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Fully Electric Switching of Magnetic Domains in a van der Waals Multiferroic Heterostructure

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Abstract. Multiferroic materials offer a powerful and efficient way to control magnetism using electric fields, a goal that has been pursued for many years. To date, the fully electric switching of magnetism still remains challenging. Here, we report a van der Waals (vdW) heterostructure multiferroic comprising magnetic trilayer CrI_3 and ferroelectric (FE) α-In_2Se_3. Using a magneto-optoelectric coupled system, we observed fully electric switching of magnetic domains in the multiferroic heterostructure at zero magnetic field. Theoretical calculations reveal that the electric switching of magnetic states originates from the modulation of interlayer coupling in trilayer CrI_3 by ferroelectric polarization. Our work provides an ideal platform for creating strong magnetoelectric coupling multiferroics at the atomic layer limit, with potential applications in low-energy multistate magnetic memory and brain-inspired neuromorphic computing.

Keywords: Van der Waals multiferroic heterostructure; magnetoelectric coupling; spintronics device.

1. Introduction

The use of pure electric fields, rather than magnetic fields, to toggle magnetism in storage media offers a promising avenue for next-generation low-power spintronics and memory technologies [1-3]. Magnetoelectric coupling, particularly in single-phase multiferroic systems, facilitates digital information control by leveraging the interplay between magnetic and ferroelectric orders [4-6]. However, single-phase multiferroic materials are scarce due to the inherent conflict between magnetism and electric polarization, leading to typically weak magnetoelectric coupling [7]. Some studies have demonstrated the control of magnetization in multiferroic heterostructure films via strain-mediated indirect coupling [8, 9]. Yet, achieving magnetization switching at zero magnetic field through direct magnetoelectric coupling remains a significant challenge. In van der Waals heterostructures, the interlayer coupling between adjacent atomically thin layers allows electronic wave functions to span multiple layers, transforming the layer degree of freedom from a spatial variable to a quantum mechanical index. The recent discovery of 2D van der Waals magnets and ferroelectrics has sparked widespread attention [10-12], as these atomically thin crystals offer new possibilities for studying the interface coupling between magnetic and ferroelectric orders through van der Waals engineering.

Here, we report a multiferroic heterostructure composed of trilayer (3L) CrI3 and ferroelectric α-In2Se3. Using low-temperature RMCD imaging, we observed that reversing the gate voltage without an external magnetic field can switch the magnetic domains. Spin density calculations suggest that opposing ferroelectric polarization configurations produce distinct interlayer coupling arrangements, leading to magnetic state transitions and a nonreciprocal magnetoelectric coupling effect. This study reveals unique and complex physical phenomena emerging from the 2D magnetic-ferroelectric interface and presents a new pathway for continuous electric control of magnetism in ultralow-power spintronics.

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2. Experimental and Computational Section

2.1 Device Fabrication

3L CrI3, hBN, α-In2Se3, and graphene nanoflakes were mechanically exfoliated from bulk crystals using PDMS films inside a glovebox. These materials were sourced from Chengdu Mouton Tech companies. The exfoliated CrI3, α-In2Se3, hBN, and graphene flakes were sequentially transferred onto pre-patterned Au electrodes (50 nm thick) on SiO2/Si substrates within the glovebox, forming the multiferroic heterostructure. The completed structure was then in-situ loaded into the cold head for subsequent optical analysis.

2.2 Optical Measurements

Raman spectroscopy was conducted using a Witec Alpha 300R Plus confocal Raman microscope designed for low-wavenumber measurements. A 633 nm laser (~1 mW), aligned parallel to the X-axis (0°), was focused onto the samples via a 50× long working distance objective (Zeiss, NA = 0.55). RMCD (reflective magnetic circular dichroism) measurements were performed using the same Witec Alpha 300R Plus microscope, combined with a magneto-optical system (Mottec MO-100, Chengdu Mouton Tech), a closed-cycle superconducting magnet, and a closed-cycle helium optical cryostat, integrated with an electronic transport measurement system. A free-space 633 nm laser (~3 μW), modulated by a photoelastic modulator (50 kHz), was reflected by a non-polarizing beamsplitter cube (R/T = 30/70) and focused on the sample using the same objective. The RMCD signal, reflecting the sample's out-of-plane magnetization, was detected by a photomultiplier tube following its passage through the same beamsplitter. The signal was processed using a lock-in amplifier and Witec photocurrent scanning system. The 155 voltage sources were used to apply the gate voltage.

2.3 Computational Details

All first-principles calculations were carried out utilizing the Vienna ab initio simulation package (VASP). Further information regarding the computational methods is available in earlier publications [13].

3. Results and Discussion

3.1 Construction of 3L CrI3/In2Se3 Multiferroic Heterojunction Devices

In the two-dimensional plane, Cr3+ ions are arranged in a honeycomb lattice, where each Cr atom is coordinated with six iodine atoms, forming a distorted octahedral structure (left panel of Fig. 1a). Within each layer, the magnetic moments of the Cr3+ cations align, while interlayer interactions are antiferromagnetic (AFM; right panel) [14]. This results in a net ferromagnetic (FM) state at zero magnetic field in the 3L CrI3. The top and side views illustrating the atomic arrangement and crystal structure of α-In2Se3 are depicted in Fig. 1b. Ferroelectric α-In2Se3 consists of dislocated quintuple layers, inducing a break in spatial inversion symmetry. It is confirmed that out-of-plane ferroelectric polarization is consistently present in hexagonal α-In2Se3, independent of the flake thickness [15]. The vertically stacked multiferroic heterostructure consists of a 3L CrI3 and a ferroelectric α-In2Se3 nanoflake, positioned between two graphene electrodes (Fig. 1c, d). Fig. 1e presents a typical RMCD signal for 3L CrI3 under an applied perpendicular magnetic field. When the absolute value of the magnetic field |μ0H| exceeds 1.2 T, the spins in all three CrI3 layers align in the same direction, as illustrated in the spin configuration diagram. At zero magnetic field, the 3L CrI3 retains a magnetic memory, with the magnetization pointing upwards in state FM1+ and downwards in state FM1−, depending on whether the magnetic field was ramped down from a positive or negative direction [14]. In Fig. 1f, Raman modes are observed around 94, 159, 179, and 256 cm^{-1}, corresponding to the E2, E3, E4, and A14 modes of α-In2Se3

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[16]. Additionally, four Raman peaks marked with a five-pointed star are assigned to the Ag and Eg modes of magnetic CrI3 [17].

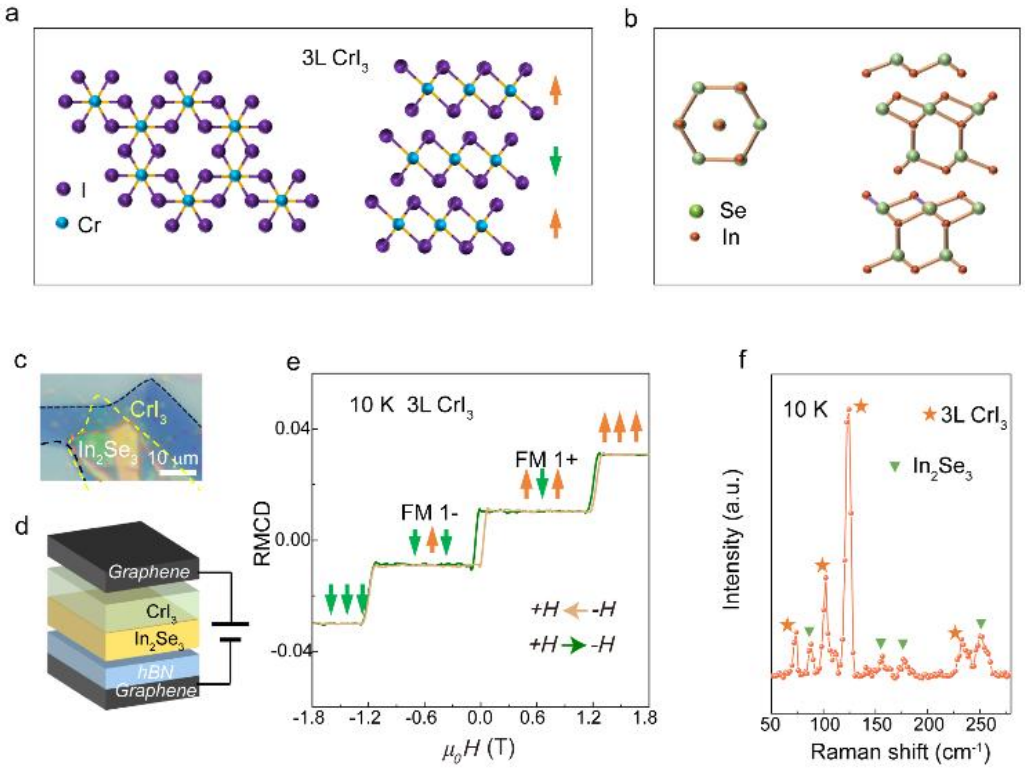

Fig. 1 3L CrI3/In2Se3 heterostructure multiferroic device. Atom structure models for the top and side views of α-In2Se3 (a) and 3L CrI3 (b). (c) Optical micrograph of multiferroic device. (d) A schematic side view of multiferroic device sandwiched between graphene contacts and hBN. (e) RMCD signal of a 3L CrI3 as a function of out-of-plane magnetic field. (e) Raman spectra of multiferroic heterojunction at 10 K.

3.2 Voltage-Controlled Magnetic Domain Switching

To observe voltage-controlled magnetic domain switching, we employed scanning RMCD microscopy to image and measure the magnetic domains of freshly exfoliated trilayer CrI3. Polar RMCD imaging is a reliable and powerful tool that reveals two-dimensional magnetism on a microscopic scale, with RMCD intensity proportional to the out-of-plane magnetization. All magneto-optical measurements were performed using a 633 nm laser to achieve optimal detection sensitivity [11]. Fig. 2a-i shows RMCD images of trilayer CrI3 on α-In2Se3 as the voltage is swept from +8 V to -8 V without any external magnetic field. At +8 V, the RMCD image shows upward magnetization (red) across the entire area. As the voltage decreases to 0 V, although part of the region switches to downward magnetization (blue), the majority of the area retains upward magnetization. At -2 V, the area of downward magnetization increases further, with a corresponding decrease in upward magnetization. As the negative voltage increases to -8 V, nearly the entire region switches to downward magnetization. This voltage-driven magnetic domain switching, in the absence of an external magnetic field, demonstrates strong interfacial magnetoelectric coupling, which is critical for the development of low-power spintronic devices.

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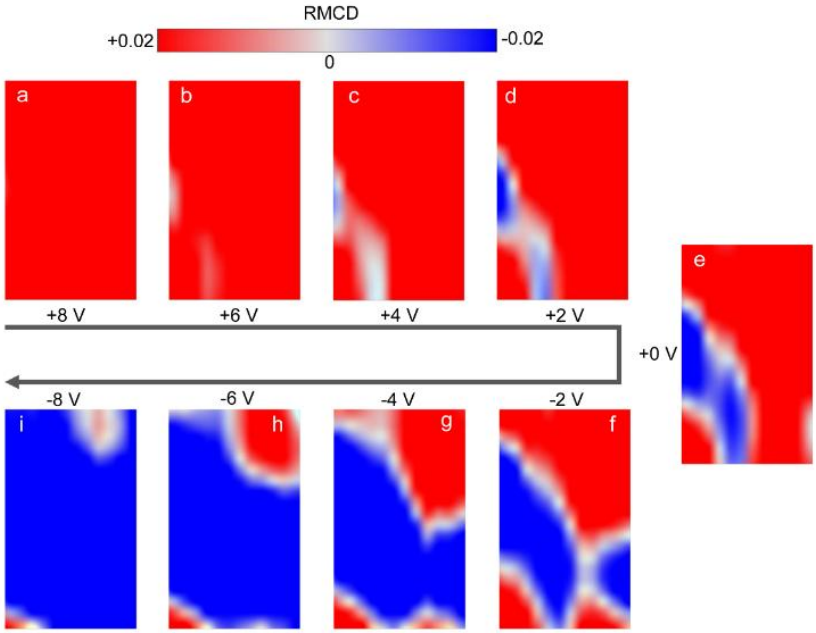

Fig. 2 Fully electric switching of magnetic domains in 3L CrI3/In2Se3 heterostructure multiferroic. (a)-(i) Polar RMCD maps upon a 633 nm laser with diffraction-limited spatial resolution (see Methods), collected at 10 K and selected voltage. The scanned area is 2×4 μm.

3.3 Mechanism of Voltage-Controlled Magnetic Domain Switching

Raman spectroscopy, a widely used technique for stress characterization in two-dimensional materials, reveals in Fig. 3a that the Raman features of CrI3 within multiferroic heterostructure devices remain unchanged across different applied voltages. This invariance indicates the absence of strain-mediated magnetoelectric coupling, a phenomenon commonly observed in conventional multiferroic heterostructures [8, 9]. To elucidate the underlying mechanism driving the interlayer magnetic phase transition in trilayer CrI3 through the switching of ferroelectric polarization in In2Se3, we examine the spin density (SD) for the no-P state (Fig. 3b) and the differential spin density (DSD) of interlayer I atoms in the 3L CrI3/In2Se3 heterostructure under FM (left panel in Figs. 3c, d) and AFM (right panel in Figs. 3c, d) configurations for the Pdown and Pup states. Actually, the interlayer magnetic coupling in CrI3 has been demonstrated to arise from the direct interaction between adjacent I atoms in each CrI3 monolayer [18]. The DSD in the FM configuration, as shown in Fig. 3c (left panel), reveals that the downward ferroelectric polarization primarily increases the charge density in the spin-down component of the I orbitals at interface 1. This enhanced spin density significantly strengthens the FM hopping between the two I atomic layers at interface 1. In the AFM configuration (right panel in Fig. 3c), the upward ferroelectric polarization mainly increases the charge density in either the spin-down or spin-up component of the I orbitals at interface 2. Consequently, the enhanced spin density markedly reinforces the AFM hopping between the adjacent I atomic layers at interface 2. The upward ferroelectric polarization, corresponding to the -8V applied in experiments, triggers an interlayer electronic density reconstruction in the 3L CrI3, consistent with the FM 2- magnetic state depicted in the Fig. 3c. When the polarization is switched downward, corresponding to the +8V applied in the experiments, the spin-down component of the I orbitals at interface 2 shows a notable increase in charge density, thereby strengthening the FM hopping between the I atomic layers (left panel in Fig. 3d). In contrast, in the AFM configuration, the downward polarization reduces the charge density in either the spin-down or spin-up component of the I orbitals at interface 2, significantly weakening the AFM hopping between the adjacent I atomic layers (right panel in Fig. 3d). This downward polarization leads to an interlayer electronic density reconstruction in 3L CrI3, consistent with the FM 2+ phase.

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These mechanisms align with the experimentally observed voltage-induced net magnetization reversal (Fig. 2a, i).

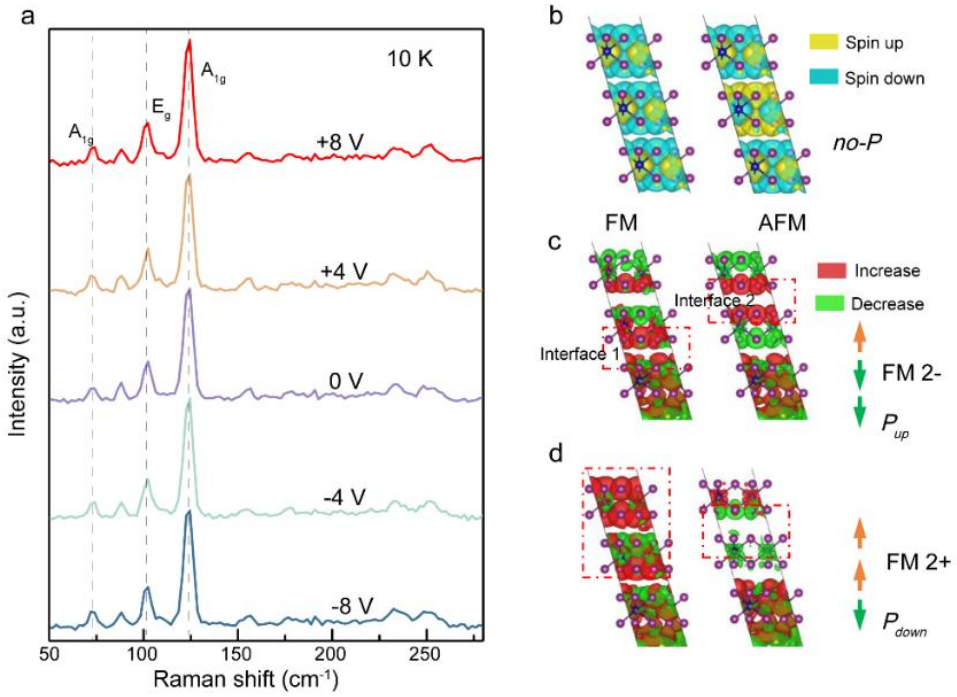

Fig. 3 Mechanism of voltage-controlled magnetic state switching. (a) Raman spectra of the multiferroic device under varying positive and negative voltages at 10 K. (b) SD of 3L CrI3 for no-P. The yellow and cyan isosurface in refer to spin-up and -down density, respectively. (c), (d) DSD of 3L CrI3 for Pup and Pdown in FM configuration (left side) and AFM configuration (right side). The increase and decrease of spin density (absolute value) are shown with red and green isosurface, respectively.

4. Summary

We observed a gradual reversal of magnetic domains driven by applied voltage, without the need for an external magnetic field, in the van der Waals 3L CrI3/In2Se3 heterostructure multiferroic. Unlike the conventional strain-mediated mechanisms in thin-film multiferroic heterojunctions, theoretical calculations suggest that ferroelectric polarization induces a reconstruction of interlayer spin density, acting as a switch between the net upward and downward magnetization states in 3L CrI3. This magnetic-field-free ferroelectric polarization switching of magnetic states paves the way for the development of a new class of ultralow-power, ultrafast, and multistate spintronic devices.

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