FROM THE EDITOR

“Stacking two-dimensional materials can be used to create designer structures with specific properties. The properties of these van der Waals materials can also be manipulated by adjusting the twist angle between layers. Here, the authors explore the effect of twist angle in double bilayers of chromium triiodide (CrI_3) and discover coexisting magnetic phases. Notably, electrical gating is shown to influence the magnetism of the CrI_3 devices.” **Katharina Zeissler, Associate Editor, *Nature Electronics*.**