

Supporting Information

Templated Growth and Intercalation Pathways of Nickel Nanoclusters on Graphene/Ir(111)

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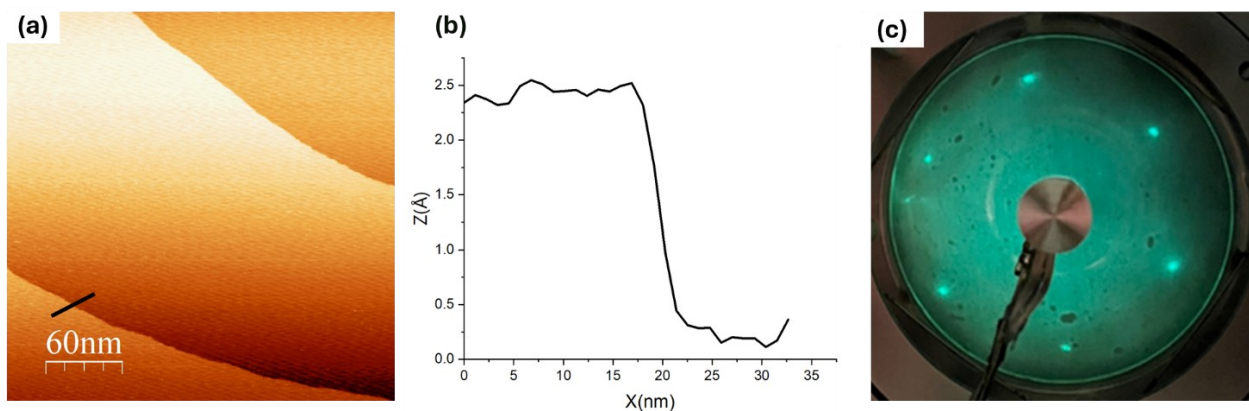


Figure S1. (a) A large-area STM image ($300 \times 300 \text{ nm}^2$) of clean Ir(111), suitable for subsequent graphene growth (scanning parameter: 2 V, 200 pA). (b) Line profile along the black line marked in (a), showing the height of a single step edge. (c) The LEED pattern of the Ir(111) crystal in (a), recorded at 77 eV, displaying sharp diffraction spots arranged in a hexagonal symmetry, characteristic of the Ir(111) Surface.

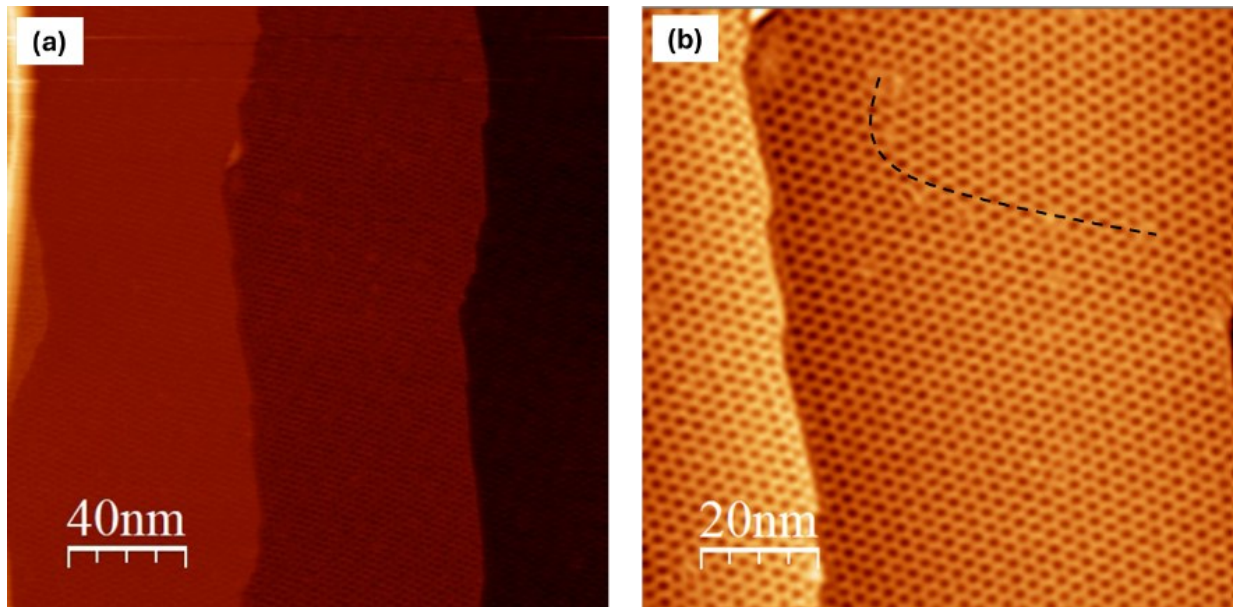
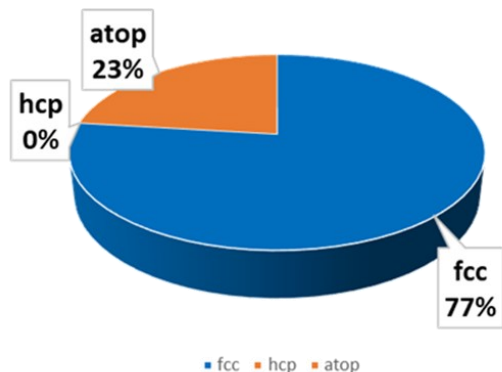


Figure S2. (a) A large-area STM image ($200 \times 200 \text{ nm}^2$) of Gr/Ir(111), showing continuous graphene coverage across multiple Ir(111) terraces, with no uncovered regions or visible defects at this scale, indicating uniform epitaxial growth (scanning parameter: -0.6 V , 1200 pA). (b) A $100 \times 100 \text{ nm}^2$ STM image of graphene spanning an Ir step edge, revealing distinct moiré domains with different orientations. One domain boundary is outlined by a black dashed line.

(a) Nucleation Site Occupation Chart



(b)

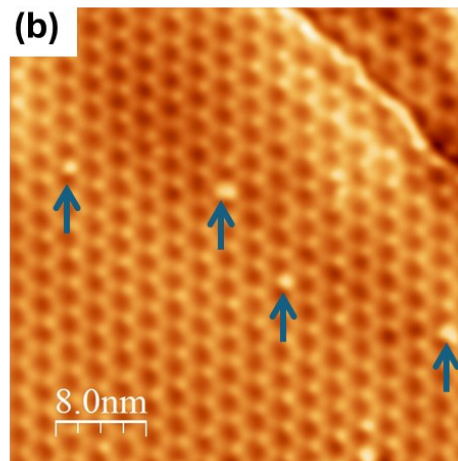


Figure S3. (a) Pie chart depicting the relative occupancy of Ni nucleation sites on the Gr/Ir(111) moiré template, evaluated from 15 nucleation events. (b) STM image showing early-stage Ni nucleation on Gr/Ir(111), with nucleation sites indicated by blue arrows. Scanning parameters: -0.6 V, 1100 pA.

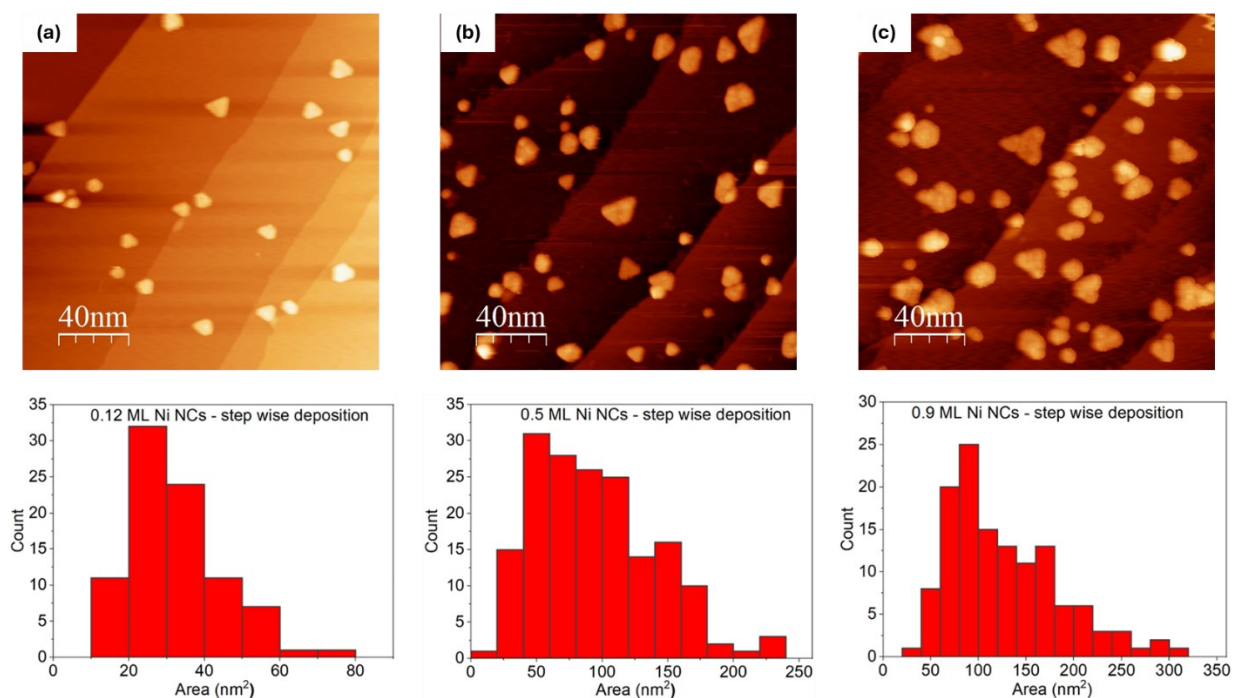


Figure S4. STM characterization of Ni nanoclusters grown on Gr/Ir(111) at room temperature. (a–c) STM images after Ni deposition at coverages of (a) 0.12 ML, (b) 0.5 ML, and (c) 0.9 ML, showing progressive growth of triangular nanoclusters with increasing size and density. Scanning parameters: (a) $+2.0$ V, 100 pA; (b) $+2.0$ V, 300 pA; (c) -2.7 V, 100 pA. Corresponding histograms of nanocluster area distributions are shown below each image, demonstrating the increase in average cluster area and the broadening of the distribution with coverage.

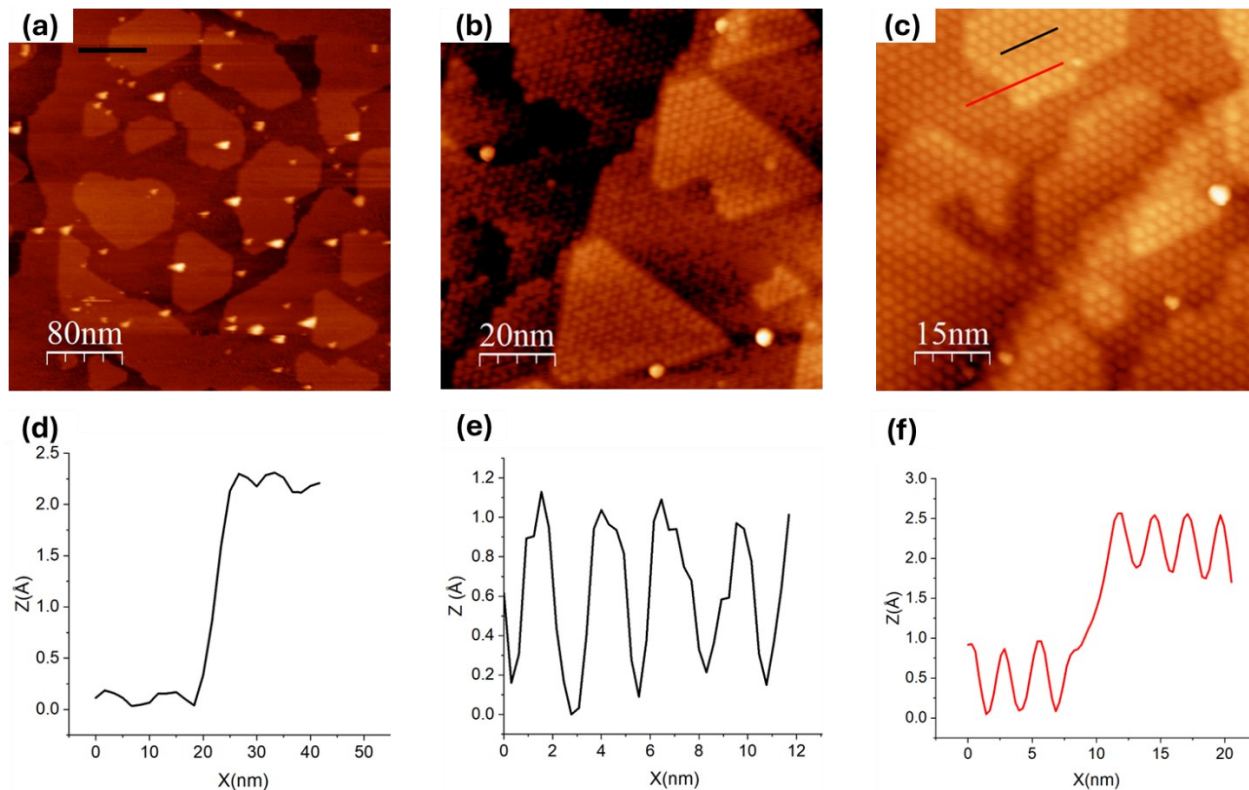


Figure S5. STM characterization of Ni intercalation between graphene and Ir(111), after deposition of 0.9 ML of Ni. (a) A large-area STM image ($400 \times 400 \text{ nm}^2$) showing intercalated Ni beneath graphene. (b) A $100 \times 100 \text{ nm}^2$ STM image revealing the round and clover-like moiré contrast of graphene after Ni intercalation. (c) A $75 \times 75 \text{ nm}^2$ STM image depicting the mostly round moiré appearance observed in some regions after intercalation. (d–f) Line profiles along the black line in (a) and the black and red lines in (c), respectively, highlighting the apparent corrugation associated with the moiré patterns. Scanning parameters: (a) $V = -1.6 \text{ V}$, $I = 400 \text{ pA}$; (b) $V = -0.9 \text{ V}$, $I = 600 \text{ pA}$; (c) $V = -0.9 \text{ V}$, $I = 500 \text{ pA}$.

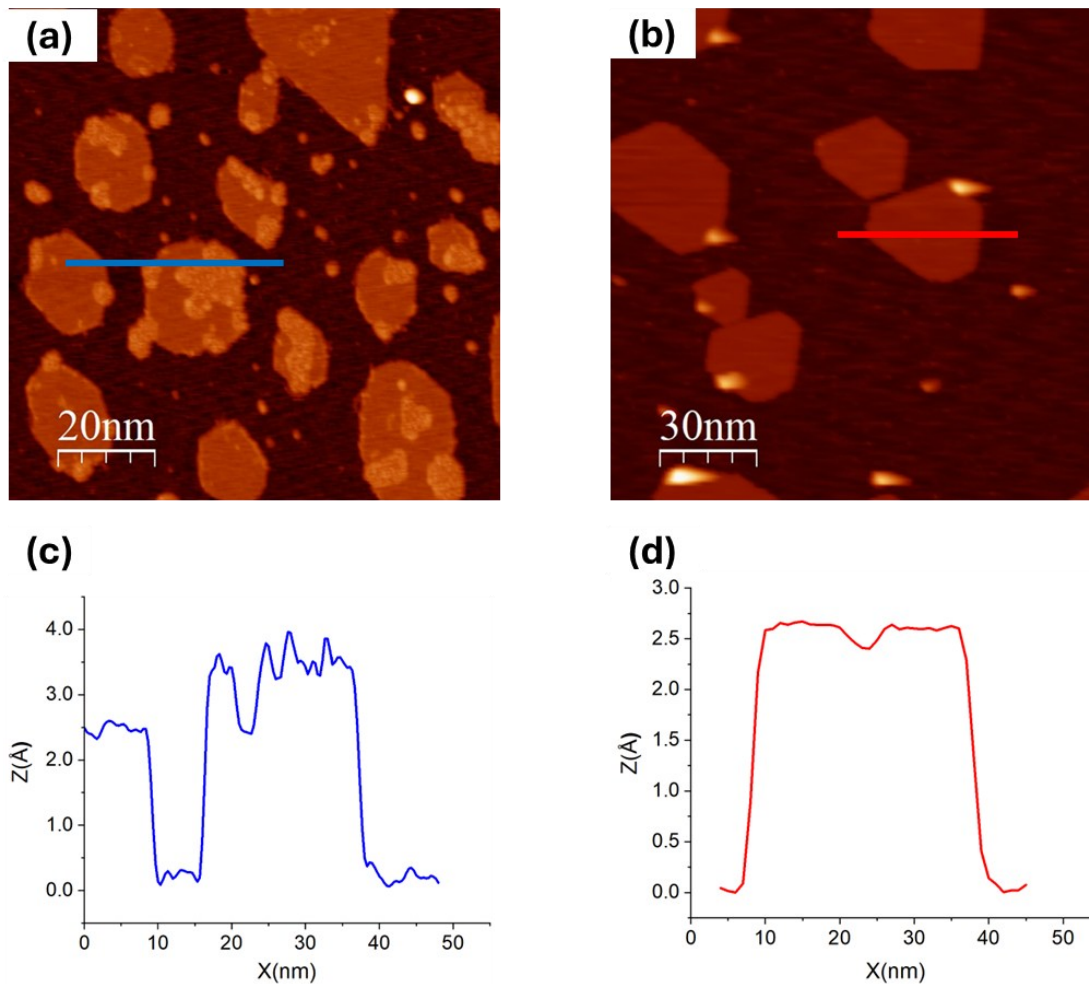


Figure S6. (a) STM topograph of 0.9 ML Ni deposited on Ir(111) and annealed to 900 K, revealing a two-layer Ni film with a rough second-layer morphology characteristic of kinetically limited growth ($V = -2.0$ V, $I = 200$ pA). (b) STM topograph of the same 0.9 ML Ni film after subsequent annealing to 1500 K, showing smoother Ni islands consistent with thermodynamically stabilized Ni overlayers and a reduced Ni coverage of ~ 0.4 ML due to high-temperature Ni desorption, analogous to the behavior of intercalated Ni beneath Gr/ Ir(111) ($V = +2.0$ V, $I = 200$ pA). (c),(d) Line profiles extracted along the lines indicated in (a) (blue) and (b) (red), respectively, illustrating the morphological variations.

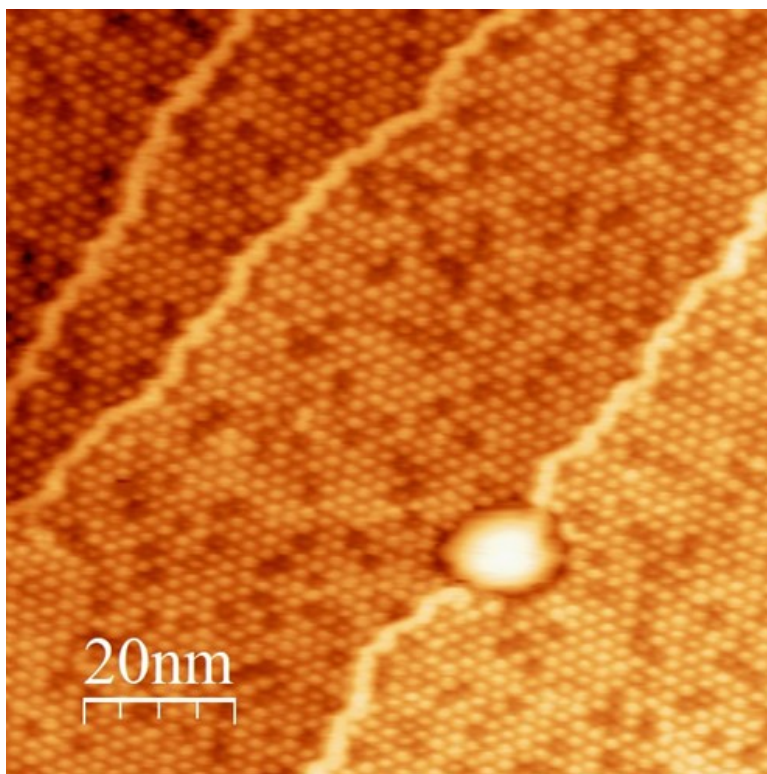


Figure S7. STM topography of Gr/Ir(111) after annealing at 1500 K. The sample was initially intercalated with 0.9 ML of Ni. The graphene moiré remains visible with a reversed contrast but retains its characteristic periodicity, indicating that the graphene lattice remains intact while the interfacial Ni becomes mobile at elevated temperatures and diffuses away from the interface, either toward step edges or desorbs. Scanning parameters: $V = -1.0$ V, $I = 200$ pA.