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Discovery of Intrinsic Ferromagnetism in Two-Dimensional van der Waals Crystals

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Abstract: We report the experimental discovery of intrinsic ferromagnetism in $Cr_2Ge_2Te_6$ atomic layers by scanning magneto-optic Kerr microscopy. In this 2D van der Waals ferromagnet, unprecedented control of transition temperature is realized via small magnetic fields.
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Atomically thin, layered van der Waals (vdW) crystals provide ideal 2D material systems hosting exceptional physical properties. Emerging functional devices (e.g., ultrafast photodetector) have been derived primarily from the electron charge degree of freedom, whereas 2D spintronics based on vdW crystals is still in its infancy, severely hindered by the lack of long-range ferromagnetic order that is crucial for macroscopic magnetic effects. The emergence of ferromagnetism in 2D vdW crystals, if possible, combined with their rich electronics and optics, could open up numerous opportunities for 2D magnetic, magnetoelectric, and magneto-optic applications. Tremendous efforts have been attempted to extrinsically induce magnetic responses by defect engineering, by adding magnetic species via intercalation or substitution, or by magnetic proximity effect. In contrast, if realizable, intrinsic ferromagnetism originating from the parent 2D lattice itself is fundamental for both understanding the underlying physics of electronic and spin processes, and device applications.

Whether or not long-range ferromagnetic order that exists in bulk can persist in 2D regime is a fundamental question, because the strong thermal fluctuations may easily destroy the 2D ferromagnetism, according to the Mermin-Wagner theorem [1]. However, with the presence of a spin wave excitation gap arising from magnetic anisotropy, long-range ferromagnetic order can exist at finite temperature. Most of the reported ferromagnetic vdW bulk crystals so far are magnetically soft with small easy-axis anisotropy [2]. The challenge of harnessing the long-range ferromagnetic order in 2D vdW crystals hinges on the strength of magnetic anisotropy retained in the 2D regime. In this work, we report the first experimental discovery of long-range ferromagnetic order in pristine $Cr_2Ge_2Te_6$ atomic layers by temperature- and magnetic field-dependent Kerr effect via scanning magneto-optic Kerr microscopy. **Fig. 1** shows a magnetic hysteresis loop of a six-layer $Cr_2Ge_2Te_6$ sample, demonstrating an intrinsic soft ferromagnetism. The small single-domain remanence is strong evidence that thermal fluctuations rather than the formation of multi-domains (usually in the bulk) is the reason for the reduced magnetization in 2D $Cr_2Ge_2Te_6$.

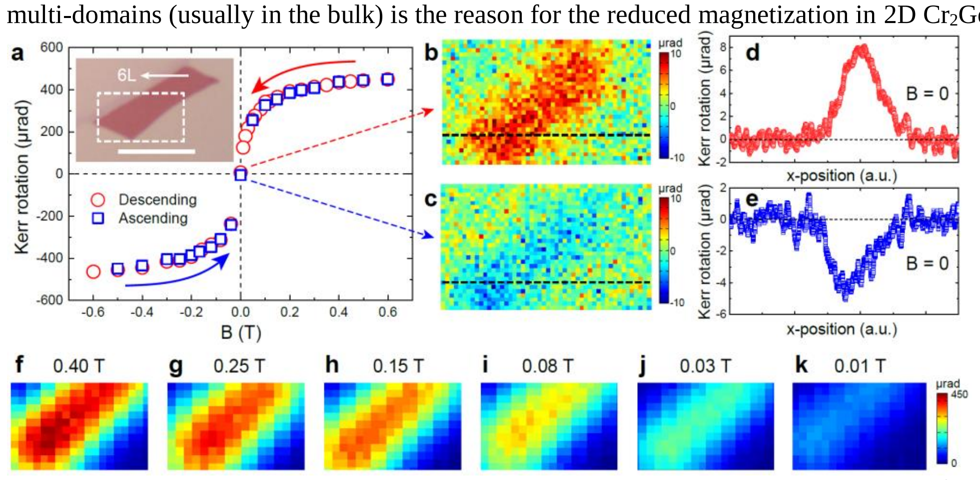

Fig. 1. Ferromagnetic hysteresis loop with single domain remanence in a six-layer (6L) $Cr_2Ge_2Te_6$. a, Hysteresis loop of a 6L flake at 4.7 K showing saturating trend at 0.6 T and small non-vanishing remanence. The solid red (blue) arrow represents the descending (ascending) field. Loop starts from 0.6 T. Inset is an optical image of the flake and the bar is 10 μm. **b, c,** Scanned Kerr rotation images of the flake at zero field after withdrawing the field from 0.6 T and -0.6 T, respectively. **d, e,** Line scanning crossing the flake. The approximate line positions are

indicated by black dashed lines in **b** and **c**, but extending further out of the window. **b-e** show a clear signature of small but definitive remanent Kerr rotation angles (about 2% of the "saturated" Kerr rotation at 0.6 T) with opposite signs on ascending and descending branches. **f-k,** Kerr images of the highlighted area in **a** (within dashed rectangle), at different descending fields, showing the persistence of a magnetic single domain through out.

Due to the very-small intrinsic magnetic anisotropy, 2D $Cr_2Ge_2Te_6$ provides an intriguing experimental platform in which even a small magnetic field can effectively engineer the anisotropy, open a large spin-wave excitation gap, and therefore control the transition temperatures. This scenario is not possible to occur in 3D regime, intrinsically because the transition temperature of 3D material is determined by the exchange interaction rather than magnetic anisotropy (note that the typical value of exchange interaction is orders of magnitudes larger than magnetic anisotropy). As shown in **Fig. 2**, we experimentally demonstrate, in our 2D soft ferromagnetic vdW crystal, the unprecedented magnetic field control of ferromagnetic transition temperature by surprisingly small fields ($\leq$ 0.3 T).

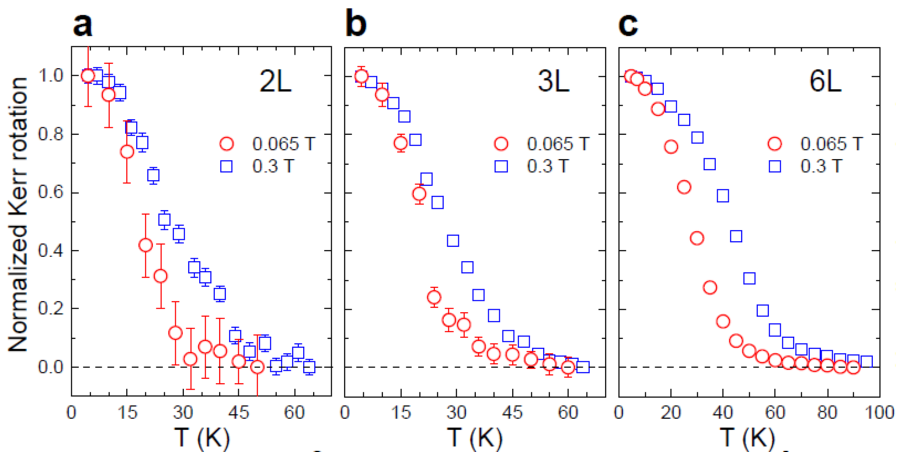

Fig. 2. Magnetic field control of transition temperature of few-layer $Cr_2Ge_2Te_6$. a-c, Normalized Kerr rotation angle as a function of temperature, under two different magnetic fields: 0.065 T and 0.3 T, for bilayer (2L), three-layer (3L) and six-layer (6L), respectively. The 0.3 T field shifts the curve drastically, with respect to 0.065 T, indicating a strong renormalization of the transition temperature in few-layer $Cr_2Ge_2Te_6$. Error bars (the standard deviation of sample signals) are smaller than the plotted dot if not shown. In 2D limit, when single ion anisotropy is negligibly small, the transition temperature will be very low, and can be easily tuned with a small magnetic field (e.g., B < 0.5 T). In bulk limit, due to the 3D nature, such tuning ability does not hold.

In summary, we report the first experimental observation of intrinsic ferromagnetism in 2D ferromagnetic vdW crystal $Cr_2Ge_2Te_6$ [3], where a strong dimensionality effect arises from the low-energy excitations of magnons. Through the effective engineering of the magnetic anisotropy by small magnetic fields, we discover the unprecedented magnetic field control of transition temperatures in 2D soft ferromagnetic vdW crystal. Our experimental observations are confirmed by the renormalized spin wave theory analysis and calculations, which can provide generic understanding of the ferromagnetic behaviors of many 2D vdW soft ferromagnets such as $Cr_2Si_2Te_6$ and CrI_3. Our discovery of 2D soft ferromagnetic vdW crystal $Cr_2Ge_2Te_6$ provides a nearly ideal 2D Heisenberg ferromagnet for exploring fundamental physics, and opens new possibilities for applications such as ultra-compact spintronics.

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