Supplemental material for publication

ab initio design of a new family of 2D materials: transition metal carbon nitrogen compounds (MCNs)

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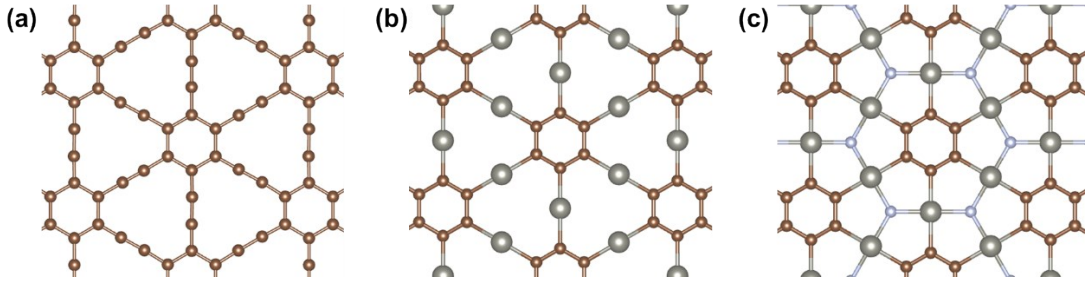

Figure S1 The proposed process for the formation of MCNs: from (a) γ-graphyne to (b) γ-graphyne-like MC to (c) the final MCN with N atoms filled in the original vacancies in γ-graphyne.

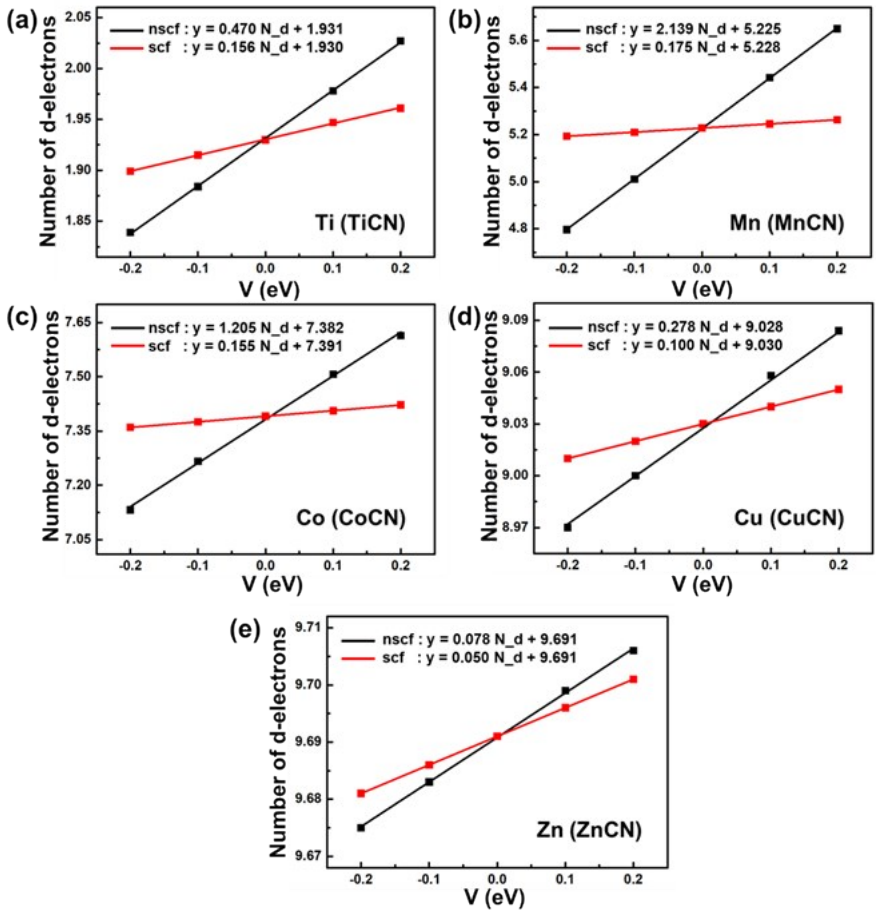

Figure S2 The number of d-electrons on transition metals in MCNs (M= Ti, Mn, Co, Cu and Zn) as a function of the additional potential V.

The U parameters for MCNs are found from:

$$U = \chi^{-1} - \chi_0^{-1} \approx \left(\frac{\partial N_I^{scf}}{\partial V_I}\right)^{-1} - \left(\frac{\partial N_I^{nscf}}{\partial V_I}\right)^{-1}$$

where χ is the selfconsistent response function, χ_0 is non-selfconsistent response function, N is the number of *d*-electrons and V is the additional potential. The calculated U parameters from linear response approach are 4.28, 5.25, 5.62, 6.40 and 7.18 eV for the transition metals in TiCN, MnCN, CoCn, CuCN and ZnCN, respectively.

Table S1 Lattice parameters of optimized MCN monolayers and formation energies of MCNs per atom (E_f/eV).

MCN	a=b/Å	L_{M-N}/Å	L_{M-C}/Å	A_{M-N-M}/°	A_{C-C-C}/°	E_f/eV
ScCN	6.52	2.03	2.37	104	115/130	0.02
TiCN	5.98	1.98	2.16	99	120	-0.15
CrCN	6.12	1.87	1.95	110	120	0.46
MnCN	5.85	1.83	1.93	106	120	0.48
FeCN	10.43	1.81	1.89	125/108	119/121	0.67
VCN	10.03	1.90	2.06	96/106	120	0.16
NiCN	10.82	1.82	1.91	125/110	115/124	0.91
CoCN	10.58	1.79	1.91	130/100	116/124	0.79
CuCN	11.48	1.91	1,95	120	118/121	1.23
ZnCN	11.77	1.96	2.01	120	120	0.92

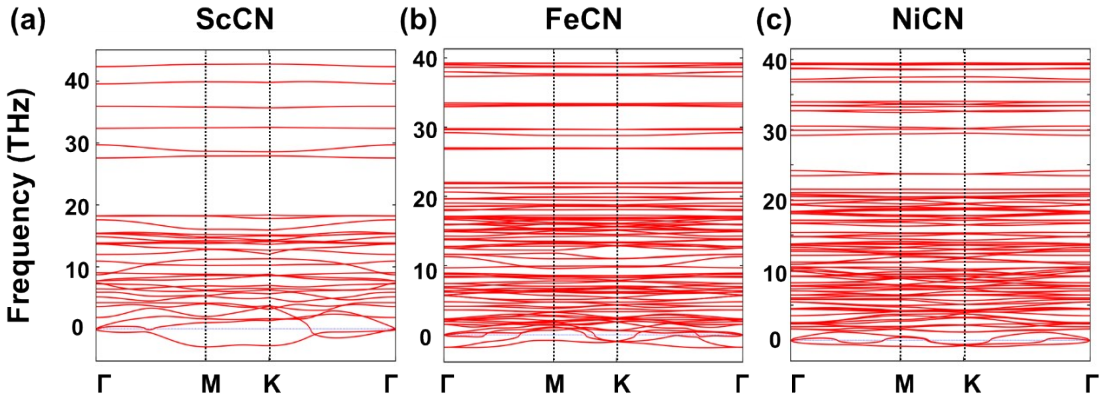

Figure S3 Phonon spectra of (a) ScCN, (b) FeCN, and (c) NiCN monolayers.

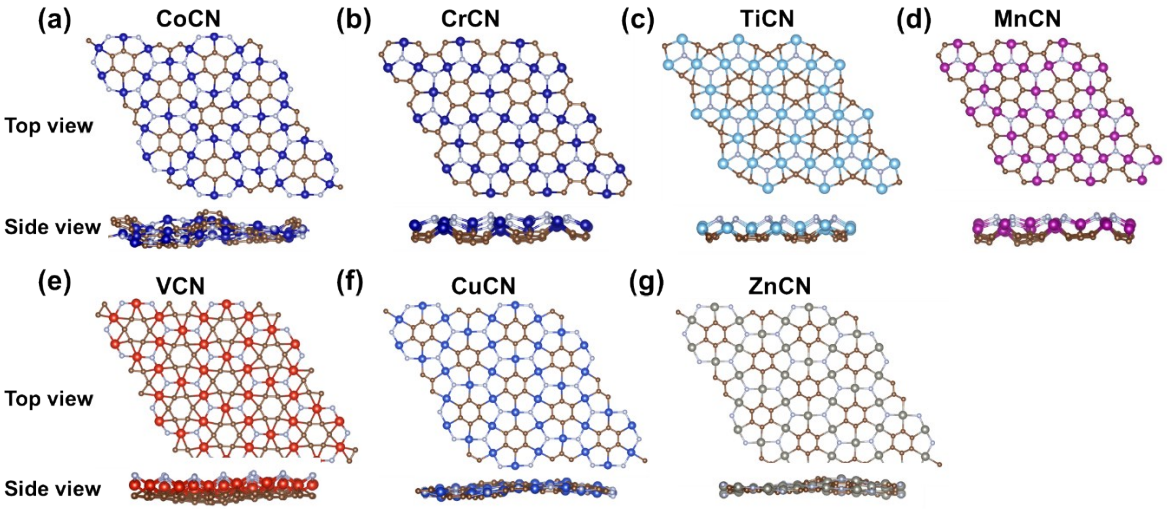

Figure S4 Final structures of MCNs after AIMD simulations for 8 ps at 300K.

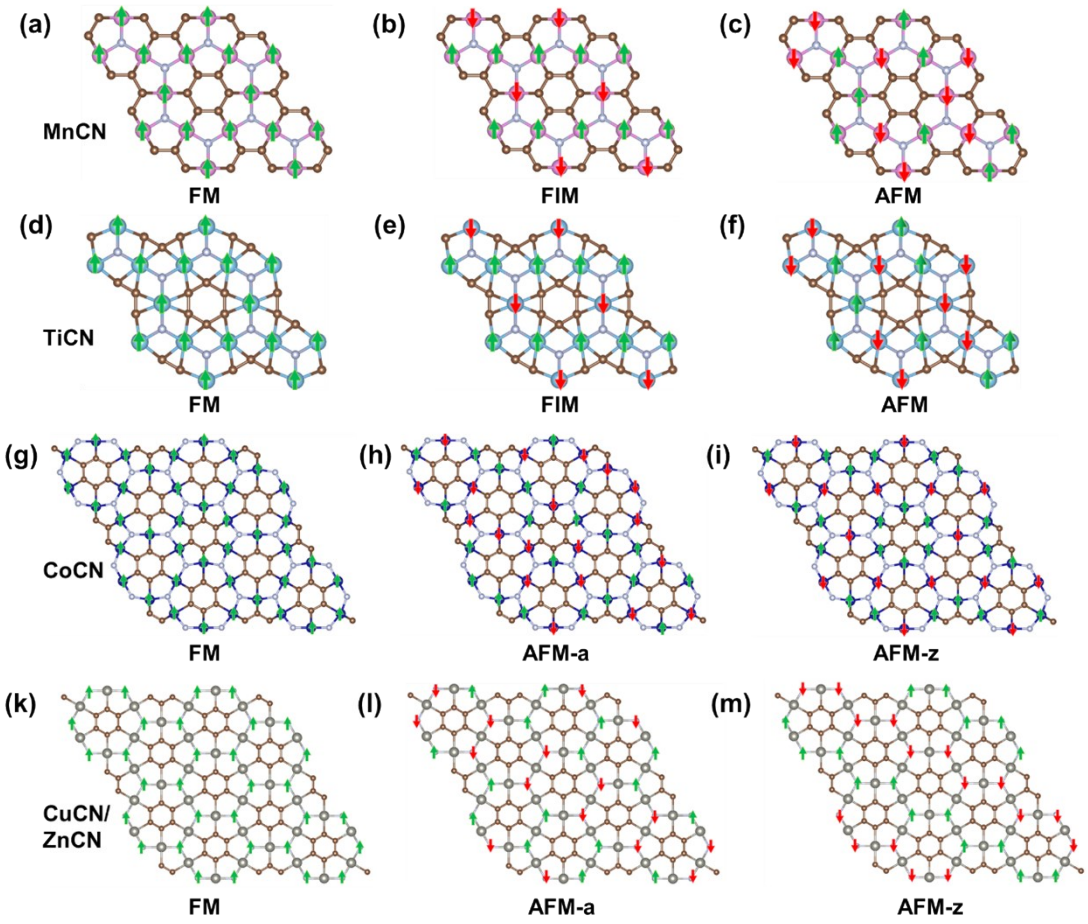

Figure S5 Schematic illustrations of ferromagnetic (FM), ferrimagnetic (FIM) and antiferromagnetic couplings in MnCN (a)-(c), TiCN (d)-(f) and CoCN (g)-(i). Ferromagnetic (FM), antiferromagnetic along arm orientation (AFM-a) and antiferromagnetic along zigzag orientation (AFM-z) couplings in CuCN and ZnCN (k)-(m).

Table S2 GGA collinear magnetic energies in MCN monolayers per s.f. ($M_3C_6N_2$)

MCN	E_{NM} /eV	E_{FM} /eV	E_{FIM} /eV	E_{AFM} /eV	E_{AFM}-E_{FM} /meV	E_{AFM-a} /eV	E_{AFM-z} /eV	E_{AFM-a}-E_{FM} /meV	E_{AFM-z}-E_{FM} /meV	Ground state
TiCN	-96.13	NM	NM	NM	-	-	-	-	-	NM
MnCN	-92.17	-92.54	-92.63	-92.77	-237.47	-	-	-	-	AFM
CoCN	-83.35	-83.37	-83.37	FIM	-	-	-	-	-	FM/FIM
CuCN	-70.13	-70.31	-	-	-	-70.40	-70.36	-88.60	-48.66	AFM-a
ZnCN	-63.89	-63.98	-	-	-	-63.95	-63.96	24.81	22.40	FM

Table S3 GGA+U atomic magnetic moments and total magnetic moments (/s.f.) in MCN monolayers

MCN	M_C/μ_B	M_N/μ_B	M_M/μ_B	M_{tot}/μ_B
MnCN	±0.21/±0.17/±0.12	±0.09	±3.33/±3.31/±3.14	0
CoCN	-0.13/-0.12/-0.09	-0.31	1.63/1.28	9.76
CuCN	0/±0.01/±0.02	±0.60	0/±0.05	0
ZnCN	0.01	0.34	0.01	2.21

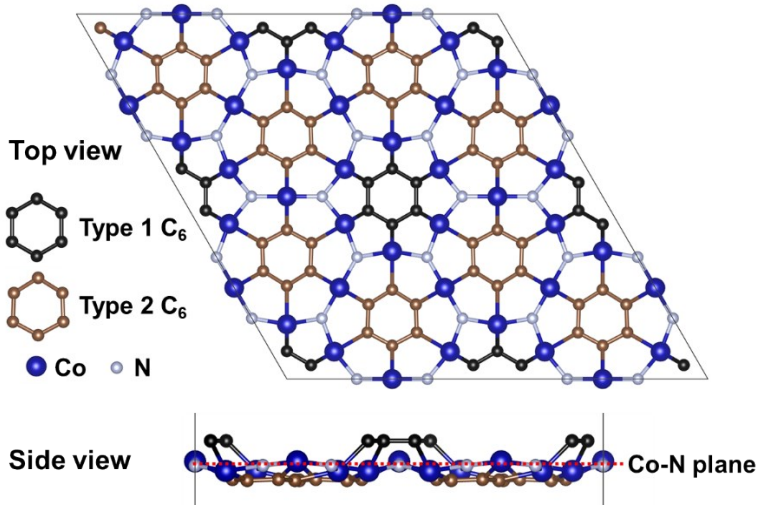

Figure S6 Top view and side view of 2 × 2 × 1 CoCN supercell. Type 1 carbon rings marked in black have a large vertical distance from the Co-N plane and type 2 carbon rings marked in brown are almost in the same horizontal position with the Co-N plane.

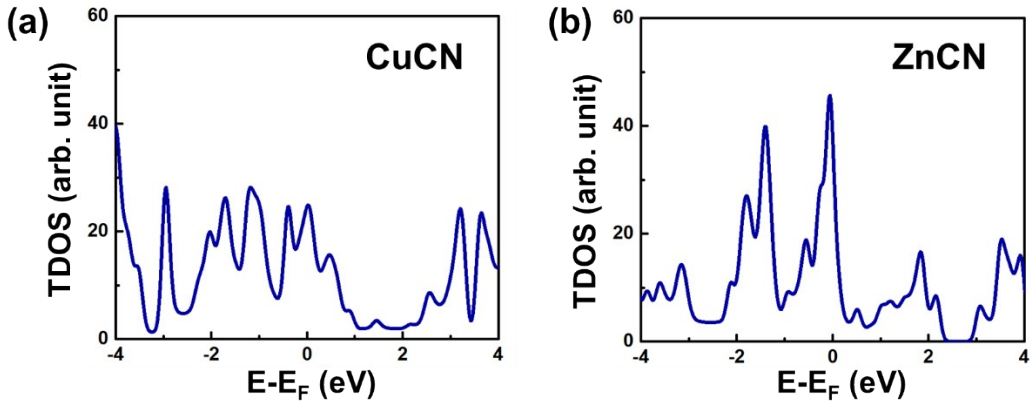

Figure S7 TDOSs of (a) CuCN and (b) ZnCN.

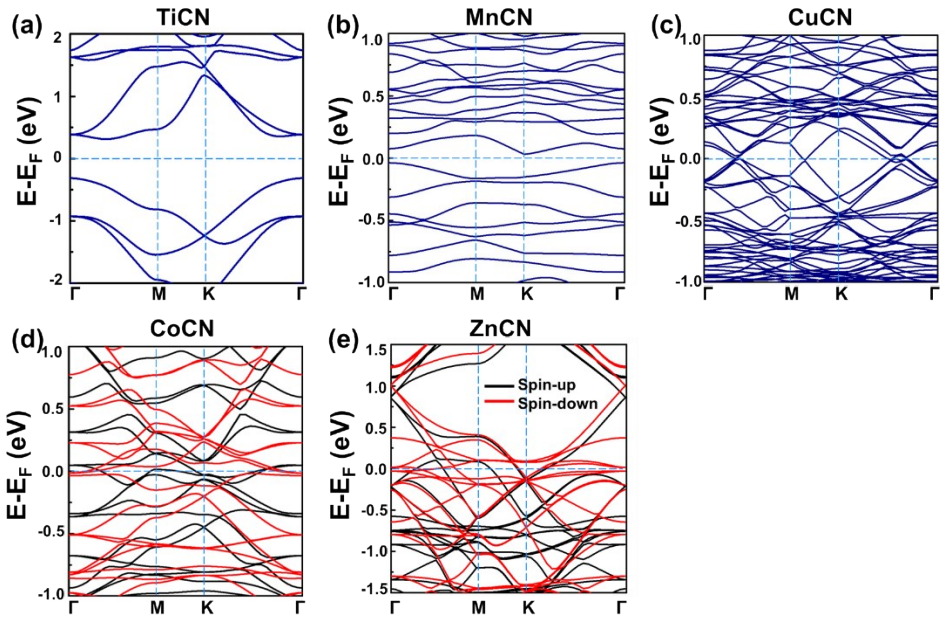

Figure S8 The GGA electronic band structures of (a) TiCN, (b) MnCN, (c) CuCN, (d) CoCN and (e) ZnCN monolayers near the Fermi level. The Fermi level was set to 0 eV. Γ (0, 0, 0), M (1/2, 0, 0), and K (1/3, 1/3, 0) are highly symmetric points in reciprocal space. (Red – spin down, black – spin up)