Supplementary Information
Halogen Functionalization-Induced Modulation of Ferromagnetism and Electronic Phases in CrXY Monolayers

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1. The AIMD simulation results and phonon dispersion of the $CrSSeF_2$ monolayer

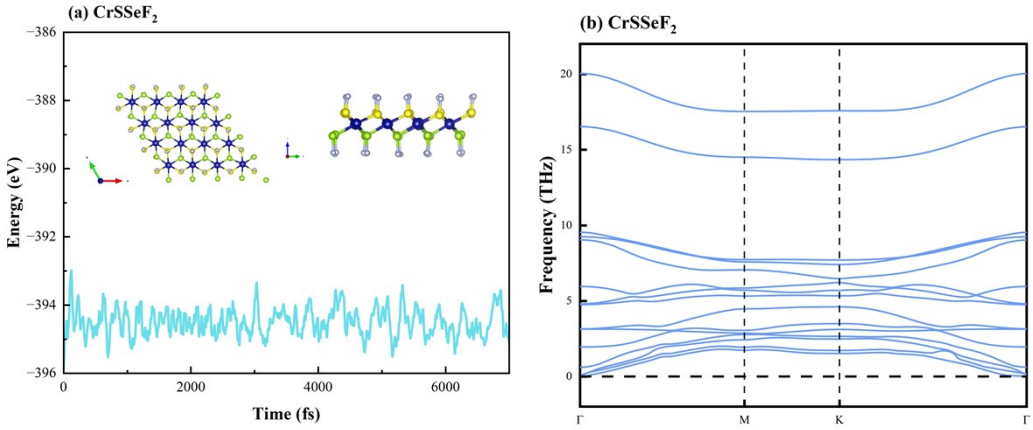

Fig. S1 (a) The variation of temperature as a function of step at temperatures of 300 K for the $CrSSeF_2$ monolayer. (b) The phonon dispersion of $CrSSeF_2$ monolayer.

2. The schematic plot of PDOS of the CrSSe monolayer

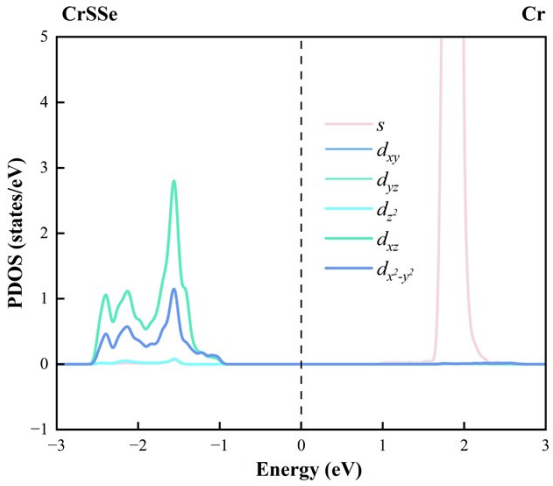

Fig. S2 The PDOS of the CrSSe monolayer.

3. Orbital-resolved magnetic anisotropy energies of the $CrSTeBr_2$ monolayer

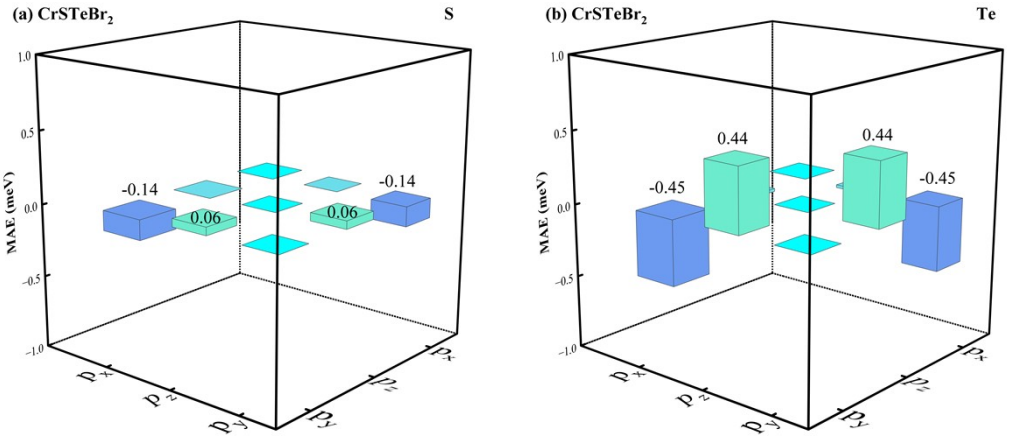

Fig. S3 Orbital-resolved MAE of the $CrSTeBr_2$ monolayer.

4. The difference of spin-orbital angular momentum matrix elements

Table S1 The matrix differences for *p* orbitals between magnetization along *x* [100] and *z* [001] directions. u^-, o^+, o^- represent unoccupied spin-down states, occupied spin-up and spin-down states, respectively.

u^-	o^+			o^-		
	p_y	p_z	p_x	p_y	p_z	p_x
p_y	0	-1	1	0	1	-1
p_z	-1	0	0	1	0	0
p_x	1	0	0	-1	0	0

Table S2 The matrix differences for *d* orbitals between magnetization along *x* [100] and *z* [001] directions.

u^-	o^+					o^-				
	d_{xy}	d_{yz}	d_{z^2}	d_{xz}	$d_{x^2-y^2}$	d_{xy}	d_{yz}	d_{z^2}	d_{xz}	$d_{x^2-y^2}$
d_{xy}	0	0	0	1	-4	0	0	0	-1	4
d_{yz}	0	0	3	-1	1	0	0	-3	1	-1
d_{z^2}	0	3	0	0	0	0	-3	0	0	0
d_{xz}	1	-1	0	0	0	-1	1	0	0	0
$d_{x^2-y^2}$	-4	1	0	0	0	4	-1	0	0	0

5. The Curie temperature T_C of the CrXY and $CrXYT_2$ monolayers

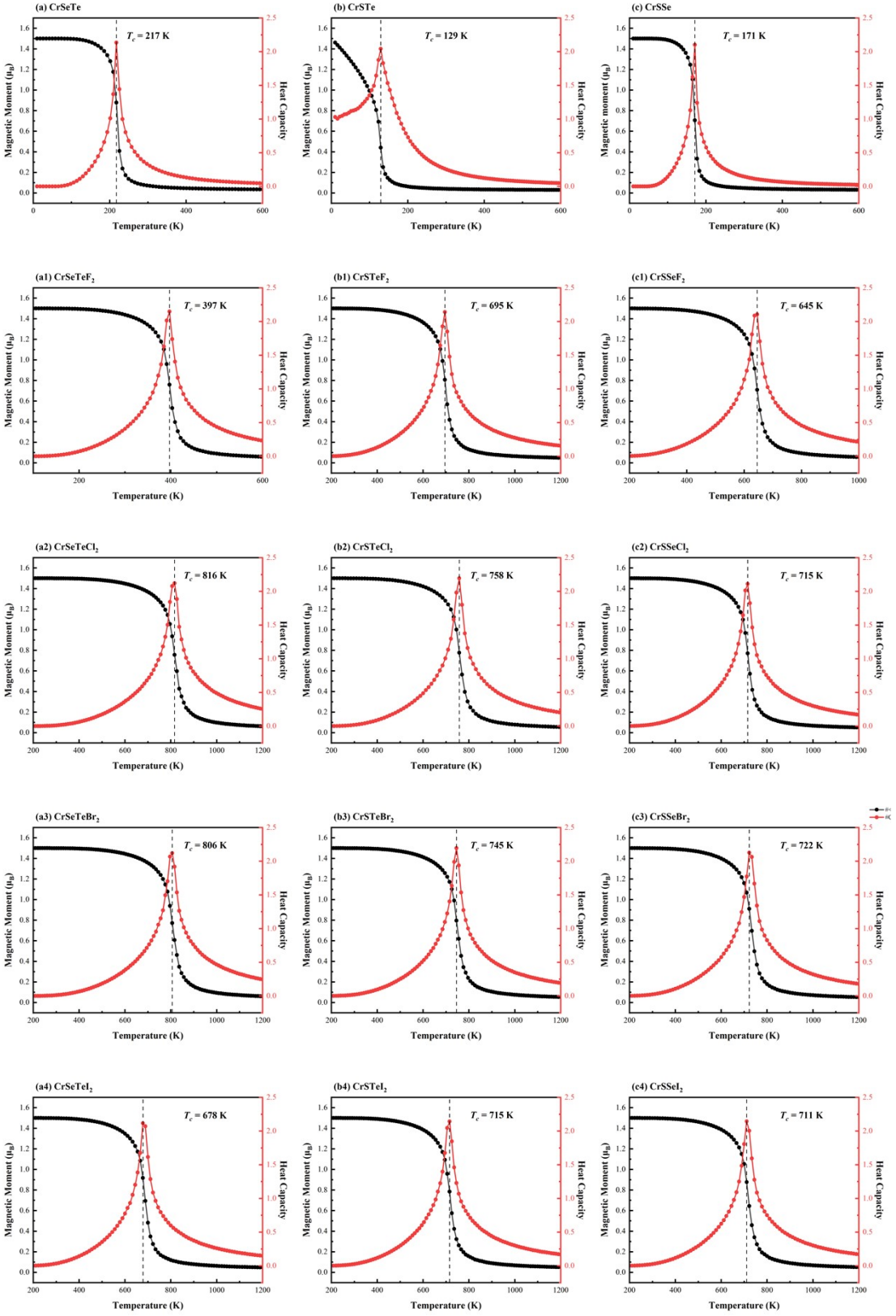

Fig. S4 Temperature dependence of the magnetic moment M and specific heat capacity C_v. The T_C for CrXY and $CrXYT_2$ monolayers are identified by the peak position in C_v and the transition point of M.