Supporting Information for

Prediction of Two-dimensional Large-gap Magnetic Semiconductors in Transition Metal Superhalogenides

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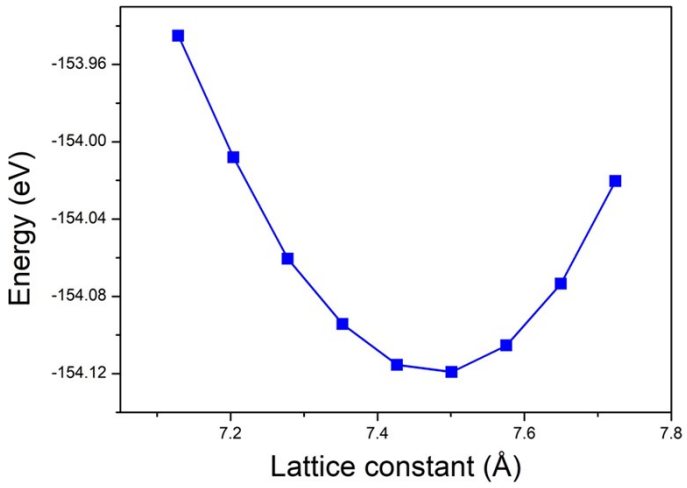

Fig. S1. Total energy as a function of lattice constants for $Co[B(CN)_4]_2$ monolayer.

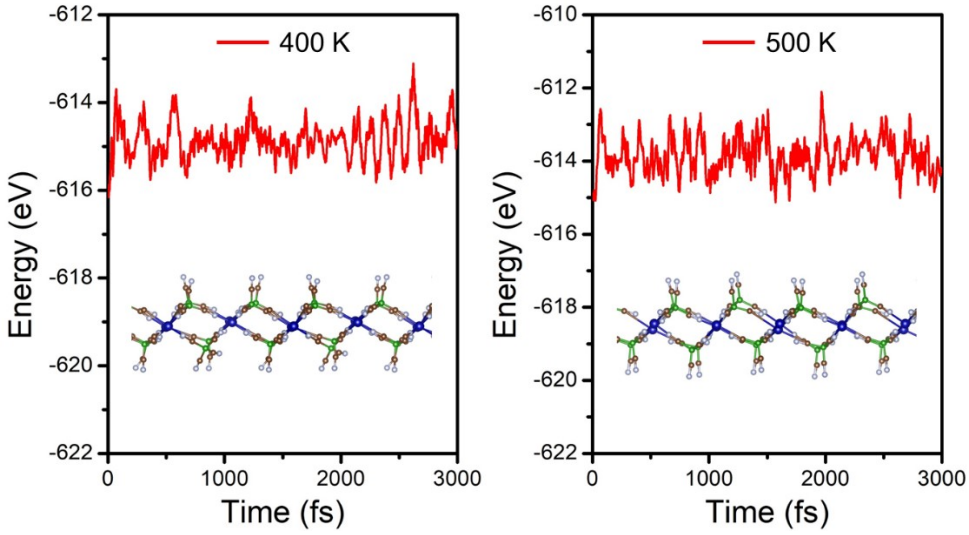

Fig. S2. Fluctuation of the total energy during the molecular dynamical simulation process at 400 and 500 K. Inset: Side view of $Co[B(CN)_4]_2$ monolayer after a simulation time of 3000 fs.

We have also further performed AIMD simulations at 800 K. The results show that the bonds between $B(CN)_4$ and Co atoms are broken and the atomic structural framework is destroyed during the simulation process. Therefore, the $Co[B(CN)_4]_2$ monolayer may not survive at a high temperature of 800 K, but could be stable at a temperature up to 500 K.

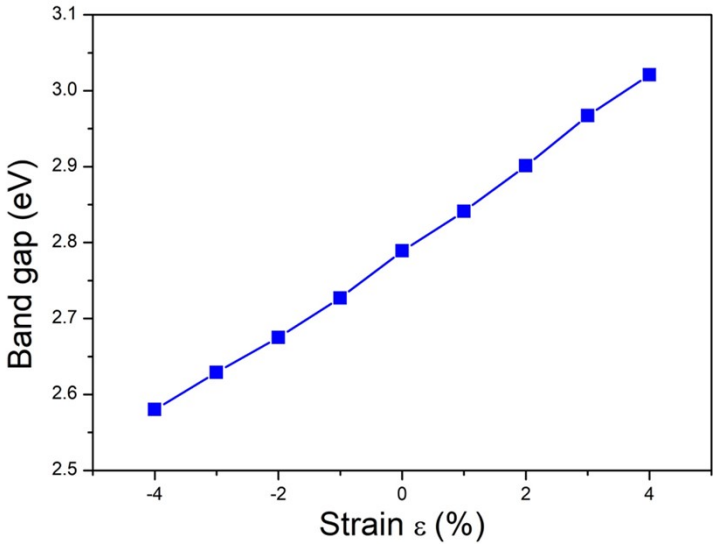

Fig. S3. Electronic band gap as a function of in-plane biaxial strain for the $Co[B(CN)_4]_2$ monolayer calculated by PBE+U (3 eV) method.

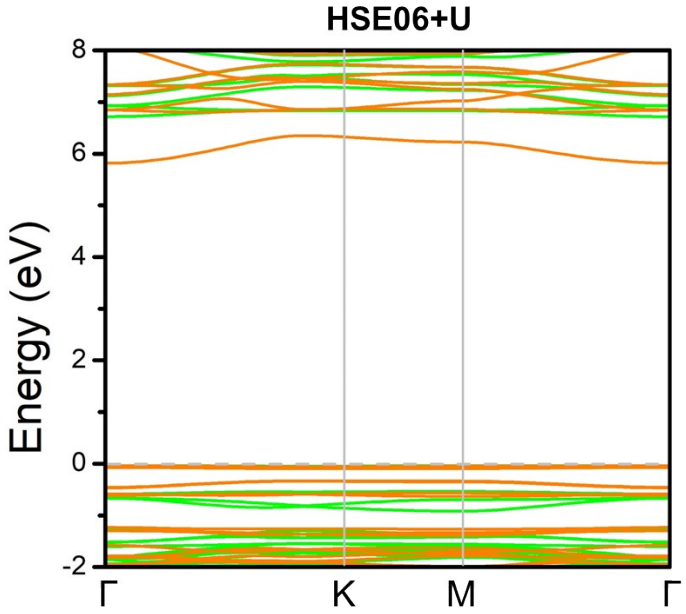

Fig. S4. Electronic band structure of $Co[B(CN)_4]_2$ monolayer calculated by HSE06+U method. Green and orange profiles represent spin-up and -down bands, respectively.

Table S1. The electronic band gap (G), total magnetic moment per unitcell (Mtot), magnetic exchange energy ($E_{ex} = E_{AFM} - E_{FM}$) and estimated Curie temperature (T_C) for $Co[B(CN)_4]_2$ monolayer calculated by different methods.

Method	G (eV)	M_{tot} (μ_B)	E_{ex} (meV)
PBE	0.51	3	12.7
PBE+U(1 eV)	1.22	3	15.5
PBE+U(2 eV)	2.03	3	17.3
PBE+U(3 eV)	2.78	3	19.2
PBE+U(4 eV)	3.49	3	20.7
HSE06	4.18	3	