Table S1. Calculated Bader charge at atomic sites in the M_4N phases

Compound	Atom	I-M_4N	II-M_4N
Sc_4N	M1	+0.98	+0.69
	M2	+0.43	+0.33
	N	-2.25	-20.5
Ti_4N	M1	+0.96	+0.73
	M2	+0.45	+0.34
	N	-2.31	-2.13
V_4N	M1	+0.96	+0.49
	M2	+0.43	+0.36
	N	-2.24	-1.70
Cr_4N	M1	+0.96	+0.45
	M2	+0.35	+0.34
	N	-2.01	-1.57
Mn_4N	M1	+1.08	+0.47
	M2	+0.23	+0.27
	N	-1.76	-1.48
Fe_4N	M1	+0.87	+0.43
	M2	+0.26	+0.29
	N	-1.66	-1.39
	M1	+0.87	+0.45
Co_4N	M2	+0.28	+0.22
	N	-1.70	-1.36
Ni_4N	M1	+0.81	+0.40
	M2	+0.22	+0.16
	N	-1.46	-1.13
Cu_4N	M1	+0.67	+0.46
	M2	+0.28	+0.12
	N	-1.52	-1.16
Zn_4N	M1	+0.62	+0.65
	M2	+0.34	+0.04
	N	-1.62	-1.37

Table S2. Calculated characteristics of the electronic structures and electronic configurations and local magnetic moments in the atomic spheres in M_4N with Wigner-Seitz radius 1.40 Å for metal and 1.00 Å for N atoms. Some selected theoretical results are included for comparison. The Stoner parameter *I* for each element from Brooks [12] is included. *Only Sc_pv potential available, in which the semi-core Sc 3s and Sc 3p electrons are included in the potential. The electrons in Sc 4s, 4p states were not specified in the output of the calculations.

	I-M_4N		II-M_4N	
M /I	Electr.Chara. Width(eV).	Elec. Conf.	Electr.Chara. Width(eV).	Magn
Sc / 0.625	N 2s: –14.2 to –13.1/1.1 N 2p: –4.7 to –2.2 /2.5 M13d: -1.6 to 4.2 M23d: -1.6 to 6.5	N: $2s^{1.45}\ 2p^{2.92}$ Sc1: * $3d^{0.96}$ Sc2: * $3d^{1.05}$	N 2s: –14.5 to –13.4 /1.1 N 2p: –5.0 to –2.8/ 2.2 Sc1 3d: -1.4 to 6.4 Sc2 3d: -1.4 to 6.6	N: $2s^{1.46}\ 2p^{2.99}$ Sc1: * $3d^{1.03}$ Sc2: * $3d^{1.01}$
Ti / 0.615	N 2s: –15.5 to –14.0 /1.5 N 2p: –6.0 to –3.3 /2.7 Ti1 3d: -2.2 to 4.3 Ti2 3d -2.8 to 4.3	N: $2s^{1.46}\ 2p^{3.03}$ Ti1: $4s^{0.30}\ 4p^{0.31}\ 3d^{2.01}$ Ti2: $4s^{0.29}\ 4p^{0.37}\ 3d^{2.14}$	N 2s: –14.3 to –13.1/1.2 N 2p: –6.0 to –3.4 / 2.6 Ti1 3d: -2.3 to 4.4 Ti2 3d: -2.2 to 4.4	N: $2s^{1.46}\ 2p^{3.042}$ Ti1: $4s^{0.28}\ 4p^{0.38}\ 3d^{2.12}$ Ti2: $4s^{0.29}\ 4p^{0.34}\ 3d^{2.07}$
V / 0.640	N 2s:–17.1 to –14.9 /2.2 N 2p: –7.6 to -4.1 /3.5 V1 3d: -3.2 to 3.4 V2 3d -3.3 to 3.6	N: $2s^{1.46}\ 2p^{3.12}$ V1: $4s^{0.37}\ 4p^{0.39}\ 3d^{3.11}$ V2: $4s^{0.37}\ 4p^{0.53}\ 3d^{3.29}$	N 2s: –16.2 to –15.1 / 1.1 N 2p: –6.9 to -4.5 / 2.4 V1 3d: -3.2 to 4.0 V2 3d: -3.2 to 4.1	N: $2s^{1.46}\ 2p^{3.05}$ V1: $4s^{0.36}\ 4p^{0.48}\ 3d^{3.32}$ V2: $4s^{0.39}\ 4p^{0.52}\ 3d^{3.27}$
Cr / 0.690	N 2s: ↑-17.5 to -15.5 / 2.0 ↓-17.5 to -15.4 /2.1 N 2p ↑ -7.8 to -4.7 ↓ -8.0 to -4.7 Cr1: ↑ -3.3 to 2.3 ↓ +0.7 to 2.8 Cr2: ↑--3.4 to 3.2 ↓ -3.7 to 2.7	N: 0.08 μ_B ↑ $4s^{073}\ 4p^{1.61}$ ↓ $4s^{0.74}\ 4p^{1.53}$ Cr1: 2.20 μ_B ↑ $4s^{0.23}\ 4p^{0.24}\ 3d^{3.14}$ ↓ $4s^{0.19}\ 4p^{0.21}\ 3d^{1.05}$ Cr2: -1.15 μ_B ↑ $4s^{0.20}\ 4p^{0.31}\ 3d^{1.64}$ ↓ $4s^{0.21}\ 4p^{0.32}\ 3d^{2.78}$	N 2s: ↑-17.3 to -15.8 / 1.5 ↓-17.3 to -15.8 / 1.5 N 2p ↑ -7.8 to -4.7 ↓ -7.8 to -4.7 Cr1: ↑ -3.6 to 3.0 ↓ -3.6 to 3.0 Cr2: ↑--3.7 to 3.1 ↓ -3.7 to 3.1	N: NM $2s^{1.47}\ 2p^{3.12}$ Cr1: 0.00 $3s^{0.45}\ 3p^{0.65}\ 3d^{4.41}$ Cr2: NM $3s^{0.43}\ 3p^{0.57}\ 3d^{4.37}$
Mn / 0.775	N 2s: ↑-18.0 to -16.0 / 2.0 ↓-17.9 to -16.0 /2.1 N 2p ↑ -8.5 to -5.4 ↓ -8.6 to -5.4 Mn1: ↑ -4.3 to 1.2 ↓ +0.3 to 2.4 Mn2: ↑ -4.1 to 2.5 ↓ -4.1 to 2.3	N: 0.10 μ_B (0.07) [34] ↑ $4s^{074}\ 4p^{1.61}$ ↓ $4s^{0.73}\ 4p^{1.52}$ Mn1: 3.40 μ_B (3.23) [34] ↑ $4s^{0.25}\ 4p^{0.28}\ 3d^{4.22}$ ↓ $4s^{0.19}\ 4p^{0.22}\ 3d^{0.82}$ Mn2: -0.81 μ_B (-0.80) [34] ↑ $4s^{0.23}\ 4p^{0.33}\ 3d^{2.37}$ ↓ $4s^{0.22}\ 4p^{0.34}3d^{3.16}$	N 2s: ↑-17.3 to -15.8 / 1.5 ↓-17.3 to -16.0 /1.3 N 2p ↑ -8.2 to -5.3 ↓ -8.0 to -5.5 Mn1: ↑ -4.2 to 1.0 ↓ -2.0 to 2.3 Mn2: ↑ -1.1 to 2.5 ↓ -4.3 to 1.7	N: -0.06 μ_B ↑ $4s^{072}\ 4p^{1.53}$ ↓ $4s^{0.73}\ 4p^{1.58}$ Mn1: 2.49 μ_B ↑ $4s^{0.24}\ 4p^{0.33}\ 3d^{3.93}$ ↓ $4s^{0.21}\ 4p^{0.31}2d^{1.48}$ Mn2: -2.27 μ_B ↑ $4s^{0.20}\ 4p^{0.25}\ 3d^{1.57}$ ↓ $4s^{0.23}\ 4p^{0.30}3d^{3.77}$
Fe / 0.900	N 2s: ↑-17.5 to -15.8 / 1.7	N: 0.03 μ_B (0.01) [22] ↑ $4s^{0.72}\ 4p^{1.52}$	N 2s: ↑-17.4 to -16.1 / 1.3	N: -0.03 μ_B (-0.18) [25] ↑ $4s^{073}\ 4p^{1.52}$

	↓-17.4 to -15.7 / 1.7 N 2p ↑ -8.4 to -5.4 ↓ -7.8 to -4.8 Fe1: ↑ -4.6 to -0.7 ↓ -2.9 to 2.2 Fe2: ↑ -4.9 to 0.2 ↓ -2.9 to 2.4	↓ $4s^{0.73}\,4p^{1.49}$ Fe1: 2.97 μ_B (2.98) [22] ↑ $4s^{0.22}\,4p^{0.22}\,3d^{4.62}$ ↓ $4s^{0.23}\,4p^{0.25}\,3d^{1.61}$ Fe2: 2.35 μ_B(2.23) [22] ↑ $4s^{0.20}\,4p^{0.31}\,3d^{4.41}$ ↓ $4s^{0.21}\,4p^{0.32}3d^{2..05}$	↓-17.3 to -16.0 / 1.3 N 2p ↑ -8.3 to -5.6 ↓ -7.9 to -5.1 Fe1: ↑ -4.6 to 0.6 ↓ -3.4 to 2.6 Fe2: ↑ -4.6 to 0.3 ↓ -3.4 to 2.6	↓ $4s^{0.73}\,4p^{1.55}$ Fe1: 1.96 μ_B (2.03) [25] ↑ $4s^{0.24}\,4p^{0.33}\,3d^{4.25}$ ↓ $4s^{0.23}\,4p^{0.34}\,3d^{2.27}$ Fe2: 2.54 μ_B(2.50) [25] ↑ $4s^{0.23}\,4p^{0.27}\,3d^{4.46}$ ↓ $4s^{0.23}\,4p^{0.30}3d^{1.19}$
Co / 0.940	N 2s: ↑-17.9 to -16.2 / 1.7 ↓-17.7 to -16.0 / 1.7 N 2p ↑ -8.7 to -5.6 ↓ -8.2 to -5.2 Co1: ↑ -4.5 to -0.8 ↓ -3.5 to 1.2 Co2: ↑ -4.7 to -0.1 ↓ -3.5 to 1.4	N: 0.10 μ_B (0.069) [35] ↑ $4s^{072}\,4p^{1.60}$ ↓ $4s^{0.72}\,4p^{1.50}$ Co1: 1.93 μ_B(1.967) [35] ↑ $4s^{0.22}\,4p^{0.22}\,3d^{4.65}$ ↓ $4s^{0.23}\,4p^{0.25}\,3d^{2.69}$ Co2: 1.49 μ_B (1.486) [35] ↑ $4s^{0.25}\,4p^{0.36}\,3d^{4.52}$ ↓ $4s^{0.25}\,4p^{0.37}\,3d^{3.03}$	N 2s: ↑-17.7 to -16.4 / 1.3 ↓-17.5 to -16.1 / 1.4 N 2p ↑ -8.6 to -5.7 ↓ -8.2 to -5.3 Co1: ↑ -4.7 to 0.2 ↓ -3.8 to 1.6 Co2: ↑ -4.7 to 0.2 ↓ -3.8 to 1.7	N: 0.08 μ_B ↑ $4s^{073}\,4p^{1.58}$ ↓ $4s^{0.72}\,4p^{1.50}$ Co1: 1.38 μ_B ↑ $4s^{0.25}\,4p^{0.74}\,3d^{4.47}$ ↓ $4s^{0.26}\,4p^{0.75}\,3d^{3.09}$ Co2: 1.71 μ_B ↑ $4s^{0.24}\,4p^{0.29}\,3d^{4.58}$ ↓ $4s^{0.25}\,4p^{0.31}3d^{2.87}$
Ni / 0.980	N 2s: ↑-17.4 to -15.9 / 1.5 ↓-17.3 to -15.8 / 1.5 N 2p ↑ -8.1 to -5.3 ↓ -8.0 to -5.3 Ni1: ↑ -3.6 to 0.0 ↓ -3.3 to 0.7 Ni2: ↑ -4.0 to 0.5 ↓ -3.4 to 0.6	N: 0.04 μ_B ↑ $4s^{072}\,4p^{1.55}$ ↓ $4s^{0.72}\,4p^{151}$ Ni1: 0.69 μ_B ↑ $4s^{0.23}\,4p^{0.21}\,3d^{4.58}$ ↓ $4s^{0.23}\,4p^{0.21}\,3d^{3.89}$ Ni2: 0.29 μ_B ↑ $4s^{0.26}\,4p^{0.36}\,3d^{4.44}$ ↓ $4s^{0.26}\,4p^{0.36}\,3d^{4.15}$	N 2s: –17.4 to –16.2 /1.2 N 2p: –8.2 to -5.3 / 2.9 Ni1 3d: -4.6 to 0.9 Ni2 3d: -4.2 to 1.0	N: NM $2s^{1.46}\,2p^{3.02}$ Ni1: NM $4s^{0.51}\,4p^{0.71}\,3d^{8.59}$ Ni2: NM $4s^{0.49}\,4p^{0.58}\,3d^{8.56}$
Cu / 0.670	N 2s:–17.3 to –16.0 /1.2 N 2p: –8.4 to -5.5 / 2.9 Cu1 3d: -4.8 to -0.9 Cu2 3d: -4.7 to -1.0	N: $2s^{1.45}\,2p^{2.98}$ Cu1: $4s^{0.52}\,4p^{0.35}\,3d^{9.41}$ Cu2: $4s^{0.52}\,4p^{0.63}\,3d^{9.42}$	N 2s:–17.0 to –15.8 /1.2 N 2p: –8.3 to -5.4 / 2.9 Cu1 3d: -5.4 to -1.2 Cu2 3d: -5.1 to -1.1	N: $2s^{1.47}\,2p^{2.92}$ Cu1: $4s^{0.52}\,4p^{0.64}\,3d^{9.41}$ Cu2: $4s^{0.50}\,4p^{0.48}\,3d^{9.41}$