Supplementary Information for

Alloying two-dimensional VSi_2N_4 to realize ideal half-metal towards spintronics

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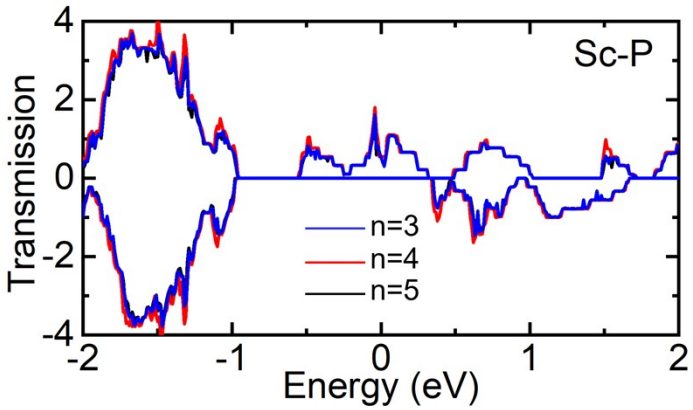

Fig. S1. The zero-bias transmission spectra of the Sc-doped VSi_2N_4 device with the central region containing 3, 4, and 5 units with parallel magnetization configuration.

Table S1. Total energy E_{total} (eV) for the pure and Sc-doped VSi_2N_4 systems with ferromagnetic (FM) and antiferromagnetic (AFM) configurations

E_{total} (eV)	FM	AFM1	AFM2	AFM3
pure	-13491.87306	-13491.87305	-13491.87304	
Sc-doped	-51199.41565	-51199.41565	-51199.41565	-51199.41565

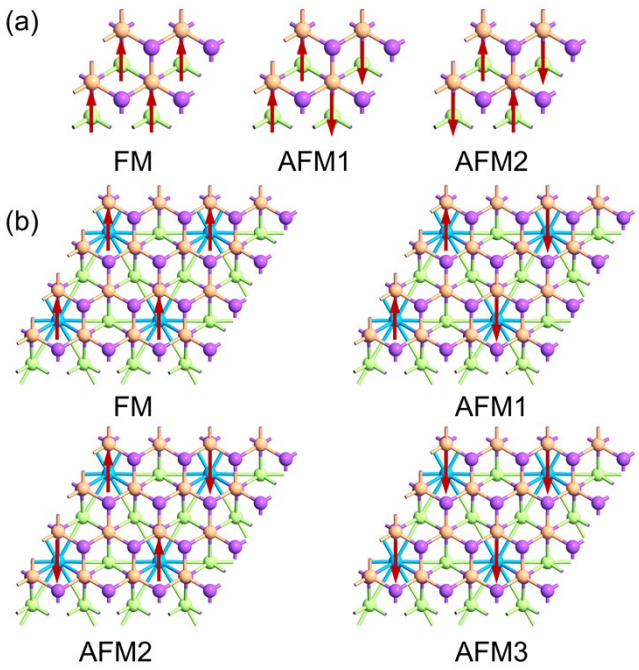

Fig. S2. Ferromagnetic (FM) and antiferromagnetic (AFM) configurations of (a) the pure VSi_2N_4 system and (b) the Sc-doped VSi_2N_4 system. All the V atoms in (b) are in spin-up magnetic order.

Table S2. The formation energy E_{form} (eV), magnetic moment M (μ_B), and band gap E_{gap} (eV) of the transition metal atom (Sc-Ni, Y-Mo) doped VSi_2N_4 system, in comparison of those with U correction (U=3.0 eV).

System	E_{form}	M	E_{gap_up}	E_{gap_down}	M ($+U$)	E_{gap_up} ($+U$)	E_{gap_down} ($+U$)
pure	---	0.98	0.00	1.74	1.00	0.05	2.00
Sc	−3.08	2.00	0.00	1.32	2.00	0.00	1.92
Ti	−5.20	3.00	0.21	1.68	3.00	1.54	1.94
Cr	−2.39	2.15	0.00	0.00	5.00	0.00	2.32
Mn	−1.80	0.00	0.23	0.23	6.00	1.24	2.23
Fe	−2.37	0.00	0.00	0.00	7.00	0.00	1.83
Co	−1.76	0.00	0.00	0.00	4.00	0.94	0.00
Ni	0.08	1.20	0.00	0.00	3.00	0.73	1.23
Y	−1.92	2.00	0.00	0.90	2.00	0.00	1.61
Zr	−6.25	3.00	0.30	1.50	3.00	1.25	1.72
Nb	−6.95	3.70	0.00	0.00	4.00	0.29	1.89
Mo	−5.40	1.93	0.00	0.00	3.43	0.00	0.00

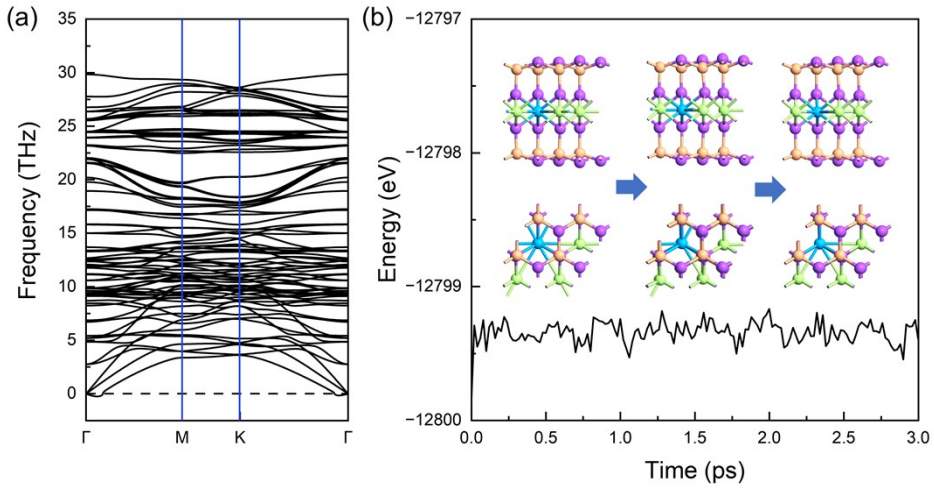

Fig. S3. (a) The phonon spectra and (b) AIMD simulations of the Sc-doped VSi_2N_4 system. The molecular dynamics at 300 K for 3 ps with 1 fs time step: total energy and structure over time.

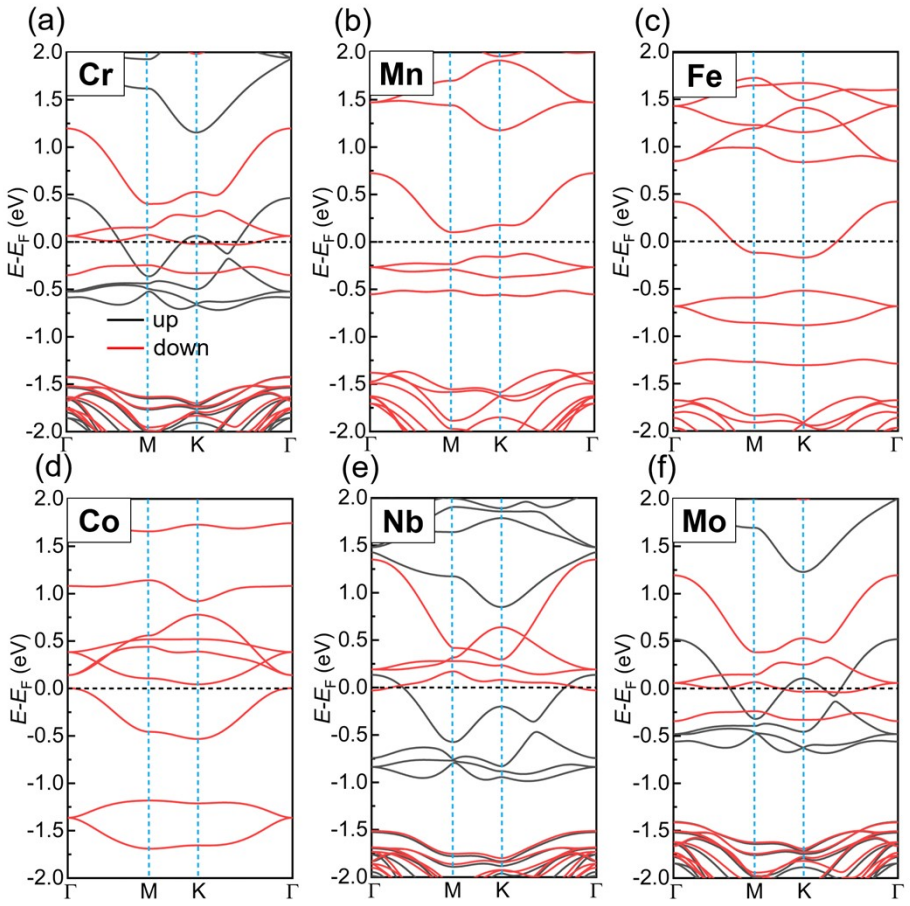

Fig. S4. The band structures of the transition metal atoms (Cr, Mn, Fe, Co, Nb, and Mo) doped VSi_2N_4 monolayers.

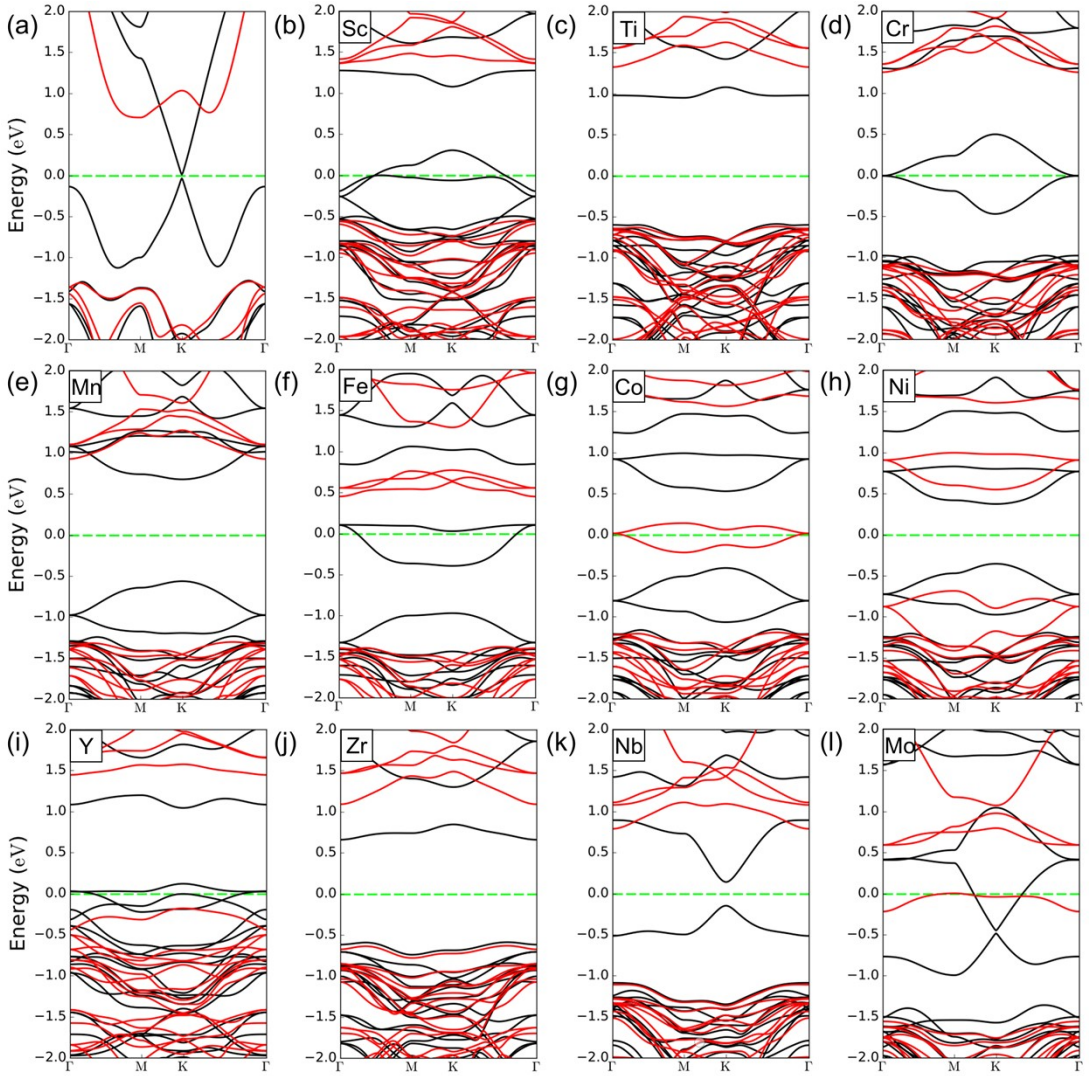

Fig. S5. The band structures of (a) the pure VSi_2N_4 and (b-l) the transition metal atom (Sc-Ni, Y-Mo) doped VSi_2N_4 systems within the Hubbard U approach (U=3.0 eV). Spin-up and spin-down bands are denoted by the black and red lines, respectively.

Table S3. Spin bandgap (E_{gap_up} and E_{gap_down}) of the Sc-doped system with one Sc atom doped in 2*2, 3*3, and 4*4 supercell.

Sc-doped system	2*2	3*3	4*4
E_{gap_up} (eV)	0.00	0.00	0.00
E_{gap_down} (eV)	1.32	1.29	1.33

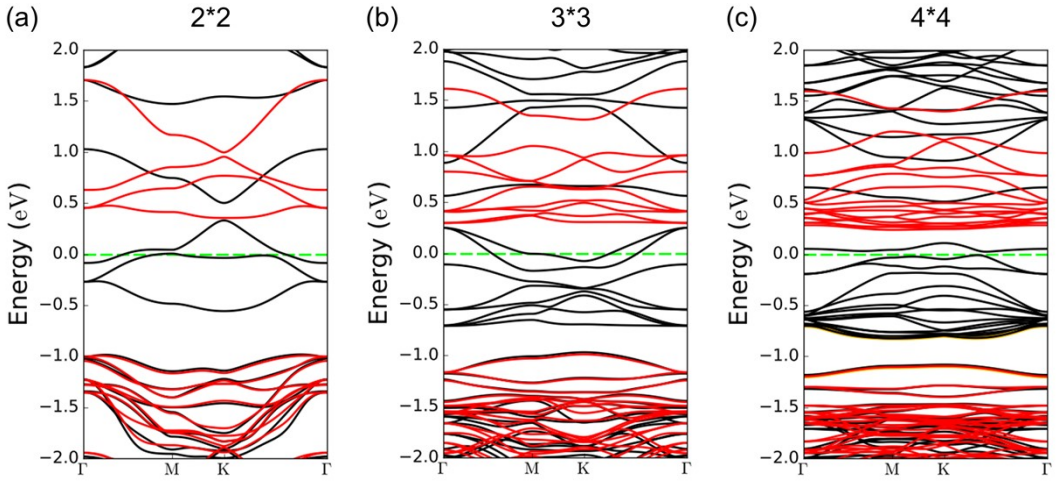

Fig. S6. The energy band of the Sc-doped system with one Sc atom doped in (a) 2*2, (b) 3*3, and (c) 4*4 supercell.

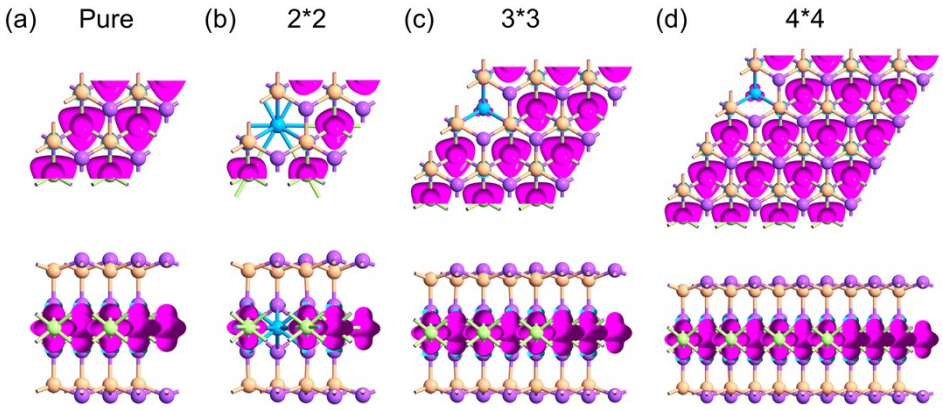

Fig. S7. Spin electron density of (a) the pure VSi_2N_4 system and the Sc-doped VSi_2N_4 system with (b) 2*2, (c) 3*3, and (d) 4*4 supercell. The pink color shows the up-spin electrons. The isovalue is 0.05.