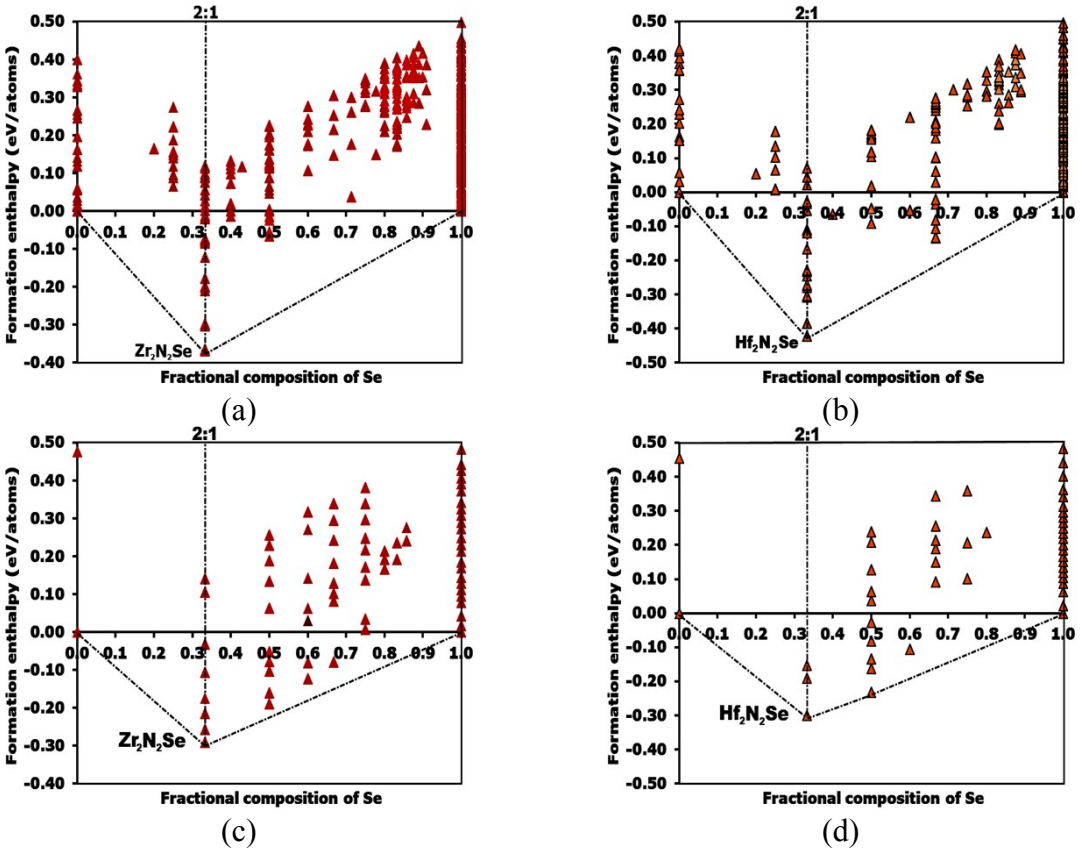
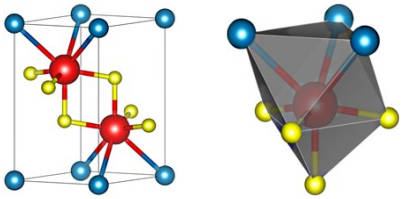
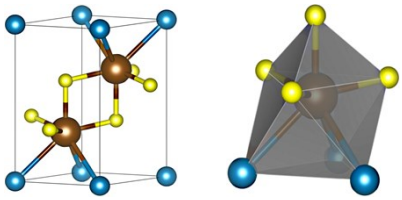
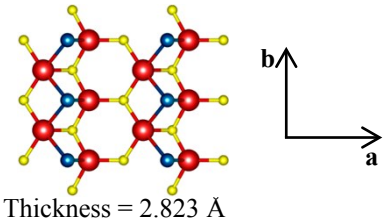
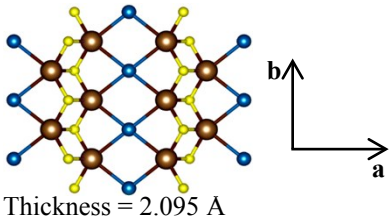
Supplementary file



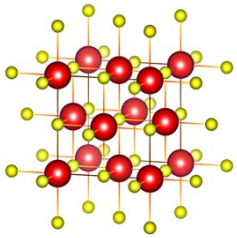
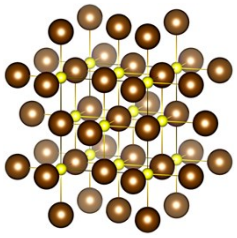
S1 Convex hull plot for (a) $P\bar{3}m1$-Zr_2N_2Se and (b) $P\bar{3}m1$-Hf_2N_2Se phases, (c) P1m1-Zr_2N_2Se and (d) C2/m-Hf_2N_2Se monolayers.

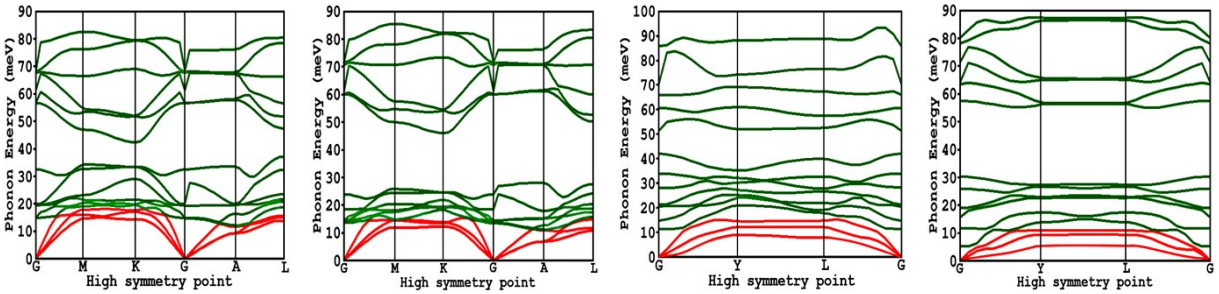
T1 Calculated structural data for energetically stable (a) α-Zr_2N_2Se and (b) α-Hf_2N_2Se phases. Top view of (c) P1m1-Zr_2N_2Se and (d) C2/m-Hf_2N_2Se monolayers. Red, brown, blue and yellow spheres indicate Zr, Hf, Se and N atoms, respectively.

Phase	Unit cell	Lattice constants / Å			Atomic Wyckoff positions
		a	*b*	*c*	
Zr_2N_2Se SG 164		3.659	3.659	6.687	Zr (0.33333, 0.66667, 0.30472) N (0.33333, 0.66667, -0.36850) Se (0.00000, 0.00000, 0.00000)
Hf_2N_2Se SG 164		3.623	3.623	6.655	Hf (0.33333, 0.66667, -0.30511) N (0.33333, 0.66667, 0.36841) Se (0.00000, 0.00000, 0.00000)

Zr_2N_2Se SG 6	 Thickness = 2.823 Å Dipole moment = 0.166 eÅ	5. 812	3.535	19.830	Zr1 (0.21562, 0.50000, -0.49546) Zr2 (-0.32906, 0.00000, 0.47402) N1 (-0.49246, 0.50000, 0.44697) N2 (0.01983, 0.00000, 0.48574) Se (0.45792, 0.00000, -0.41127)
Hf_2N_2Se SG 12	 Thickness = 2.095 Å Dipole moment = 0.006 eÅ	13.024	3.490	19.817	Hf (0.33746, 0.00000, 0.47705) N (0.23670, 0.00000, -0.44716) Se (0.00000, 0.00000, 0.50000)

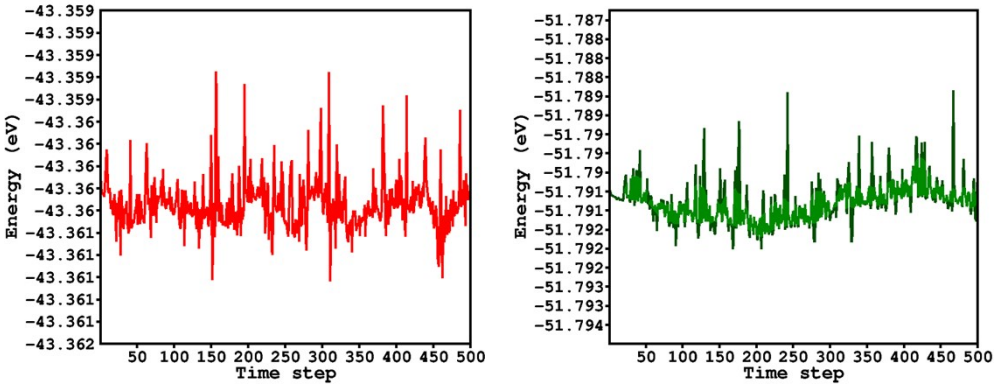
T2 Crystallographic data of experimentally proven Zr_3N_3 and Hf_4N_4 phases. Red, brown and yellow spheres denote the Zr, Hf and N atoms, respectively. Refs [1-2] indicate their experimentally verified crystal class and lattice constants.

Phase	Unit cell structure	Lattice parameters / Å	Wyckoff position
$Fm\bar{3}m$-Zr_3N_3 (SG 225)		a = 4.59537 a = 4.56750 [1]	Zr (0.00000, 0.00000, 0.00000) N (0.50000, 0.50000, 0.50000)
$Fm\bar{3}m$-Hf_4N_4 (SG 225)		a = 4.53684 a = 4.53000 [2]	Hf (0.50000, 0.50000, 0.50000) N (0.00000, 0.00000, 0.00000)



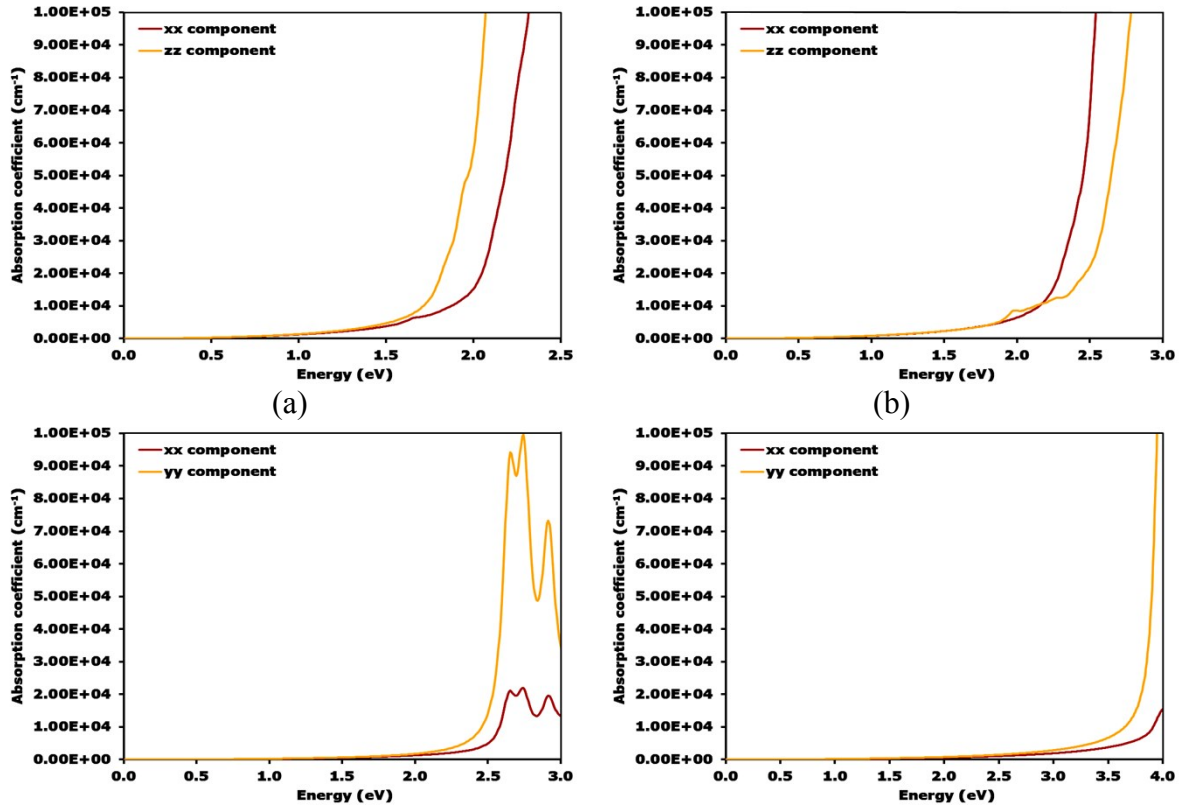
(a) (b) (c) (d)

S2 Phonon dispersion curve for (a) $P\bar{3}m1$-Zr_2N_2Se, (b) $P\bar{3}m1$-Hf_2N_2Se, (c) P1m1-Zr_2N_2Se and (d) C2/m-Hf_2N_2Se.



(a) (b)

S3 Energy profiles for (a) P1m1-Zr_2N_2Se and (b) C2/m-Hf_2N_2Se during AIMD simulations at 300 K. Instability of structures is not observed.



(a) (b)

(c) (d)

S4 Zoom-in features of absorption coefficient in the visible and infrared range for (a) $P\bar{3}m1$-Zr_2N_2Se, (b) $P\bar{3}m1$-Hf_2N_2Se, (c) P1m1-Zr_2N_2Se and (d) C2/m-Hf_2N_2Se.

References

1) M.M.S. Sirajuddeena, I.B.S. Banu (2014). AIP Advances. 4(5), 057121.
2) A. Srivastava and B.D. Diwan (2014). Can. J. Phys. 92, 1058-1061.