Supplementary Information: Two-dimensional ferromagnetism in CrTe flakes down to atomically thin layers

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**Experimental Section**

*CVD of the CrTe flake.* CrTe flakes were synthesized inside a tube furnace (Lindberg/Blue M) equipped with a 1.2-metre length and 1 inch. diameter quartz tube in an ambient pressure CVD system. Te powder (99.997%, Sigma Aldrich) and chromium chloride (CrCl3, 99.99%, Sigma Aldrich) were used as solid precursors and reactants. The Te powder was placed in a quartz boat and located upstream of the hot zone, where the temperature was approximately 500 °C. A mixture of CrCl3 powder and sodium chloride (NaCl, 99.95%, Sigma Aldrich) with a weight ratio of 10:1 was placed in the ceramic boat and placed in the center of the tube. The SiO2/Si substrate was placed on the boat with the surface downside. Before the heating process, the furnace was purged by flowing 300 sccm high-purity Ar gas for 5 min. The growth was conducted from 675–750 °C with 100 sccm Ar and 10 sccm H2 mixtures for 3 min. After synthesis, the furnace was rapidly cooled down to room temperature with the assistance of electric fans.

*Characterization of CrTe flakes.* The optical images for the CrTe flake were obtained by optical microscopy (BX51, Olympus). The thickness was measured by AFM (Dimension Icon, Bruker). The STEM and EDS images were obtained by an aberration-corrected JEOL ARM-200F. The Raman spectra were obtained by a Raman spectrometer (LabRAM HR800) with a 532 nm laser. The MOKE and RMCD measurements were obtained by a homemade polar MOKE microscope system. The laser wavelength ranged from 500 to 900 nm, and the intensity was 5 μW. The samples were placed in a 7 T superconducting magnet with the temperature range from 4.2 K to 300 K.
Supplemental Data

Fig. S1. Scanning electron microscope (SEM) results for CrTe flake. (a) SEM image of CrTe flake. (b) EDS elemental analysis of the CrTe flake. (c) and (d) Cr and Te elemental mappings of the sample shown in (a).

Tab. S1. Weight percentage and atomic percentage of Cr, Te in CrTe flake.

<table>
<thead>
<tr>
<th>Element</th>
<th>Wt%</th>
<th>At%</th>
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<tbody>
<tr>
<td>Cr</td>
<td>27.84</td>
<td>48.64</td>
</tr>
<tr>
<td>Te</td>
<td>72.15</td>
<td>51.36</td>
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Fig. S2. Raman spectra of CrTe flakes. (a) Raman spectra for 11nm CrTe flake, with Raman shift from 50 to 700 cm$^{-1}$. (b) Raman peak shift of P1 and P2 for 11 nm, 15 nm, 19 nm CrTe flakes, respectively. (c) Temperature-dependence Raman spectra for 11nm CrTe flake at 85 K, 150 K, 230 K, 300 K, respectively. (d) Raman peak shift of P1 and P2 as a function of temperature, respectively.
Fig. S3. Optical and AFM images of 11 nm CrTe flake. (a) Optical image of 11 nm CrTe flake for MOKE measurement. (b) The corresponding AFM image.
Fig. S4. MOKE signal for 11 nm CrTe flake measured before and after 2 weeks under PMMA film protected.
Fig. S5. Wavelength dependence of $\theta_K$ for 11 nm CrTe flake.
Fig. S6. Wavelength dependence of RMCD signal for 11 nm CrTe flake.
Fig. S7. Optical and AFM images of 39 nm CrTe flake. (a) Optical image of 39 nm CrTe flake for MOKE measurement. (b) The corresponding AFM image.
Fig. S8. Temperature dependence of $\theta_K$ for 39 nm CrTe flake.
Fig. S9. (a) and (b) Hysteresis loops of the 39 nm CrTe flake measured at 120 and 150 K, respectively. The second order differential curves of the Kerr rotation $\theta_K$ over the magnetic field are shown at the top.

There is a common and effective way to determine the phase transitions by the differential of the magnetization over the magnetic field. The second order differential of the Kerr rotation $\theta_K$ over the magnetic field curves for 120 K and 150 K are shown at the top of Fig. S9. There are two peaks for both two temperatures. The first peak (peak-type) shows the same property, which corresponds to the magnetic field when the spin direction changes. However, the second peak (valley-type) shows obviously difference. For MOKE signal at 120 K, which shows no intermediate state, the second peak corresponds to the saturated magnetic field. For 150 K, the second peak corresponds to the intermediate magnetic field. Therefore, the intermediate states could be determined quantitatively.