## **Supplementary Information**

## **All-Electrical Layer-Spintronics in Altermagnetic Bilayer**

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**Fig. S1** Spin-polarized band structures of CrS monolayer using different effective Hubbard U<sub>eff</sub> and HSE06. The Fermi level is set to the middle of the band gap.



**Fig. S2** (a) Phonon frequency spectrum of CrS monolayer. No imaginary modes appear in the phonon frequency spectrum. (b) Variations of the total energy of CrS monolayer at 300 K during AIMD simulations. Inset shows the snapshots of the equilibrium structure of CrS monolayer. Neither bond breakage nor structural reconstruction are observed in the snapshots after annealing for 5 ps.



**Fig. S3** Different magnetic configurations of CrS monolayer. The blue and red arrows indicate spin down and spin up, respectively.



**Fig. S4** Band structures of CrS monolayer with SOC. The Fermi level is set to the middle of the band gap.



**Fig. S5** Spin-resolved conductivity and spin polarization of the transport current in CrS monolayer.



**Fig. S6** Phonon frequency spectrum of CrS bilayer. No imaginary modes appear in the phonon frequency spectrum.



**Fig. S7** Spin-resolved conductivity using the semi-analytical transport model based on the parabolic energy dispersion fitting of the CBMs of the layer-spin subbands at X and Y valleys under the out-of-plane electric field of  $\pm 0.05$ ,  $\pm 0.10$ , and  $\pm 0.15$  V/Å.



**Fig. S8** Spin polarization of the transport current using the semi-analytical transport model based on the parabolic energy dispersion fitting of the CBMs of the layer-spin subbands at X and Y valleys as a function of Fermi level and out-of-plane electric field.

Electric field		AFM-Néel	AFM-Stripy	FM
In-plane	-0.15	0	0.48	1.31
	-0.10	0	0.48	1.31
	-0.05	0	0.48	1.31
	0.05	0	0.48	1.31
	0.10	0	0.48	1.31
	0.15	0	0.48	1.31
Out-of-plane	-0.15	0	0.48	1.30
	-0.10	0	0.48	1.30
	-0.05	0	0.48	1.30
	0.05	0	0.48	1.30
	0.10	0	0.48	1.30
	0.15	0	0.48	1.30

**Table S1.** Energy (eV) of AFM-Néel, AFM-Stripy and FM configurations of CrS monolayer under in-plane and out-of-plane electric field of  $\pm 0.05$ ,  $\pm 0.10$  and  $\pm 0.15$  V/Å. The energy of AFM-Néel is set to 0.

## **Supplementary Notes**

Based on Eq. 1, when T = 0 K, the conductivity tensor can be written as:

$$\sigma_{ij}^V = \frac{ne^2\tau}{m^*}$$

where n is the carrier density, and m\* is the effective mass. m\* can be presented as:

$$\begin{bmatrix} \frac{1}{m^*} \end{bmatrix} = \frac{1}{\hbar^2} \begin{bmatrix} \frac{\partial^2 \mathcal{E}}{\partial k_x^2} & \frac{\partial^2 \mathcal{E}}{\partial k_x \partial k_y} \\ \frac{\partial^2 \mathcal{E}}{\partial k_y \partial k_x} & \frac{\partial^2 \mathcal{E}}{\partial k_y^2} \end{bmatrix}$$

Therefore, based on Eq. 2-3, the conductivity tensors at X and Y valleys can be written as:

$$\sigma_{ij}^{X} = \begin{bmatrix} \frac{ne^{2}\tau}{m_{1}} & 0\\ 0 & \frac{ne^{2}\tau}{m_{2}} \end{bmatrix}$$
$$\sigma_{ij}^{Y} = \begin{bmatrix} \frac{ne^{2}\tau}{m_{2}} & 0\\ 0 & \frac{ne^{2}\tau}{m_{1}} \end{bmatrix}$$

As we can see, the longitudinal conductivity at X and Y are  $\sigma_{xx}^X = \frac{ne^2\tau}{m_1}$  and  $\sigma_{xx}^Y = \frac{ne^2\tau}{m_2}$ , respectively, and there is no transverse conductivity