High Curie temperature ferromagnetic monolayer T-CrSH and valley physics of T-CrSH/WS$_2$ heterostructure

Xiaole Qiu, Bing Liu, Lin Ge, Lianzhen Cao, Kai Han and Hongchao Yang*

School of Physics and Electronic Information, Weifang University, Weifang 261061, China

*Corresponding author: hc_yang90@163.com

Table S1 Relative energies (meV/Cr) of Stripe AFM, Dimer AFM, Zigzag AFM, and Non-collinear AFM states for the monolayer Janus T-CrSH with respect to that of the FM state at different strains. The energy differences between FM and AFM states can be calculated by $\Delta E = E_{AFM} - E_{FM}$, where E_{AFM} corresponds to the AFM state with the lowest energy, that is, in the range of -3%-5% strains E_{AFM} represents the energy of Dimer AFM state, while at the -4% and -5% strains E_{AFM} represents the energy of Zigzag AFM state.

Strain	FM	Stripe AFM	Dimer AFM	Zigzag AFM	Non-collinear AFM	ΔE
			(meV)			
-5 %	0.00	-17.40	-16.94	-36.61	-30.39	-36.61
-4 %	0.00	4.12	-4.19	-12.96	-4.765	-12.96
-3 %	0.00	23.49	7.22	8.315	18.41	7.22
-2 %	0.00	40.85	17.35	27.41	39.35	17.35
-1 %	0.00	56.25	26.31	44.38	58.01	26.31
0 %	0.00	53.01	27.81	43.50	56.33	27.81
1 %	0.00	81.58	40.91	72.43	89.01	40.91
2 %	0.00	91.71	46.74	83.66	101.62	46.74
3 %	0.00	100.32	51.70	93.28	112.46	51.70
4 %	0.00	107.43	55.75	101.27	121.61	55.750

5 %	0.00	113.13	58.98	107.58	129.19	58.98

Table S2 Relative energies (meV) of the T-CrSH/WS$_2$ heterostructures (a), (b), (d), (e) and (f) with respect to that of the configuration (c).

configuration	a	b	c	d	e	f
ΔE	16.67	114.78	0	91.29	143.24	93.37
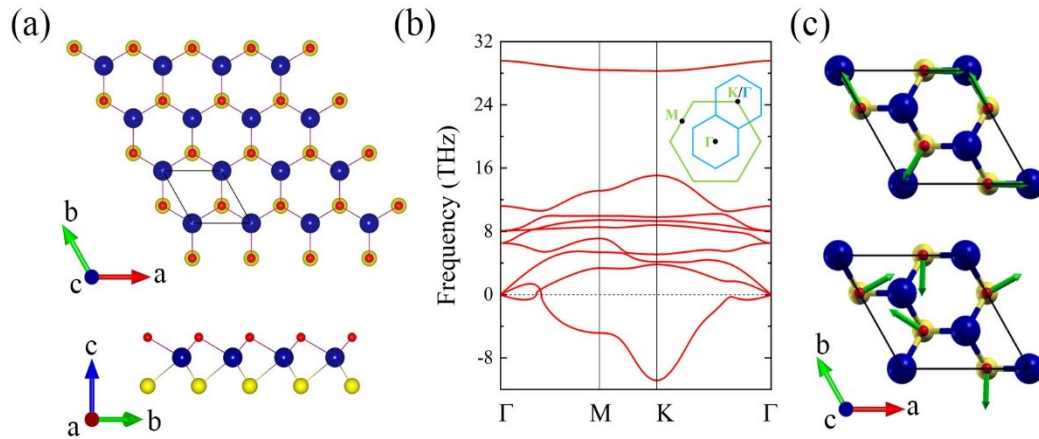
Fig. S1 (a) Top and side view of monolayer Janus H-CrSH. The unit cell is outlined by the black solid line in the top view. The blue, red, and yellow balls represent Cr, H and S atoms, respectively. (b) Phonon spectrum of the monolayer Janus CrSH along the high-symmetry paths of the BZ. Inset shows the BZs of the unit cell (green) and the $\sqrt{3} \times \sqrt{3}$ supercell (blue), where the K point of the former coincides with the Γ point of the latter. (c) Displacement patterns of two doubly degenerated imaginary phonon modes at the K point of the unit cell BZ.
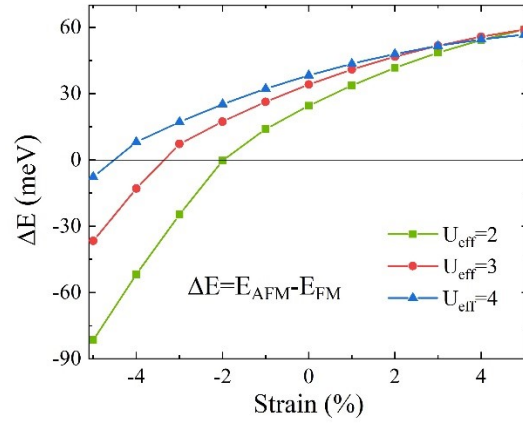
Fig. S2. Variation of the energy difference between AFM and FM states as a function of the biaxial strain of Janus T-CrSH.
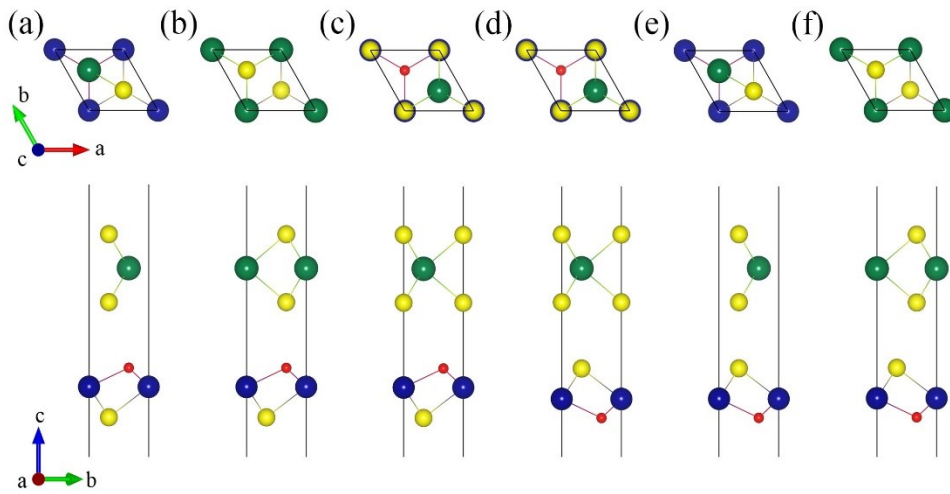

Fig. S3 (a) to (f) are top and side views of six T-CrSH/WS$_2$ heterostructures. The blue, red, yellow and green balls represent Cr, H, S and W atoms, respectively.
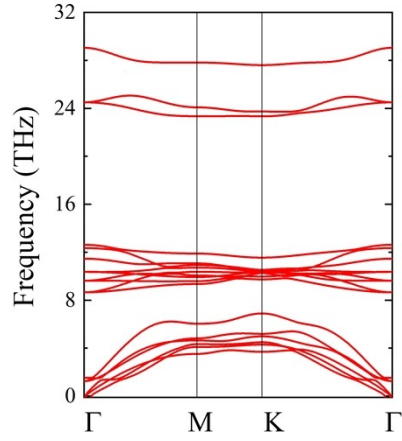
Fig. S4 Phonon spectrum of the T-CrSH/WS$_2$ heterostructure along the high-symmetry paths of the BZ.
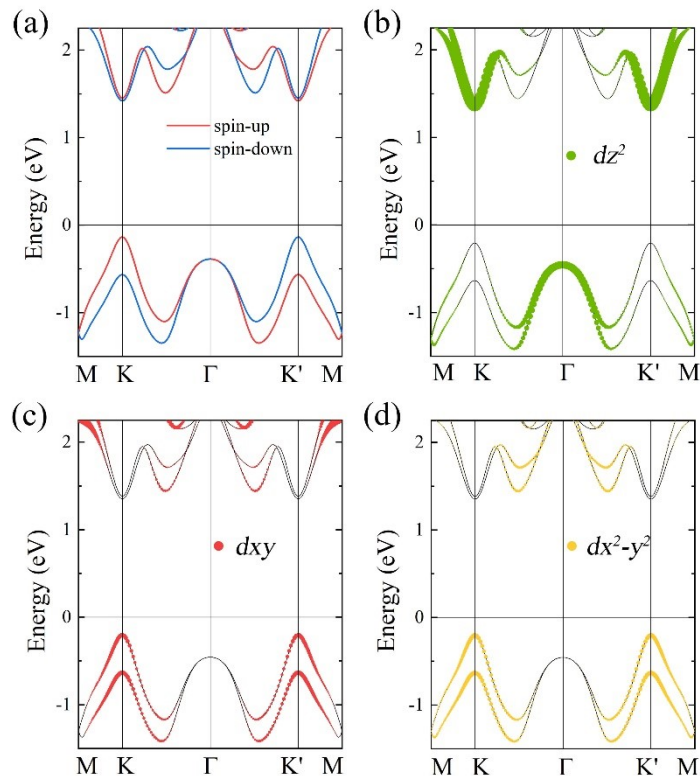

Fig. S5 (a) Spin-polarized and (b)-(d) the W-*d*-resolved band structures of monolayer WS$_2$ with SOC effect.
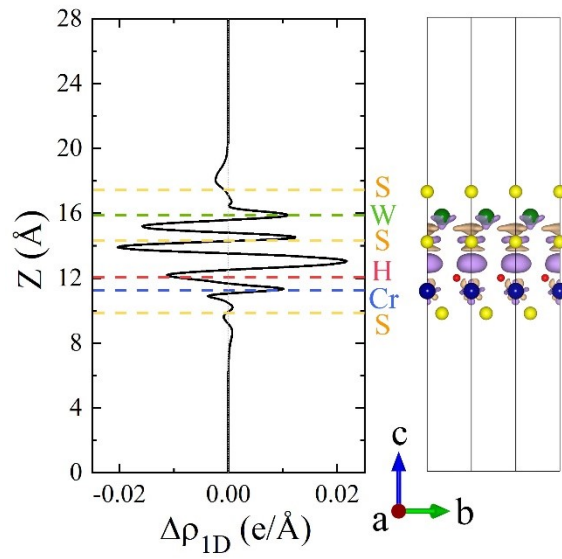
Fig. S6 Left and right panels represent one and three-dimensional charge difference densities for the T-CrSH/WS$_2$ heterostructure, respectively. In the left panel, the atomic positions are marked by the dotted lines. In the right panel, the value of the isosurface is set to 0.0004 $e\text{Å}^{-3}$, and the purple (orange) distribution corresponds to charge accumulation (depletion).