SUPPLEMENTARY MATERIAL:

Moiré-of-Moiré phases formed in twisted graphene/hexagonal boron nitride heterostructures under high pressure

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A. The Wyckoff positions and full atomic coordinates (in fractional) of *m*-BC_2N-21.78°

Table S1. The Wyckoff positions and full atomic coordinates (in fractional) of *m*-BC_2N-21.78° under zero pressure

Crystal	Groups	Wyckoff positions	Atomic coordinates
m-BC_2N (21.78°)	*P*3 (143)	C1 0.30688 0.43764 0.67442 C2 0.39830 0.12162 0.64586 C3 0.10662 0.46434 0.79021 C4 0.26816 0.20902 0.82662 C5 0.00000 0.00000 0.91236 C6 0.66667 0.33333 0.68826 N1 0.87561 0.59836 0.12926 N2 0.12858 0.44146 0.15338 N3 0.00000 0.00000 0.26108 B1 0.10735 0.64607 0.28456 B2 0.94060 0.21604 0.31233 B3 0.66667 0.33333 0.05198	C1 0.306880 0.437640 0.674420 C2 0.398300 0.121620 0.645860 C3 0.106620 0.464340 0.790210 C4 0.268160 0.209020 0.826620 C5 0.562360 0.869240 0.674420 C6 0.878380 0.276680 0.645860 C7 0.535660 0.642280 0.790210 C8 0.790980 0.059140 0.826620 C9 0.130760 0.693120 0.674420 C10 0.723320 0.601700 0.645860 C11 0.357720 0.893380 0.790210 C12 0.940860 0.731840 0.826620 C13 0.000000 0.000000 0.912360 C14 0.666670 0.333330 0.688260 N1 0.875610 0.598360 0.129260 N2 0.128580 0.441460 0.153380 N3 0.401640 0.277250 0.129260 N4 0.558540 0.687120 0.153380 N5 0.722750 0.124390 0.129260 N6 0.312880 0.871420 0.153380 N7 0.000000 0.000000 0.261080 B1 0.107350 0.646070 0.284560 B2 0.940600 0.216040 0.312330 B3 0.353930 0.461280 0.284560 B4 0.783960 0.724560 0.312330 B5 0.538720 0.892650 0.284560 B6 0.275440 0.059400 0.312330 B7 0.666670 0.333330 0.051980

B. Verification of elastic constants C_{ij} calculations

Table S2. Comparison of DFT-calculated elastic constants C_{ij} (in GPa) for diamond and *c*BN with reported experimental and DFT results.

Crystal	C_{11}	C_{22}	C_{33}	C_{44}	C_{55}	C_{66}	C_{12}	C_{13}	C_{23}	Source
Diamond	1054.4	-	-	563.1	-	-	120.0	-	-	This
	1076	-	-	577	-	-	125	-	-	Expt.[1]
	1099	-	-	590	-	-	149	-	-	Cal.[2]
*c*BN	853.4	-	-	497.4	-	-	215.8	-	-	This
	820	-	-	480	-	-	190	-	-	Expt.[3]
	823			479			185			Cal.[4]

C. Verification of electronic band structure calculations

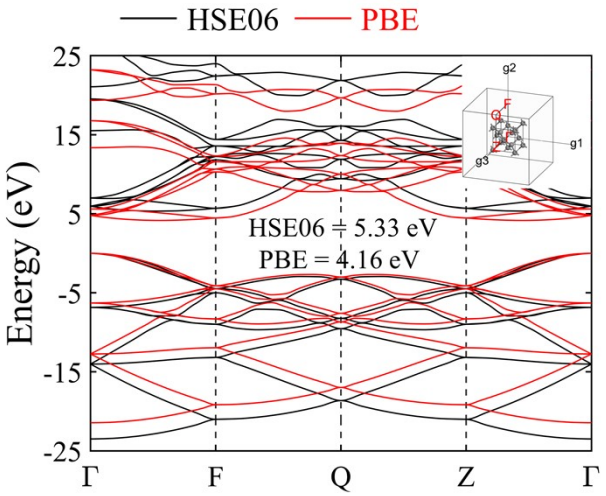

Figure S1. Electronic band structures of diamond, where the black lines are obtained from HSE06 functional, the red lines are obtained from PBE functional. It can be observed that HSE06 increases only the values of band gap without causing any qualitative changes in the band structure.

D. Verification of size-dependent effects on the structural stability of *m*-BC_2N (21.78°)

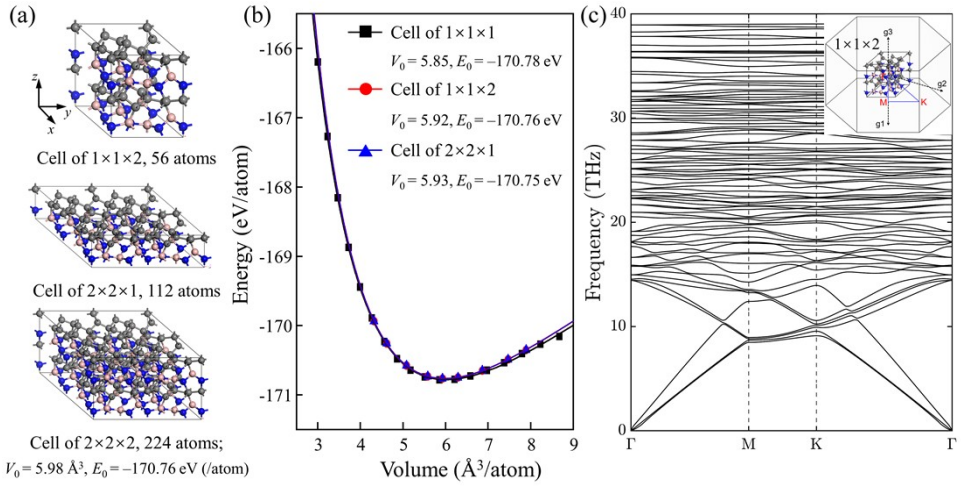

Figure S2. The validation of size-dependent effects on the structural stability of *m*-BC_2N (21.78°). (a) Upon full relaxation at zero pressure, the atomic structure of the 1×1×2, 2×2×1, and 2×2×2 supercell units maintains a perfect sp^3 configuration. (b) The total energy per atom of the 1×1×2 and 2×2×1 supercell as a function of atomic density, exhibits a high degree of consistency compared to the 1×1×1 unit. (c) Phonon dispersion relation of 1×1×2 supercell reveals no occurrence of imaginary frequencies. These findings collectively support the stability of *m*-BC_2N (21.78°) regardless of its unit cell dimensions.

F. Band structure of *m*-BC_2N (9.43°)

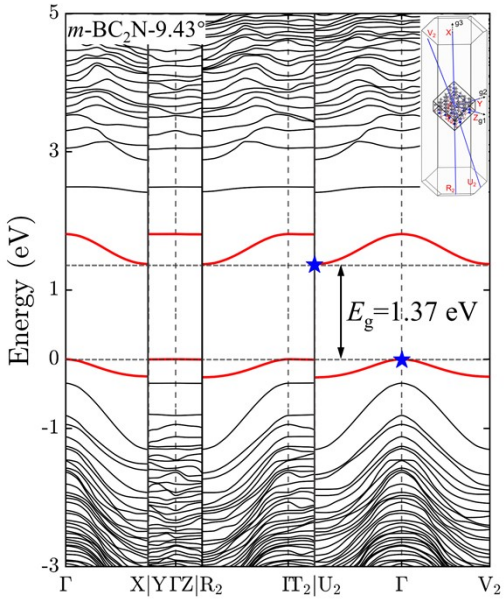

Figure S3. The electronic band structures along the high symmetry direction in the Brillouin zone of *m*-BC_2N (9.43°). Additionally, the band structure shows a characteristic of p-doped semiconductors. This finding aligns with the minute peak observed at the Fermi point in the DOS curve (Figure 9a).

References

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