Supporting information:

Ultrathin Nanosheets of $CrSiTe_3$: A Semiconducting Two-Dimensional Ferromagnetic Material

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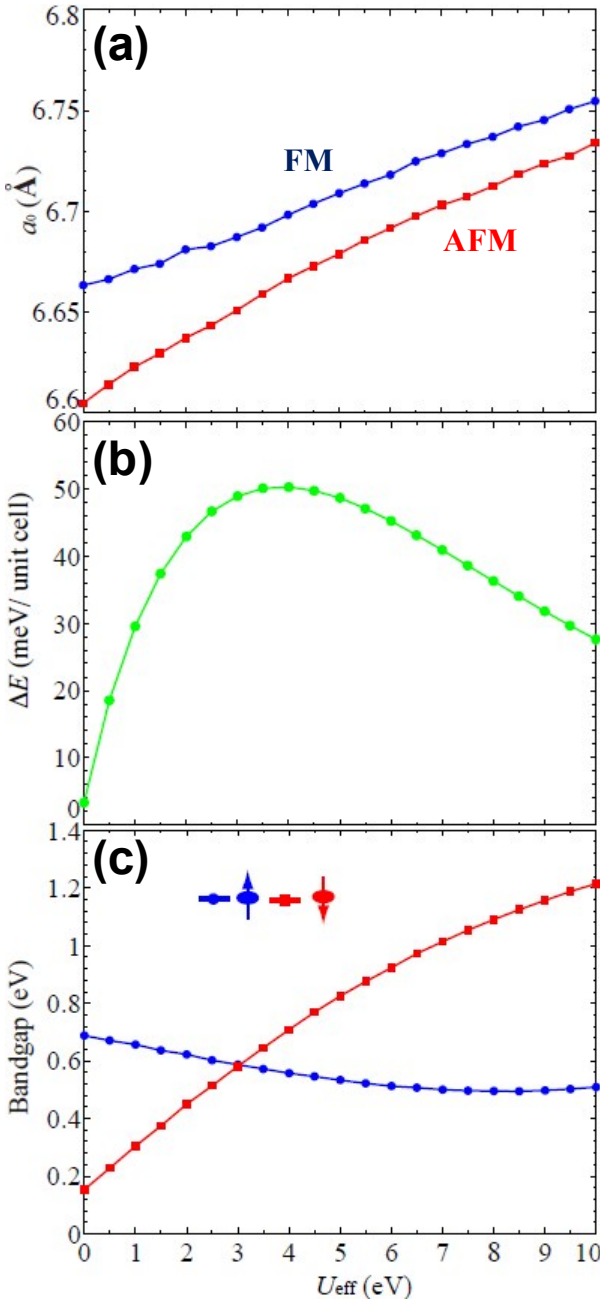

Figure S1. The U-dependent of (a) in-plane lattice constants of FM and Neel-AFM monolayer $CrSiTe_3$; (b) energy difference between FM and Neel-AFM monolayer $CrSiTe_3$; (c) spin-up (blue) and spin-down (red) bandgaps of FM monolayer.

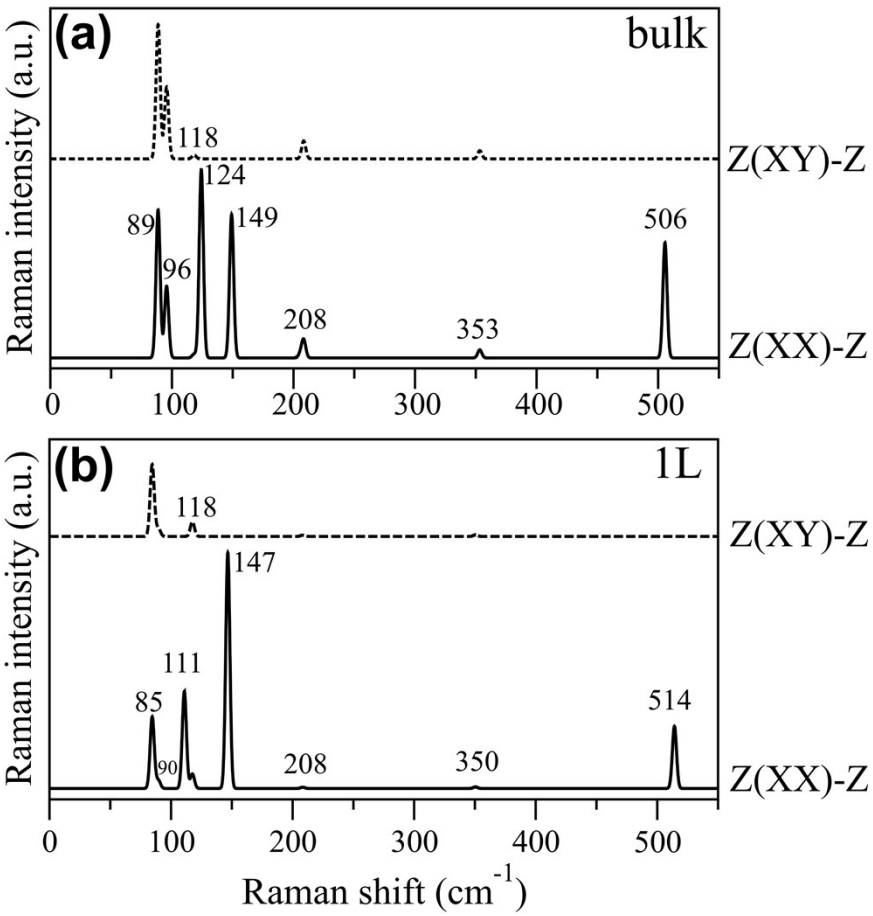

Figure S2. The Raman spectra determined from first principles simulation both in bulk (a) and in monolayer (b) of $CrSiTe_3$ are shown. Z(XX)-Z and Z(XY)-Z denote that the incident and scattering lights are in parallel and in perpendicular polarization configurations, respectively.

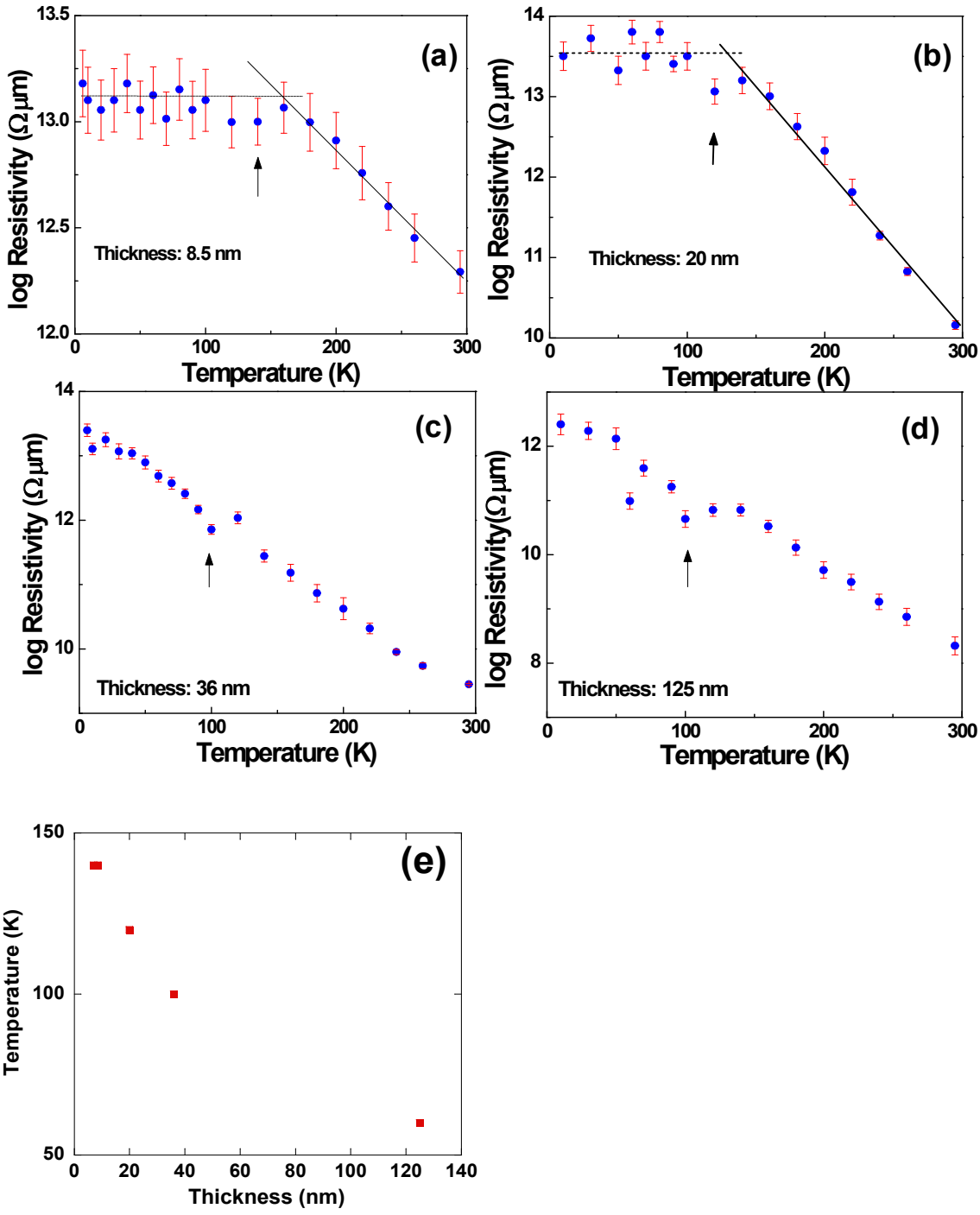
(a)
Thickness: 8.5 nm
log Resistivity (Ωμm)
Temperature (K)
(b)
Thickness: 20 nm
log Resistivity (Ωμm)
Temperature (K)
(c)
Thickness: 36 nm
log Resistivity (Ωμm)
Temperature (K)
(d)
Thickness: 125 nm
log Resistivity(Ωμm)
Temperature (K)
(e)
Temperature (K)
Thickness (nm)

Figure S3. The resistivity as a function of temperature for $CrSiTe_3$ FET devices at $V_{bg} = 0$ V with various thicknesses denoted as (a) 8.5 nm, (b) 20.0 nm, (c) 36 nm, and (d) 125 nm shows the clear change of resistivity which indicate the possible FM-AFM phase change at those temperatures. (e) The transition temperatures for different thickness of flakes show that transition temperature T_c generally increased as decreasing thickness.

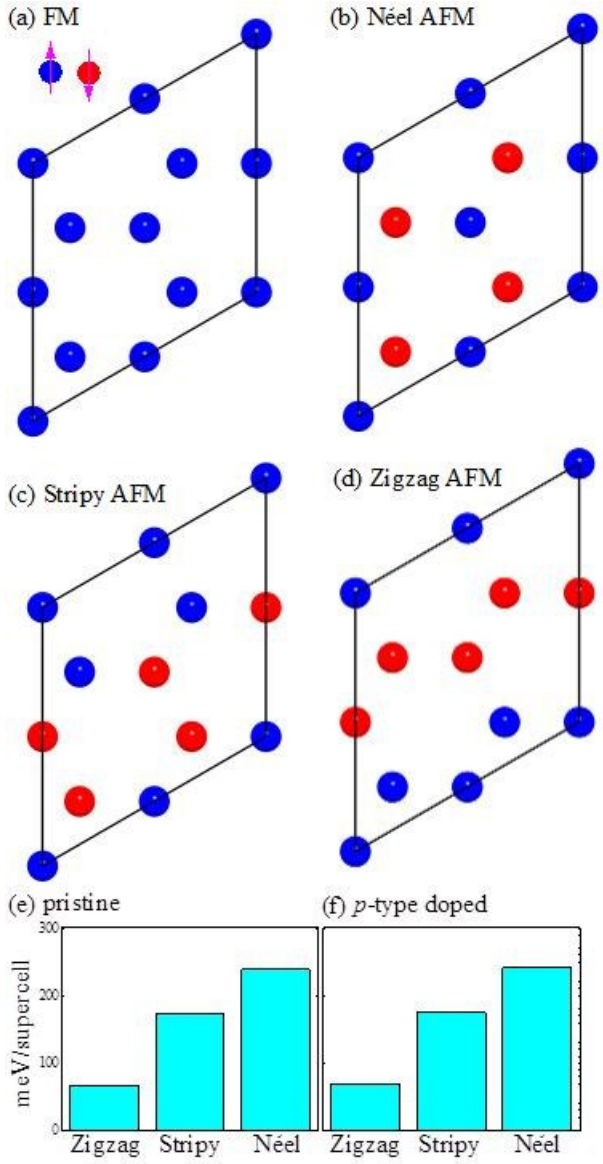

Figure S4. 2×2 supercell of single-layer $CrSiTe_3$ with (a) FM, (b) Neel, (c) strip, and (d) zigzag AFM spin textures. Only Cr atoms are shown. (e) and (f) show the energy differences of the AFM configurations with reference to the ground state FM one, which confirm the FM structure is the ground state.

The following is the detail of computing J_1, J_2, and J_3.

Followed by the notation in Ref [6], we have the following energies for single-layer $CrSiTe_3$ with the FM order and the so-called Néel, zigzag, and stripy AFM order.

$$E_{FM} = E_0 + (3J_1 + 6J_2 + 3J_3)S^2$$

$$E_{NEEL-AFM} = E_0 + (-3J_1 + 6J_2 - 3J_3)S^2$$

$$E_{Zigzag-AFM} = E_0 + (J_1 - 2J_2 - 3J_3)S^2$$

$$E_{Stripy-AFM} = E_0 + (-J_1 - 2J_2 + 3J_3)S^2$$

Here, E_0 is the energy of the spin-nonpolarzied configuration. Thus,

$$E_{NEEL-AFM} - E_{FM} = (-6J_1 - 6J_3)S^2 = 0.1193\,eV$$

$$E_{Zigzag-AFM} - E_{FM} = (-2J_1 - 8J_2 - 6J_3)S^2 = 0.0337\,eV$$

$$E_{Stripy-AFM} - E_{FM} = (-4J_1 - 8J_2)S^2 = 0.0867eV$$

Therefore, J_1, J_2, and J_3 as -2.39 meV, 0.00 meV, and +0.18 meV, respectively.