

Supplemental Material for
Mechanical, Magnetic and Electronic Properties of 2D MSX (M = Ti,
V, Co and Ni, X = Br and I)

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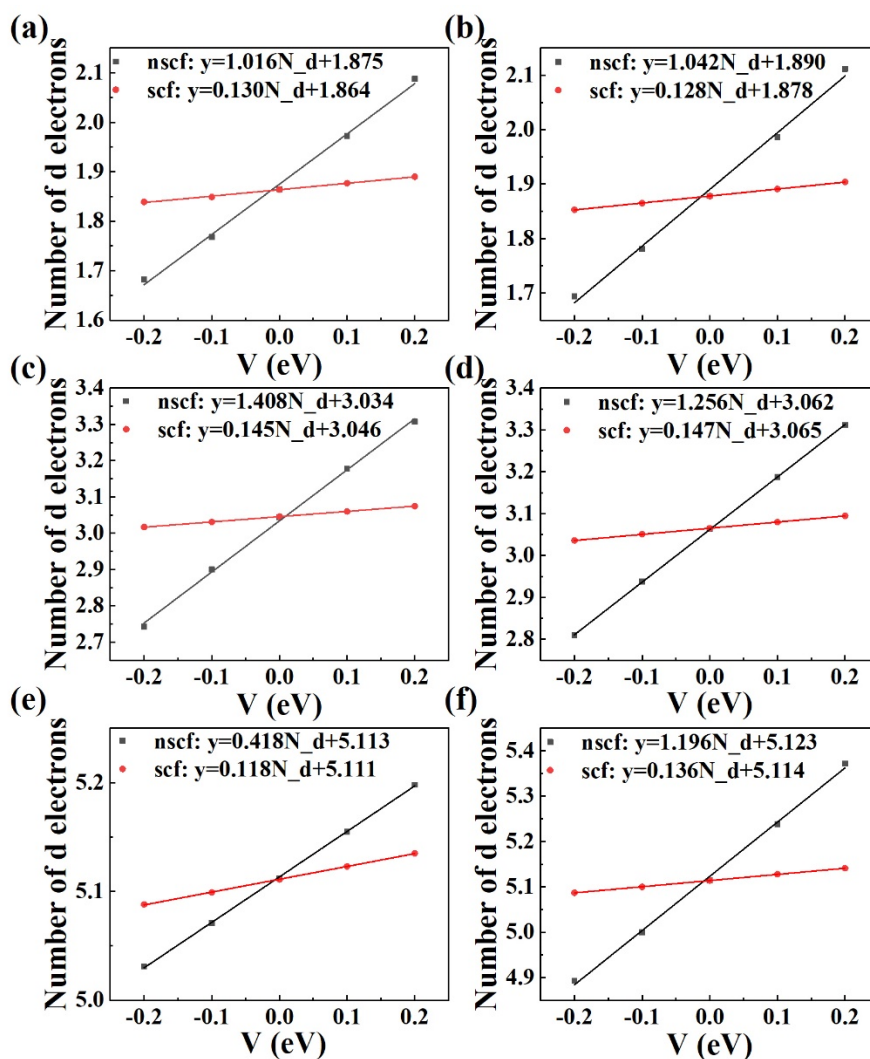


Figure S1 Linear fitting calculations for determining U_{eff} , illustrated as the relationship between the number of *d*-electrons on transition metals and the additional potential (V) for (a) TiSBr,

(b) TiSI, (c) VSBr, (d) VSI, (e) MnSBr, and (f) MnSI.

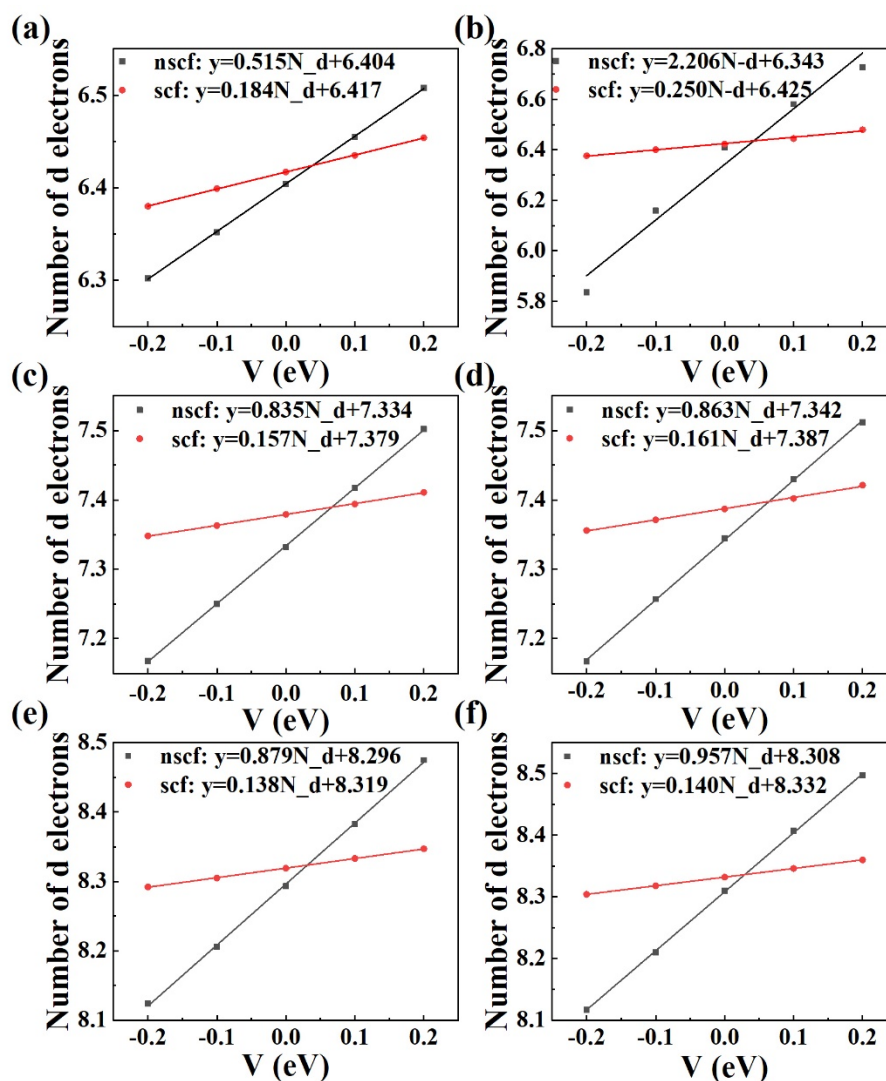


Figure S2 Linear fitting calculations for determining U_{eff} , illustrated as the relationship between the number of *d*-electrons on transition metals and the additional potential (*V*) for (a) FeSBr, (b) FeSI, (c) CoSBr, (d) CoSI, (e) NiSBr, and (f) NiSI.

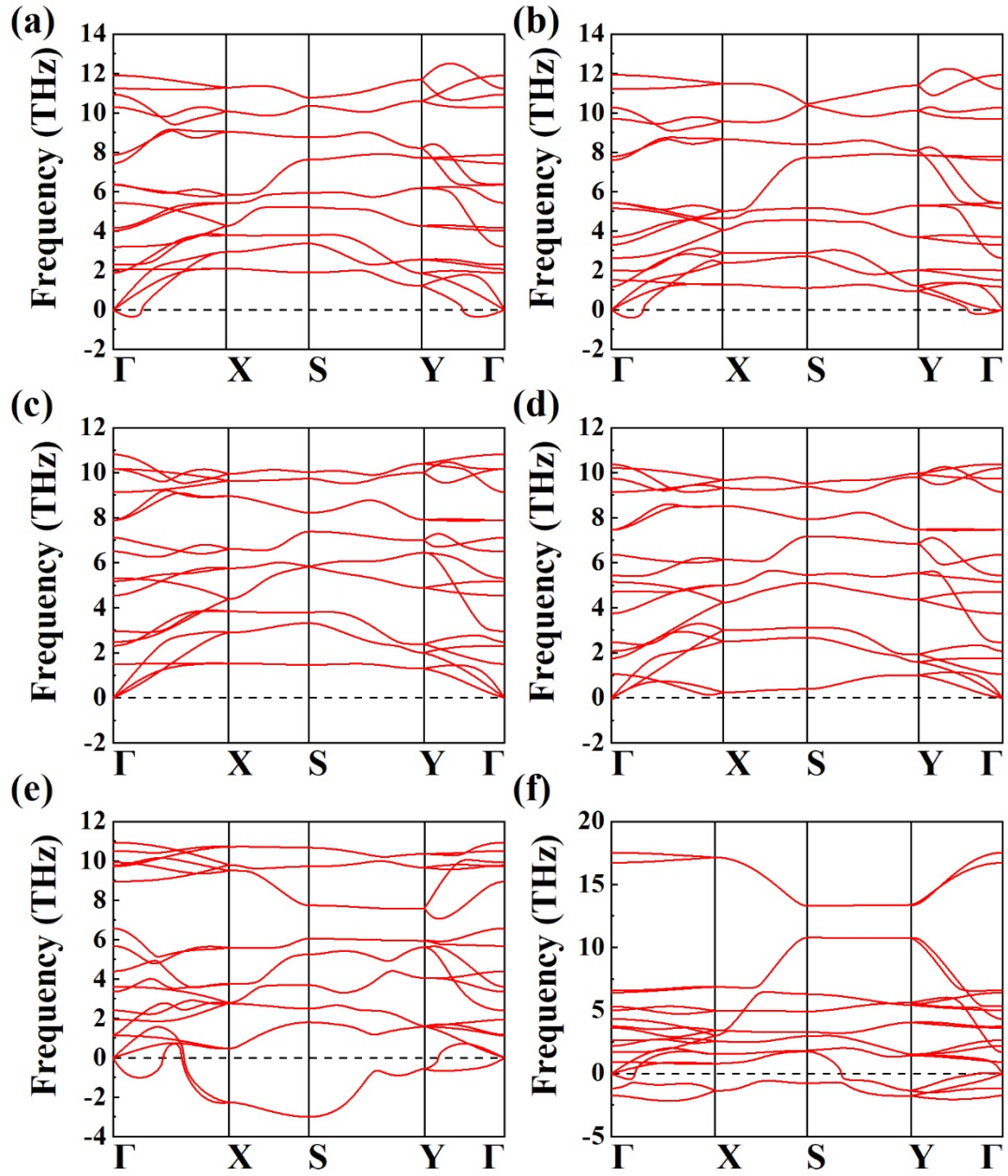


Figure S3 Phonon dispersions for (a) TiSBr, (b) TiSI, (c) VSBr, (d) VSI, (e) MnSBr, and (f) MnSI.

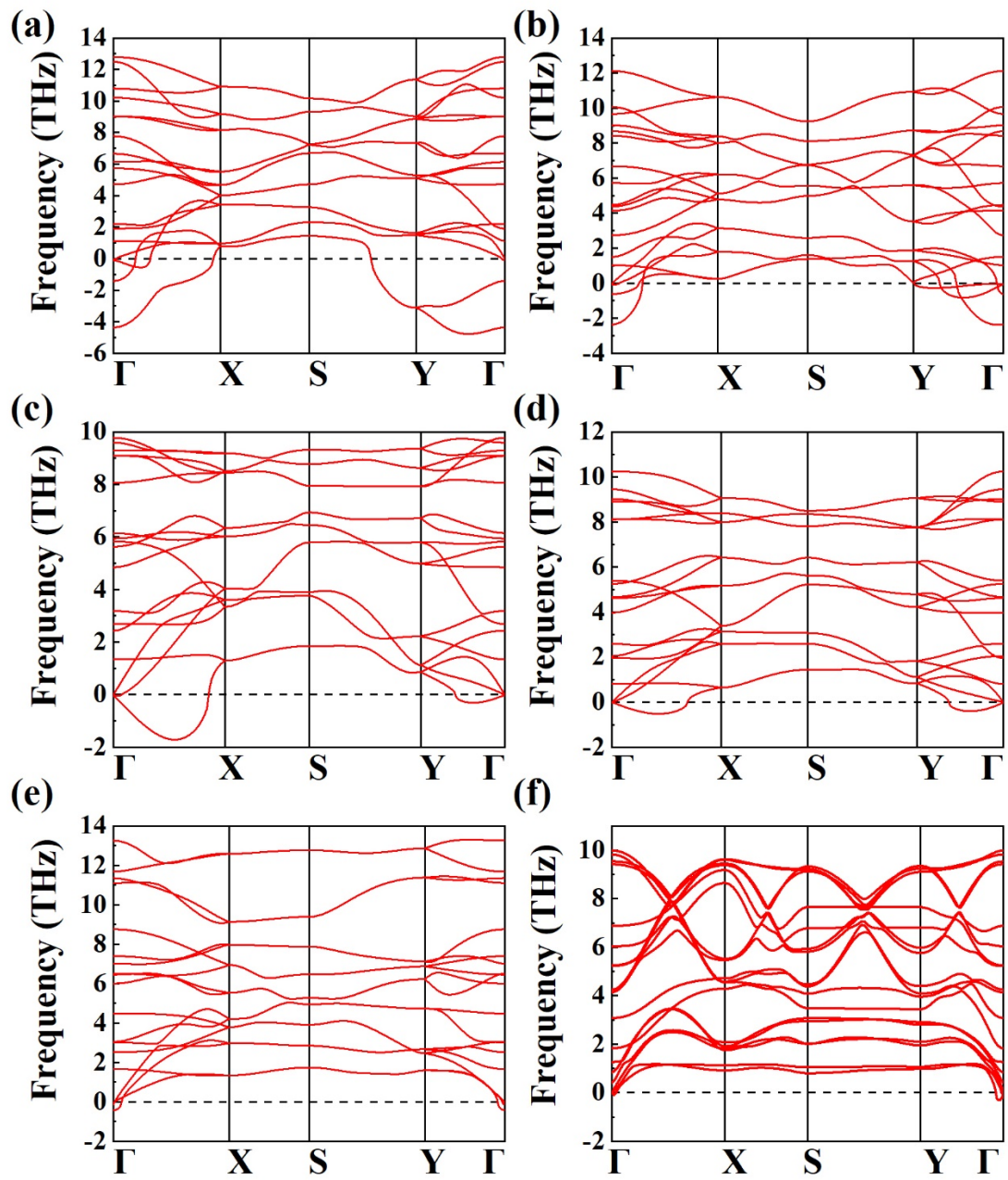


Figure S4 Phonon dispersions for (a) FeSBr, and (b) FeSI, (c) CoSBr, (d) CoSI, (e) NiSBr, and (f) NiSI.

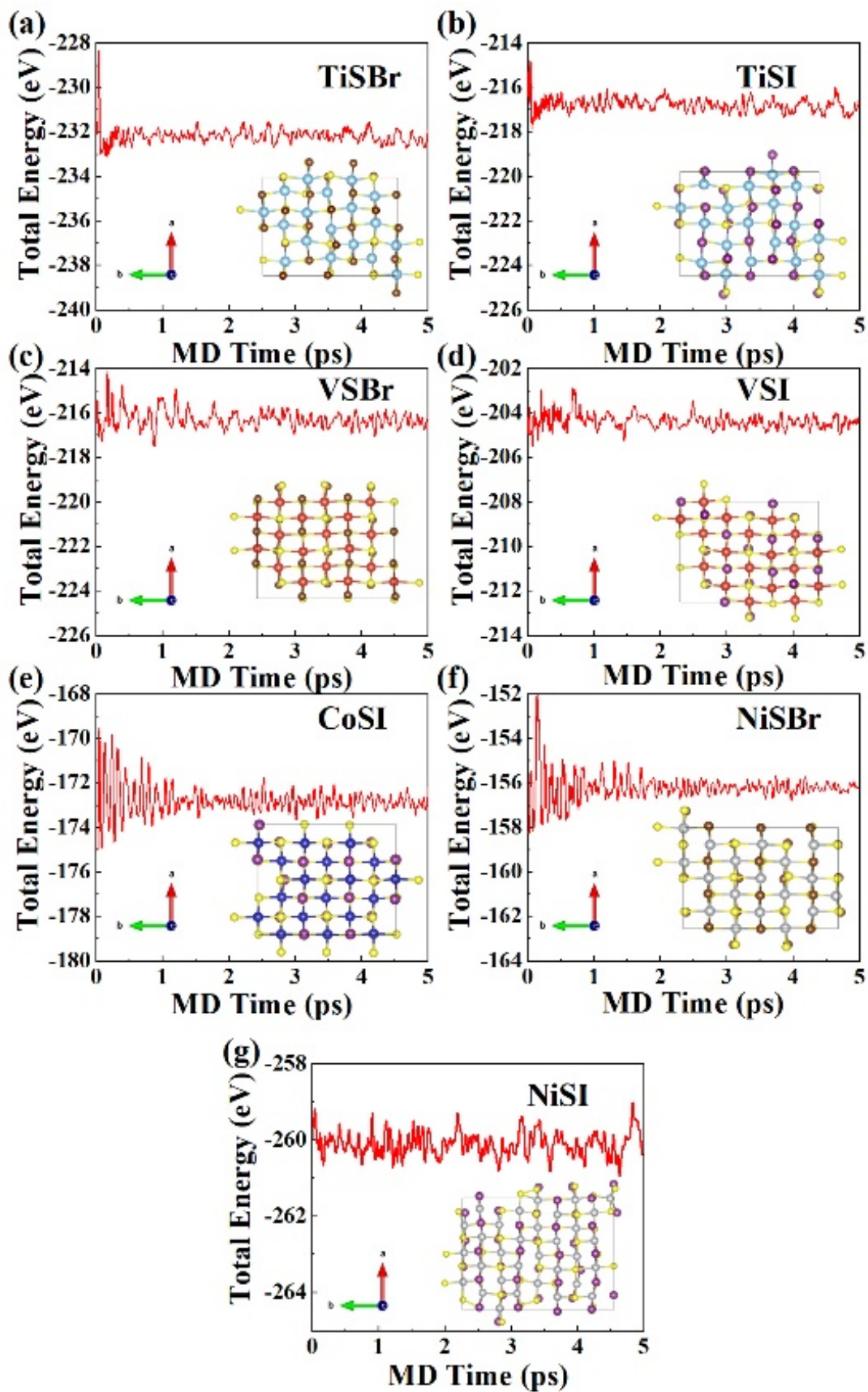


Figure S5 The AIMD simulations of (a) TiSBr, (b) TiSI, (c) VSBr, (d) VSI, (e) CoSI, (f) NiSBr, and (g) NiSI, respectively. The insets are the top views of the structure after the test.

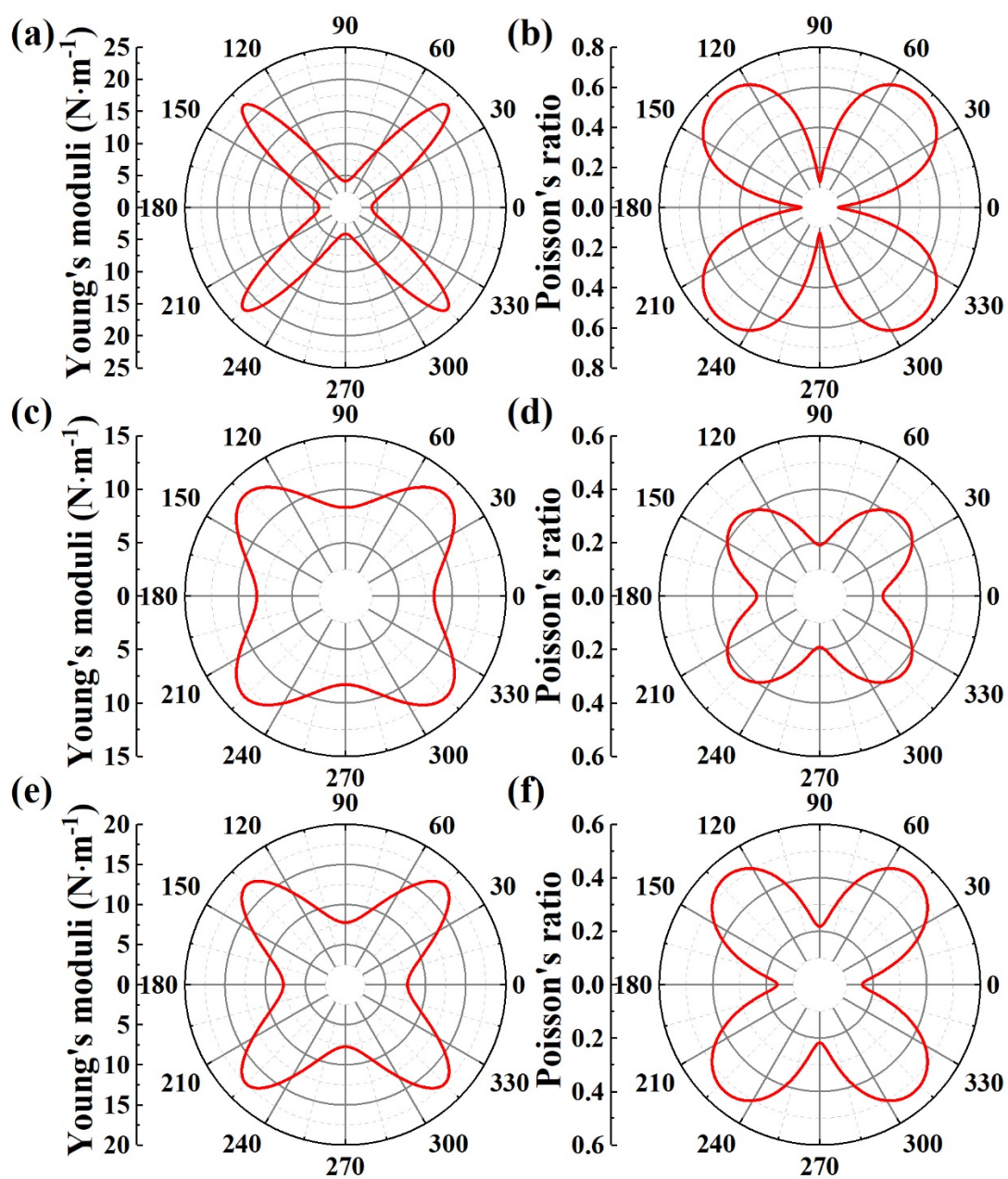


Figure S6 (a) Young's moduli and (b) Poisson's ratio of TiSI, (c) Young's moduli and (d) Poisson's ratio of VSBBr, and (e) Young's moduli and (f) Poisson's ratio of VSI.

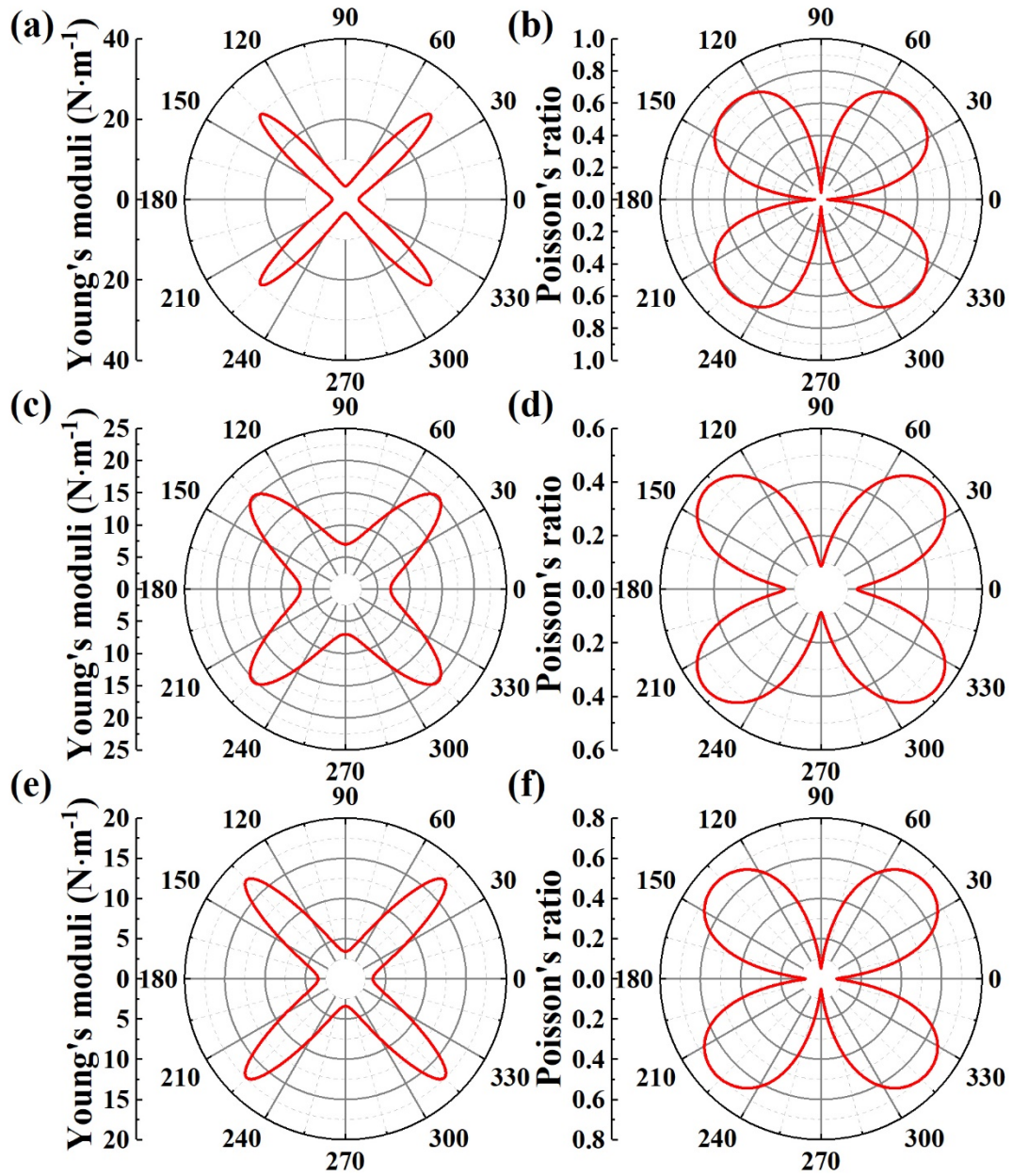


Figure S7 (a) Young's moduli and (b) Poisson's ratio of CoSI, (c) Young's moduli and (d) Poisson's ratio of NiSBr, and (e) Young's moduli and (f) Poisson's ratio of NiSI.

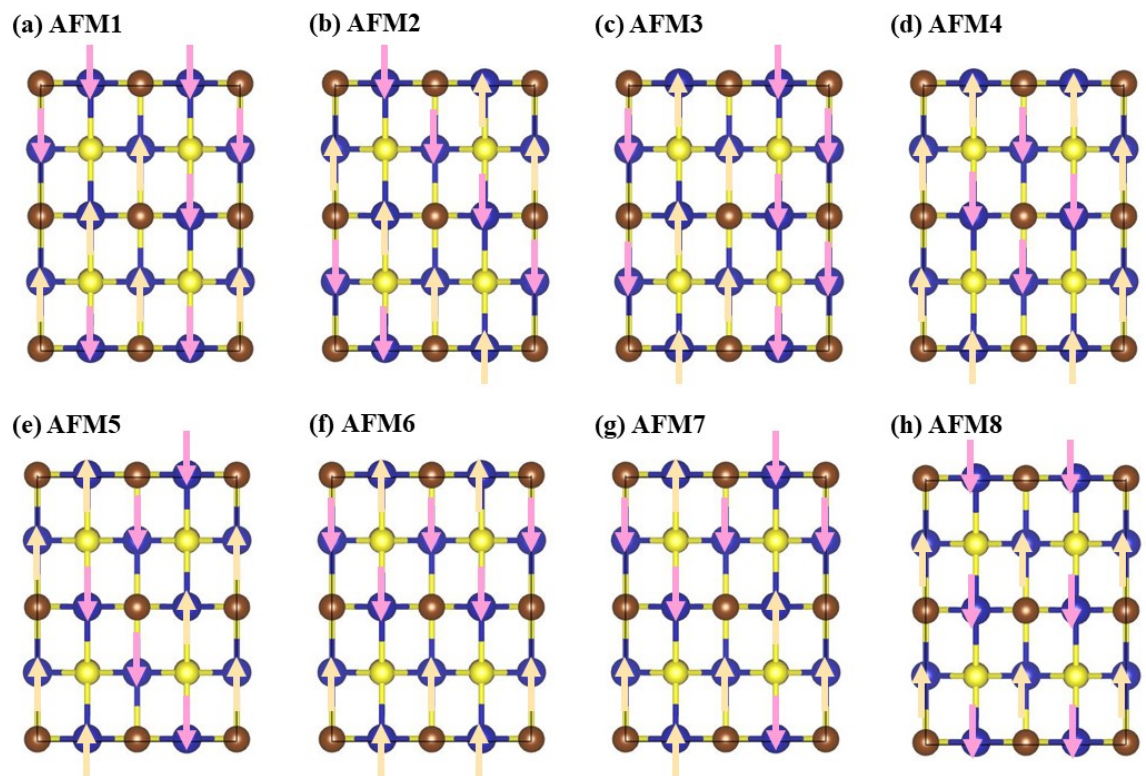


Figure S8 Top views of AFM states in a $2 \times 2 \times 1$ supercell. Blue, yellow, and brown spheres denote M, S, and X atoms, respectively. White and pink arrows represent the spin-up and spin-down moments, respectively.

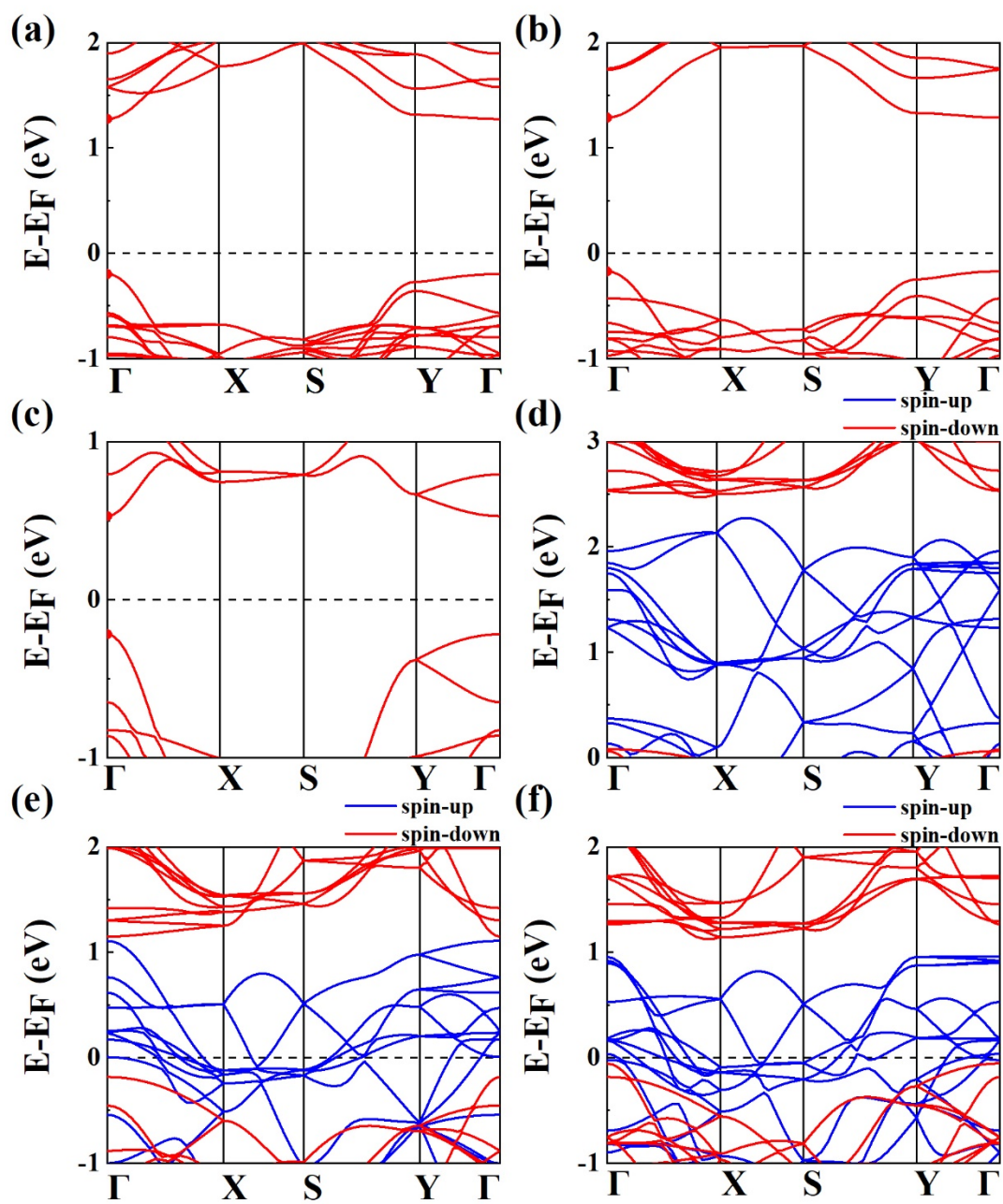


Figure S9 Band structures of (a) TiSI, (b) VSBBr, (c) VSI, (d) CoSI, (e) NiSBr, and (f) NiSI.

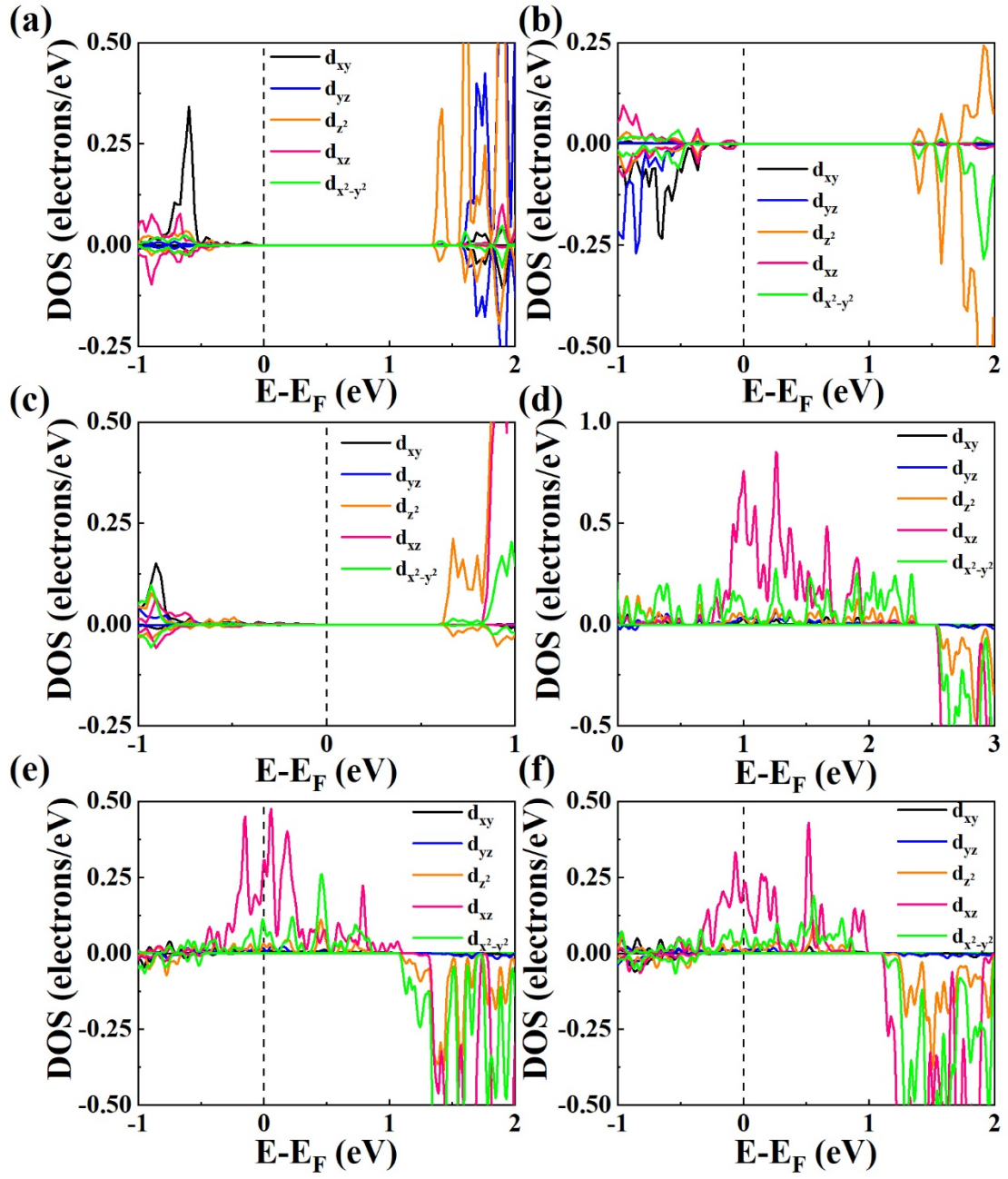


Figure S10 PDOSs of the d electrons of M atoms of (a) TiSI, (b) VSBr, (c) VSI, (d) CoSI, (e) NiSBr, and (f) NiSI.

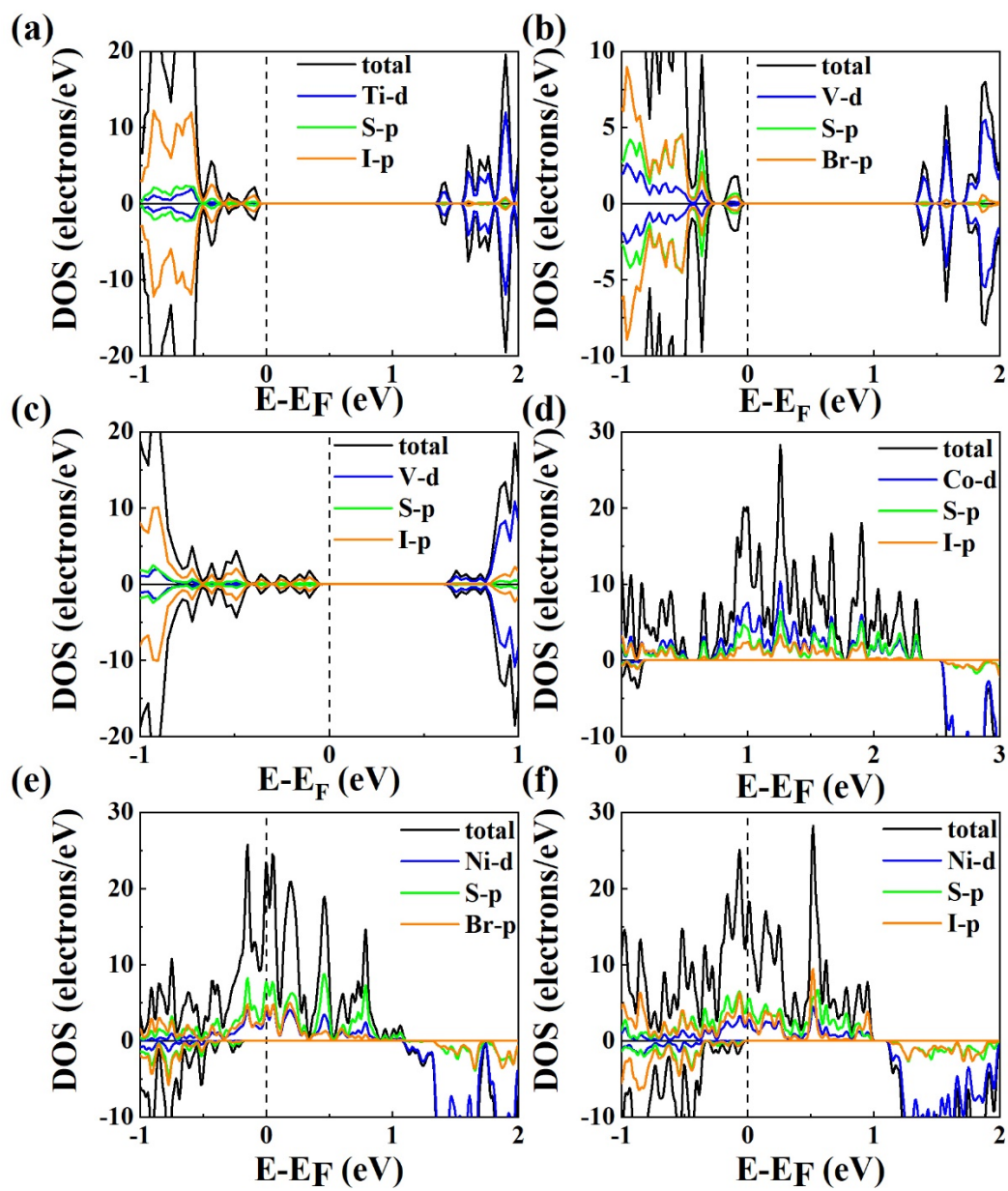


Figure S11 PDOSs of (a) TiSI, (b) VSBr, (c) VSI, (d) CoSI, (e) NiSBr, and (f) NiSI.

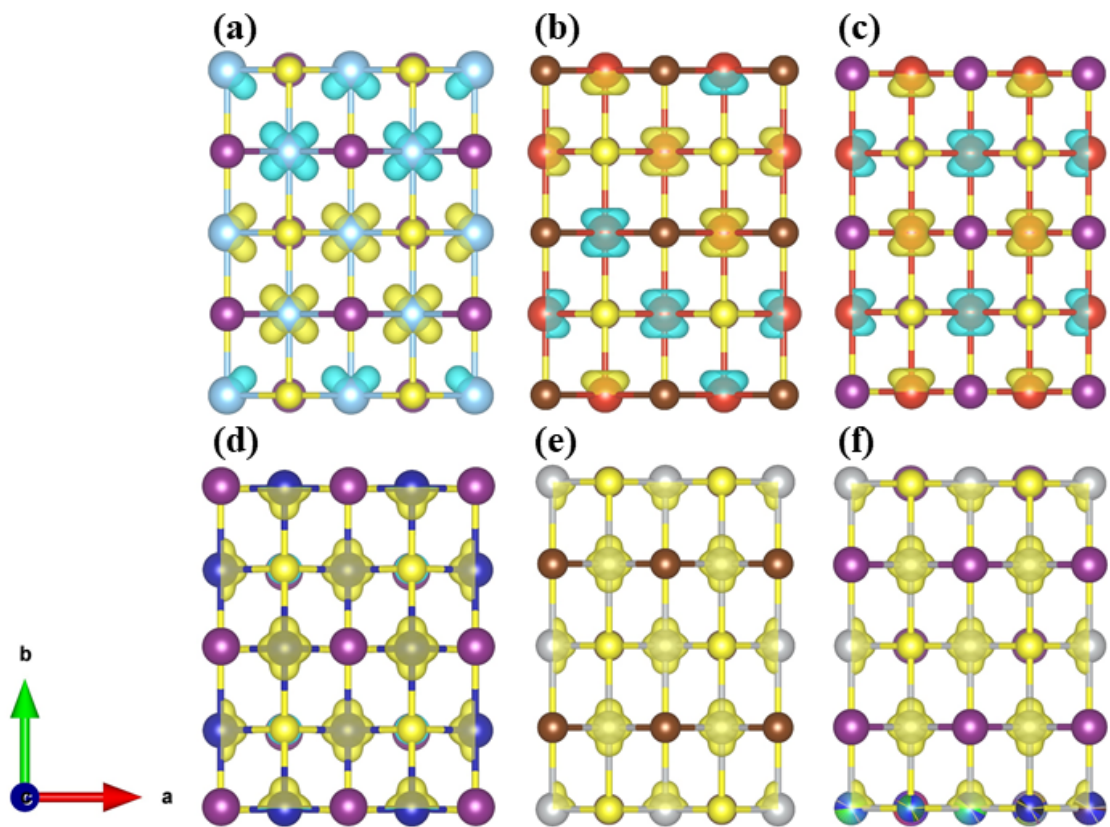


Figure S12 Top views of spin densities in (a) TiSI, (b) VSBr, (c) VSI, (d) CoSI, (e) NiSBr, and (f) NiSI, respectively. The spin-up density is shown in yellow and the spin-down density is shown in blue, respectively.

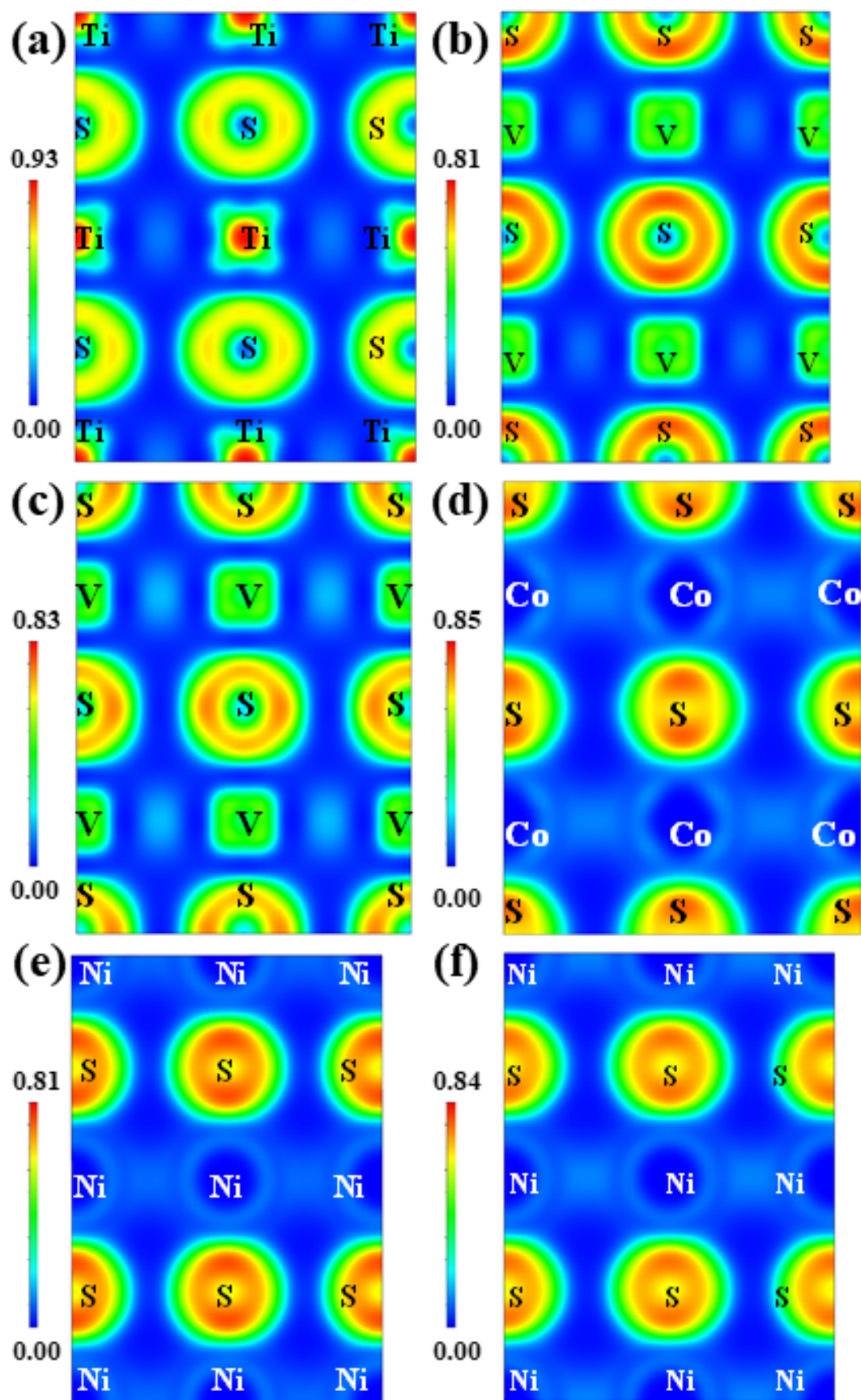


Figure S13 ELFs of (a) TiSI, (b) VSBr, (c) VSI, (d) CoSI, (e) NiSBr, and (f) NiSI on the (001) surfaces.

Table S1 Lattice parameters, bond lengths, and bond angles of the proposed structures.

MSX	a (Å)	b (Å)	l_1 (Å)	l_2 (Å)	l_3 (Å)	θ_1 (°)	θ_2 (°)	θ_3 (°)	$(\theta_1+\theta_2)/2$ (°)
TiSBr	3.643	4.978	2.664	2.494	2.493	93.888	86.295	87.885	90.091
TiSI	3.793	4.982	2.886	2.503	2.493	98.563	82.200	49.639	90.381
VSBr	3.537	5.055	2.573	2.484	2.428	91.112	87.140	54.603	89.126
VSI	3.780	5.029	2.840	2.401	2.468	98.744	79.817	47.943	89.280
MnSBr	3.402	4.909	2.502	2.335	2.475	93.507	85.637	57.489	89.572
MnSI	3.898	4.405	2.751	2.584	2.209	97.914	90.215	50.779	94.064
FeSBr	3.514	4.221	2.432	2.280	2.277	100.801	92.505	58.886	96.653
FeSI	3.611	4.247	2.593	2.320	2.268	102.211	88.256	57.194	95.233
CoSBr	3.398	4.498	2.410	2.258	2.301	97.631	89.658	56.937	93.645
CoSI	3.551	4.488	2.584	2.289	2.299	101.721	86.809	54.809	94.265
NiSBr	3.322	4.802	2.368	2.217	2.436	97.046	89.090	57.465	93.068
NiSI	3.485	4.752	2.541	2.252	2.416	101.386	86.578	55.242	93.982

Table S2 Energies of FM and AFM states of MSX.

MSX	FM (eV)	AFM1 (eV)	AFM2 (eV)	AFM3 (eV)	AFM4 (eV)	AFM5 (eV)	AFM6 (eV)	AFM7 (eV)	AFM8 (eV)	ground state
TiSI	-111.731	-111.793	-111.824	-111.751	-103.274	-111.805	-111.825	-111.825	-111.769	AFM6
VSBr	-109.794	-109.749	-109.836	-109.723	-109.769	-109.780	-109.816	-109.838	-109.610	AFM7
VSI	-102.609	-102.924	-102.942	-102.860	-102.894	-102.906	-102.979	-102.955	-102.983	AFM8
CoSI	-78.584	-78.432	-78.319	-78.516	-78.215	-78.433	-78.306	-78.314	-78.527	FM
NiSBr	-71.959	-71.699	-71.801	-71.703	-70.465	-71.701	-71.855	-71.842	-71.460	FM
NiSI	-67.941	-67.560	-67.579	-67.585	-65.926	-67.556	-67.689	-67.642	-67.404	FM

Table S3 Angular dependent MAEs of CoSI, NiSBr, and NiSI.

MSX	plane	0° (meV)	30° (meV)	60° (meV)	90° (meV)	120° (meV)	150° (meV)	180° (meV)
CoSI	xoy	0	0.287	0.813	1.005	0.813	0.287	0
	yoz	0	0.213	0.725	0.965	0.725	0.213	0
	xoz	1.005	0.954	0.965	0.965	0.965	0.954	1.005
NiSBr	xoy	0	-0.026	-0.056	-0.066	-0.056	-0.026	0
	yoz	0	0.049	0.146	0.195	0.146	0.049	0
	xoz	-0.066	0.003	0.135	0.195	0.135	0.003	-0.066
NiSI	xoy	0	0.334	0.884	1.171	0.884	0.334	0
	yoz	0	0.566	1.652	2.120	1.652	0.566	0
	xoz	1.171	1.384	1.858	2.120	1.858	1.384	1.171

Table S4 Bandgaps of semiconducting TiSI, VSBr, and VSI.

MSX	Bandgap (eV)
TiSI	1.473
VSBr	1.459
VSI	0.748