Supporting Information for 2D hybrid $CrCl_2(N_2C_4H_4)_2$ with tunable ferromagnetic half-metallicity

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Table S1. Relative total energy ΔE (meV/f.u.) and lattice constants (a, b, and c in unit of Å) for bulk structures α, β, γ, and δ of $CrCl_2$(pyrazine)$_2$ with full structural optimization, see also Fig. 5 in the main text. The optimized lattice constants of the most stable α structure agree well with the experimental ones of a = 6.90 Å, b =6.97 Å and c = 10.83 Å (Ref. 37).

Bulk	ΔE	a	b	c
α	0	6.87	6.88	10.36
β	248	6.93	6.94	10.45
γ	$\rightarrow \alpha$			
δ	$\rightarrow \alpha$			

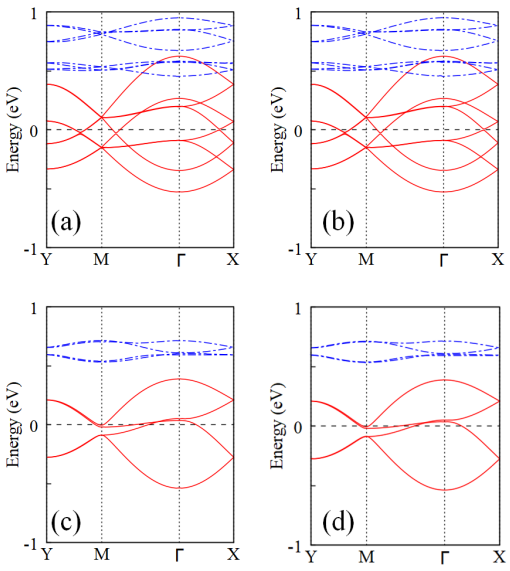

Fig. S1. Band structure of bulk $CrCl_2$(pyrazine)$_2$ calculated by (a) GGA +U and (b) GGA+U+SOC. Band structure of monolayer $CrCl_2$(pyrazine)$_2$ calculated by (c) GGA +U and (d) GGA+U+SOC. The blue (red) lines stand for the up (down) spin. The Fermi level is set at zero energy.

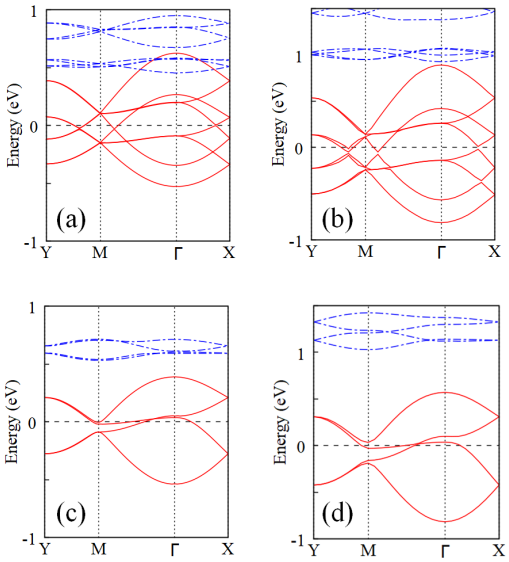

Fig. S2. Band structure of bulk $CrCl_2$(pyrazine)$_2$ calculated by (a) GGA + U and (b) HSE06. Band structure of monolayer $CrCl_2$(pyrazine)$_2$ calculated by (c) GGA + U and (d) HSE06. The blue (red) lines stand for the up (down) spin. The Fermi level is set at zero energy.