Supporting Information for

Creating a Nanospace under an h-BN Cover for Adlayer Growth on Nickel(111)

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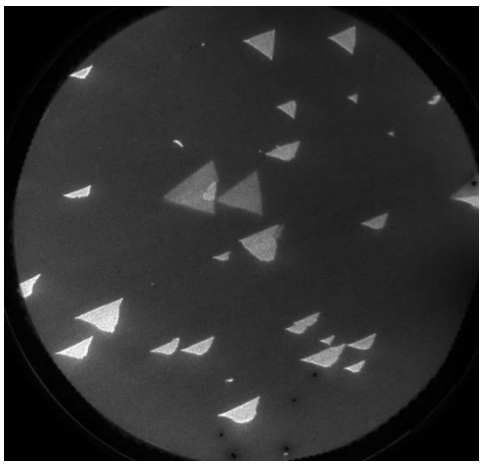

Video S1 LEEM video of h-BN overlayer growth on Ni(111). Growth conditions: temperature = 625 ^{o}C, P(BH_3NH_3) = 1.8 × 10^{-8} Torr. Imaging conditions: STV = 5.0 V, FoV = 50 μm. The whole growth process lasted for 11 min.

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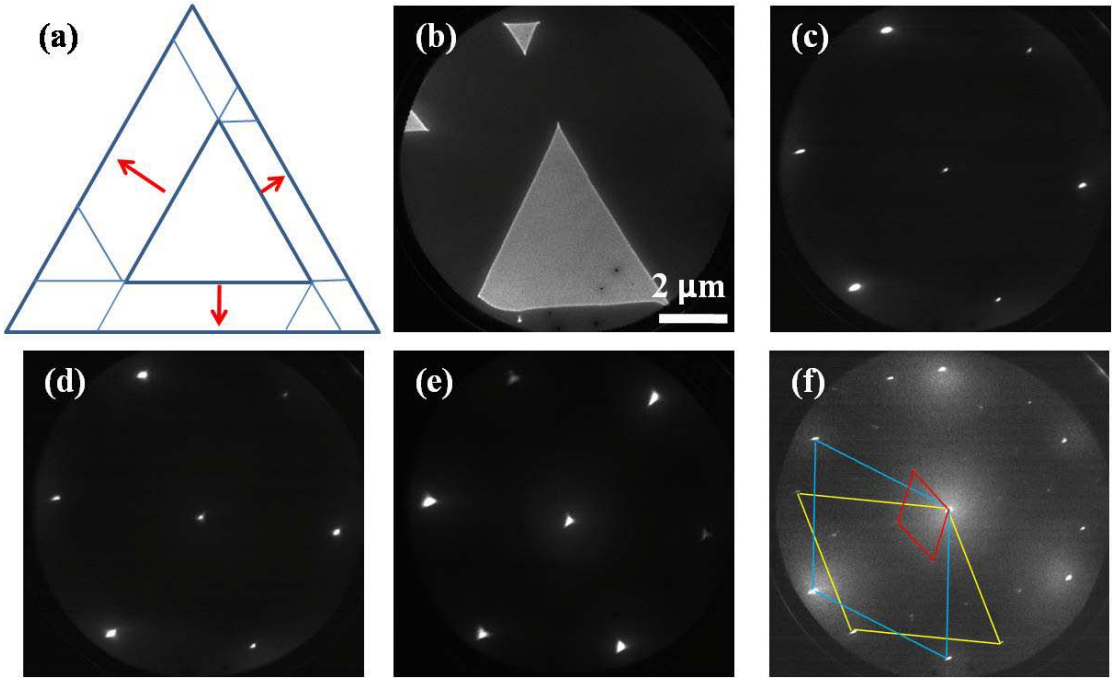


Figure S1 (a) Schematics showing the growth of a h-BN triangle island along three directions which are perpendicular to the three edges. The increase of one edge length includes the increases in all three directions. (b) LEEM image showing two kinds of type-I h-BN nanoislands. (c) LEED pattern of Ni(111). (d) LEED pattern of the smaller h-BN nanoisland in (b). (e) LEED pattern of the larger h-BN island in (b). (f) LEED pattern of another type-II h-BN nanoisland. Rhombuses with different colors: yellow, Ni; blue, type-II h-BN; red, moire superstructure. The electron energies to form LEED patterns in (c-f) are all 50 eV.

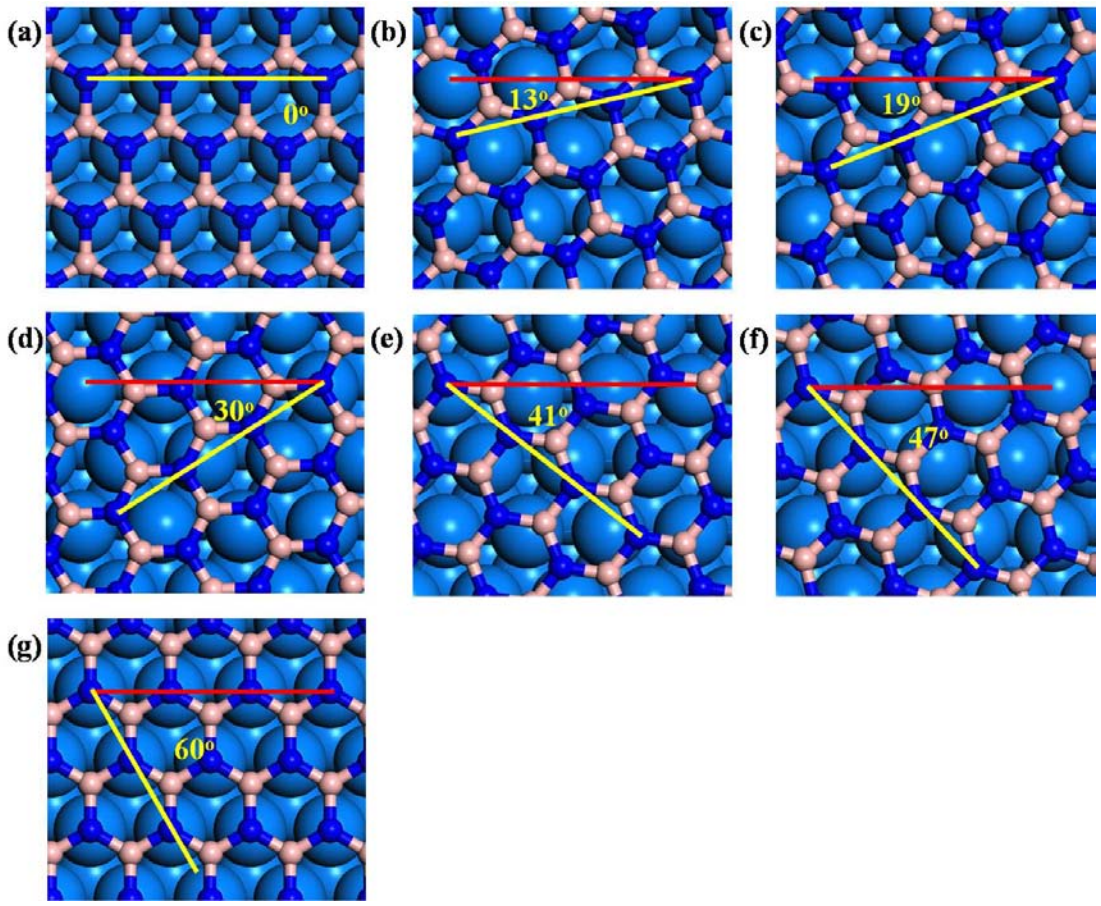
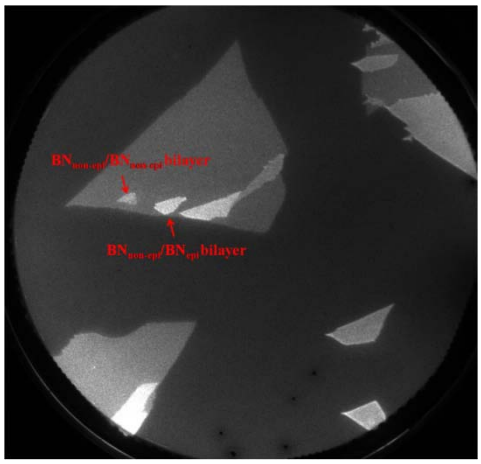


Figure S2. (a-g) Top view of optimized atomic models for the supercells with different rotation angles. a) 0 °, b) 13 °, c) 19 °, d) 30 °, e) 41 °, f) 47 °, g) 60 °. Ni: light blue balls; N: dark blue balls; B: pink balls. Red lines: orientation of the Ni(111) lattice; Yellow line: orientation of h-BN lattice.

Rotation angle (degree)	Lattice constant (Å)	Lattice mismatch (%)
0	4.98	0.33
13	10.86	0.34
19	6.59	0.32
30	8.98	0.38
41	6.59	0.32
47	10.86	0.34
60	4.98	0.33

Table S1 Lattice constant and lattice mismatch of the calculated models for different rotation angles. The lattice constant of h-BN is 2.50 Å.



Video S2 LEEM video of the formation of h-BN_{nonepi}/h-BN_{epi} and h-BN_{nonepi}/h-BN_{nonepi} bilayer structures *via* the nucleation and growth of h-BN_{epi} or h-BN_{nonepi} islands underneath an existing h-BN_{nonepi} island. Growth conditions: temperature = 640 ^{o}C, P(BH_3NH_3) = 3.5×10^{-8} Torr. Imaging conditions: STV = 5.0 V, FoV = 50 μm. The whole growth process lasted for 9 min.

http://fruit.dicp.ac.cn/images/videos2.avi

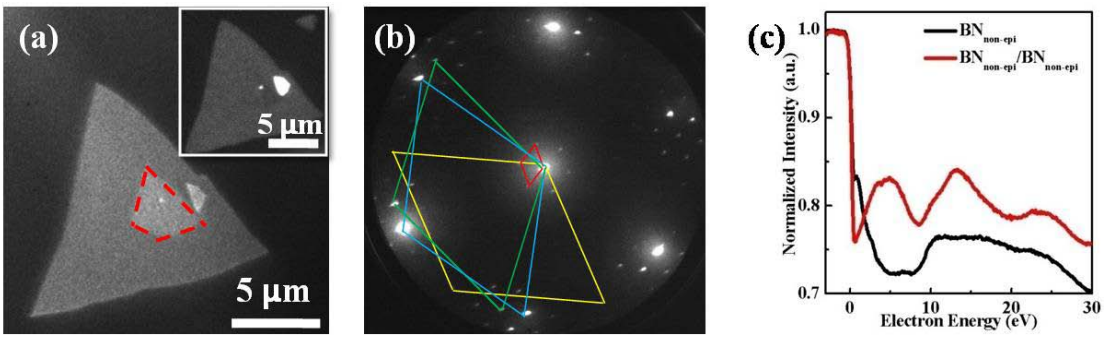
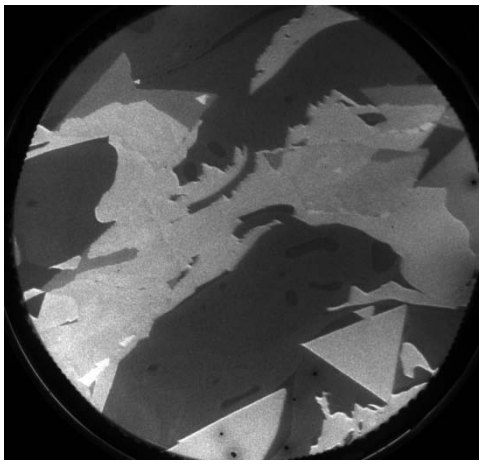


Figure S3 (a) LEEM image of a h-BN_{nonepi}/h-BN_{nonepi} bilayer highlighted by dashed red lines. Inset is the corresponding PEEM image. (b) LEED pattern (50 eV) of the bilayer structure. Rhombuses with different colors: yellow, Ni; blue, the first h-BN_{nonepi} layer; green, the second h-BN_{nonepi} layer; red, moire superstructures. (c) *I-V* curves of h-BN_{nonepi} vs h-BN_{nonepi}/h-BN_{nonepi} bilayer.



Video S3 LEEM video of a h-BN/Ni(111) surface exposed to C_2H_4. Growth conditions: temperature: 540 ^{o}C, P(C_2H_4) = 1.2 ×10^{-6} Torr. Imaging conditions: STV = 4.5V, FoV = 50 μm. The whole growth process lasted for about 1h.

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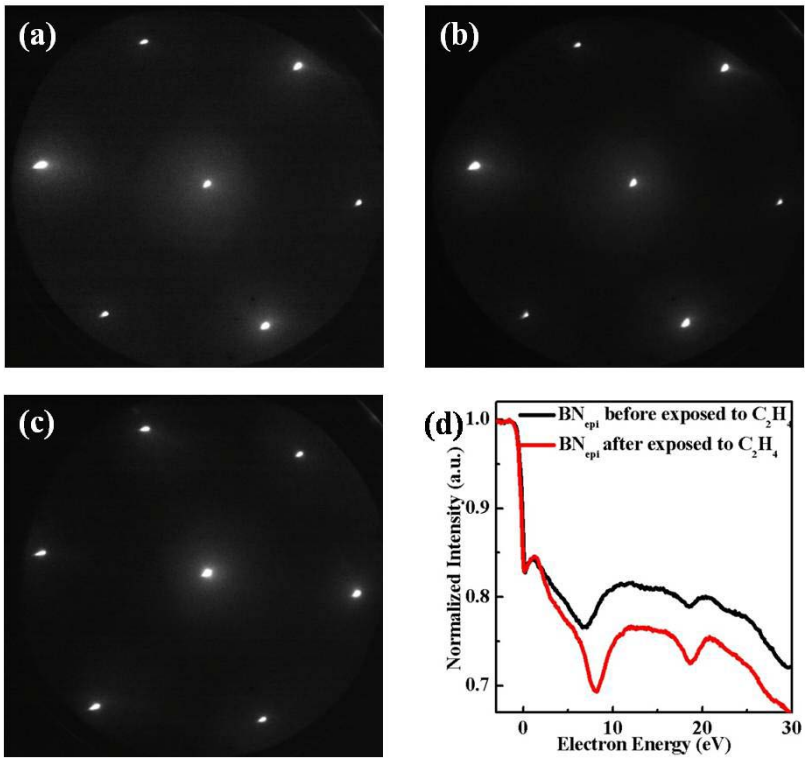
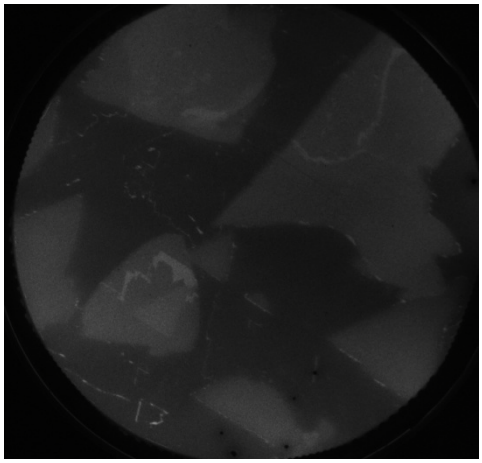


Figure S4 (a, b) LEED patterns of a h-BN_{epi} island before and after the exposure of C_2H_4. (c) LEED pattern of a graphene island grown on Ni(111). The electron energies to form LEED patterns in (a-c) are all 50 eV. (d) Corresponding *I-V* curves for (a) and (b), respectively.



Video S4 LEEM video of a h-BN/graphene/Ni(111) surface when heated from room temperature to about 620 ^{o}C in UHV. Imaging conditions: STV = 7.2 V, FoV = 50 **μ**m. The whole dissolution process lasted for 7 min.

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