

# Supporting Information

## Observation of Room-Temperature Magnetoresistance in Monolayer MoS<sub>2</sub> by Ferromagnetic Gating

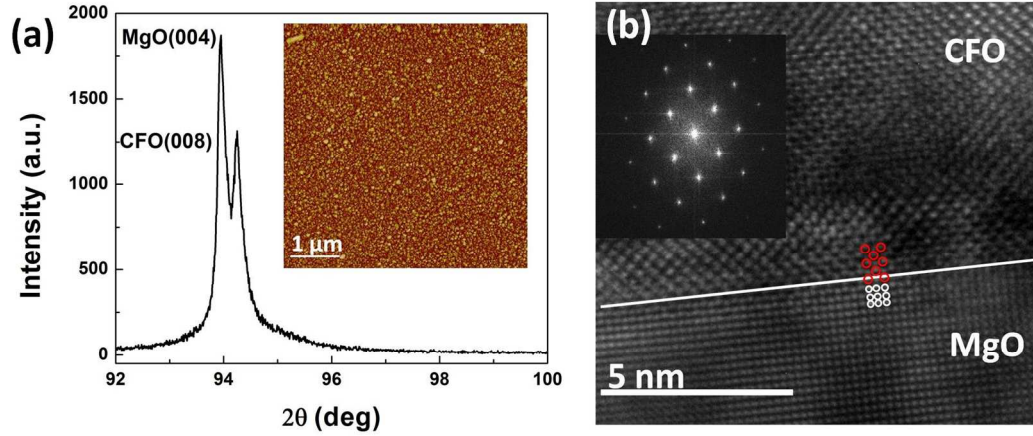
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## Supporting Figures



**Figure S1.** Characterization of the pulse-laser-deposited (PLD)  $\text{CoFe}_2\text{O}_4$  (CFO) thin films on [001]-oriented MgO substrates. (a) X-ray diffraction (XRD)  $2\theta$  scanning pattern of CFO/MgO. Inset is the atomic force microscopy (AFM) image of the surface morphology of CFO thin film over a  $5\ \mu\text{m} \times 5\ \mu\text{m}$  area. (b) Cross-sectional high-resolution transmission electron microscope (HR-TEM) image of CFO/MgO interface. The fast Fourier transform (FFT) pattern is shown in the inset.

CFO possesses cubic spinel structure with  $a = b = c = 0.8391\ \text{nm}$ . Thus, [001]-oriented MgO crystals ( $a = 0.42\ \text{nm}$ ) are selected as the substrates to grow CFO thin films by considering the small lattice mismatch (-0.12%) between them. Thus CFO thin film can be well deposited on the MgO substrate by PLD method. A sharp (008) diffraction peak of the CFO and (004) of MgO are clearly shown in the diffraction pattern, while the (004) peak of the CFO is failed to detect since it is adjacent to the strong (002) peak of the MgO substrate. Only the (00 $l$ ) peaks of the CFO

films are shown in the XRD diffraction patterns, suggesting the CFO films are *c*-axis oriented. Furthermore, the cross section of the as-prepared CFO/MgO sample is studied by HR-TEM. The thickness of the fabricated CFO thin film can be determined to be about 80 nm by TEM. The CFO thin films are cubic-on-cubic grown on the MgO substrates (one CFO cubic on 4 MgO cubic lattices). The epitaxial growth of CFO is demonstrated in the FFT pattern. Based on the XRD, AFM and TEM characterization, high-quality CFO epitaxial thin films are well prepared for the subsequent MoS<sub>2</sub> deposition.

In this work, there are mainly three reasons for us to choose CFO as the ferromagnetic gating material. Firstly, CFO is a room-temperature ferromagnetic material. This is important for the bilayer heterostructure to achieve a room temperature RM. Secondly, CFO possesses high permittivity. Thus, the CFO thin films should not divert electric current away from MoS<sub>2</sub> layer. Thirdly, epitaxial CFO thin films can be well deposited by PLD method on MgO substrate in our research group, which can help us fabricate the bilayer heterostructure.

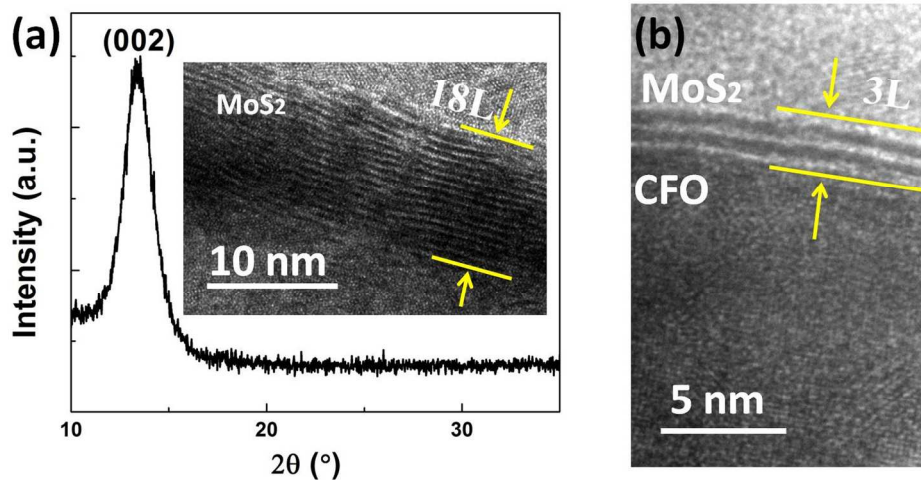
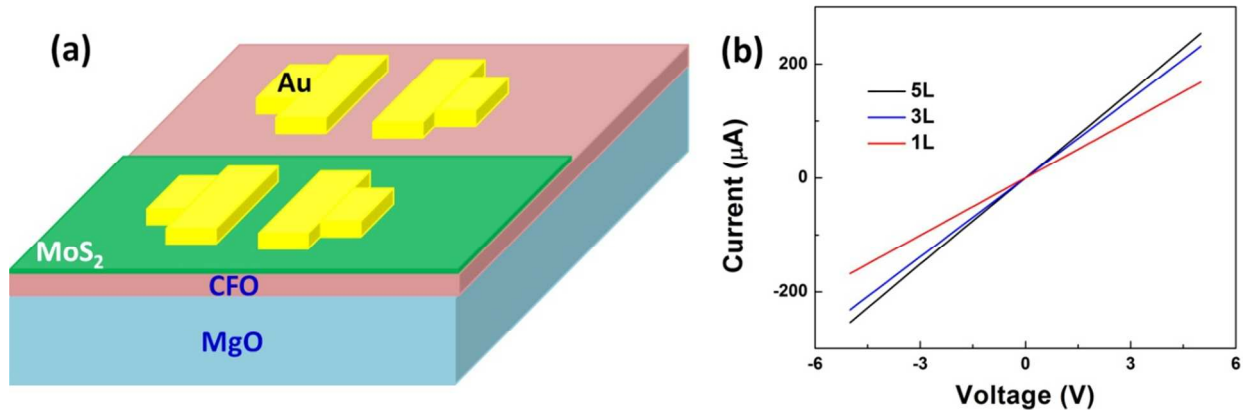


Figure S2. (a) X-ray diffraction (XRD)  $2\theta$  scanning pattern of  $\text{MoS}_2$  sample. Inset shows the cross-sectional TEM image of the  $\text{MoS}_2$  sample with thickness about 18-layer (18L). (b) Cross-sectional TEM image of the 3-layer (3L)  $\text{MoS}_2$  /  $\text{CFO}$  heterostructure.

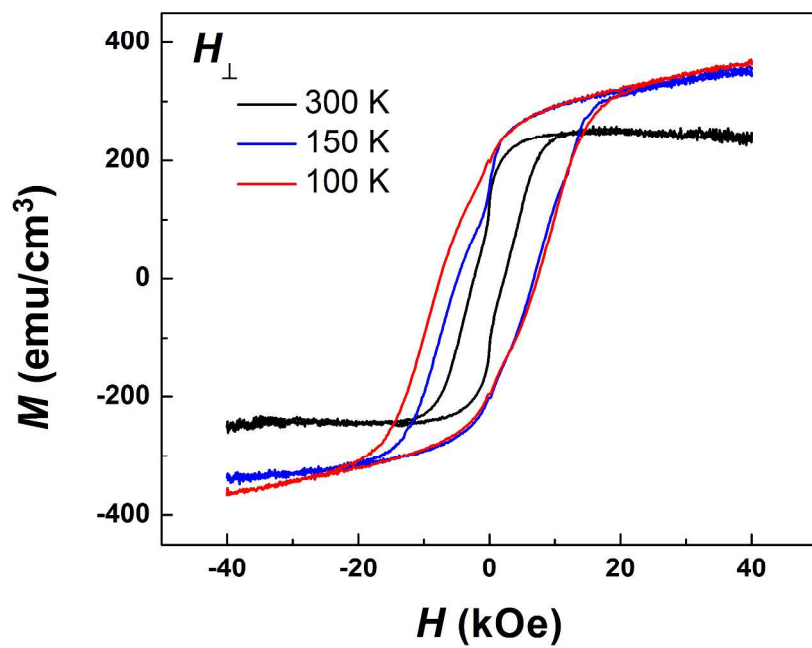
The (002) diffraction peak indicates the formation of  $c$ -axis oriented  $\text{MoS}_2$  layers. Cross-sectional TEM images of multilayer (ML)  $\text{MoS}_2$  samples show the layered structure of the PLD-grown samples. According to the deposition time and the deposited samples, the deposition rate of PLD-grown  $\text{MoS}_2$  is calculated to be about 55 laser pulses per layer.



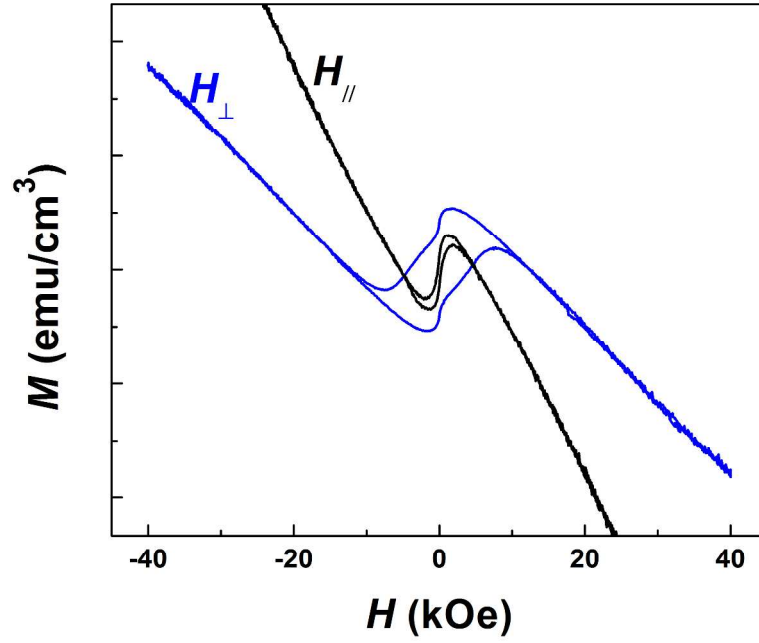
**Figure S3.** (a) Schematics of the structures for the transport properties measurements of the PLD-grown MoS<sub>2</sub> layers and the CFO thin film. (b) The current-voltage ( $I$ - $V$ ) curves of monolayer (1L), 3-Layer (3L) and 5-layer (5L) MoS<sub>2</sub> on CFO/MgO substrates.

The Au electrodes with width of 1 mm and channel length of 30  $\mu\text{m}$  were prepared on the MoS<sub>2</sub> layers and CFO thin film, respectively, by thermal evaporation method for transport measurements. The bandgap of monolayer MoS<sub>2</sub> is about 1.8 eV according to the photoluminescence measurements. The electrical properties of MoS<sub>2</sub> were characterized by the standard Hall measurements. The conductivity of monolayer MoS<sub>2</sub> is about  $150 \Omega^{-1}\text{cm}^{-1}$  and the carrier density is about  $1 \times 10^{14} \text{ cm}^{-2}$ . The  $I$ - $V$  curves show a completely linear behavior, suggesting the good ohmic contact properties between the MoS<sub>2</sub> layers and Au electrodes. The magnitude of electrical current is about 200  $\mu\text{A}$  when the applied bias is 5 V, while, no electrical current signals are achieved within an accuracy of 1 pA for CFO thin film even when the bias voltage is increased to 20 V. Thus, the CFO thin film possesses high permittivity as expected and should not divert electric current away from MoS<sub>2</sub> layers. Besides, the electrical resistance of CFO was also been measured when the external magnetic field was applied with amplitude of 9 k Oe in the PPMS system. However, during the entire process for the magnetic field changing

from -9 kOe to 9 kOe, the electrical resistance of CFO was too large to be measurable and showed no MR contribution.



**Figure S4.** Magnetic hysteresis loops of a CFO thin film when the magnetic field is applied perpendicular to the CFO plane at 300, 150, and 100 K.



**Figure S5.** Magnetic hysteresis loops of the CFO sample grown on single-crystal MgO substrate when the magnetic field is applied perpendicular or parallel to the CFO plane.

The two loops are the original data we obtained from the vibrating sample magnetometer (VSM) measurement system. Generally, to obtain the magnetization data of CFO thin film, the diamagnetic contribution from substrate is typically removed by subtracting the linear part at high magnetic field, which is a commonly used procedure to access the magnetization of ferromagnetic thin film. Through this process, the typical ferromagnetic loop of CFO can be achieved as shown in Figure 3a.