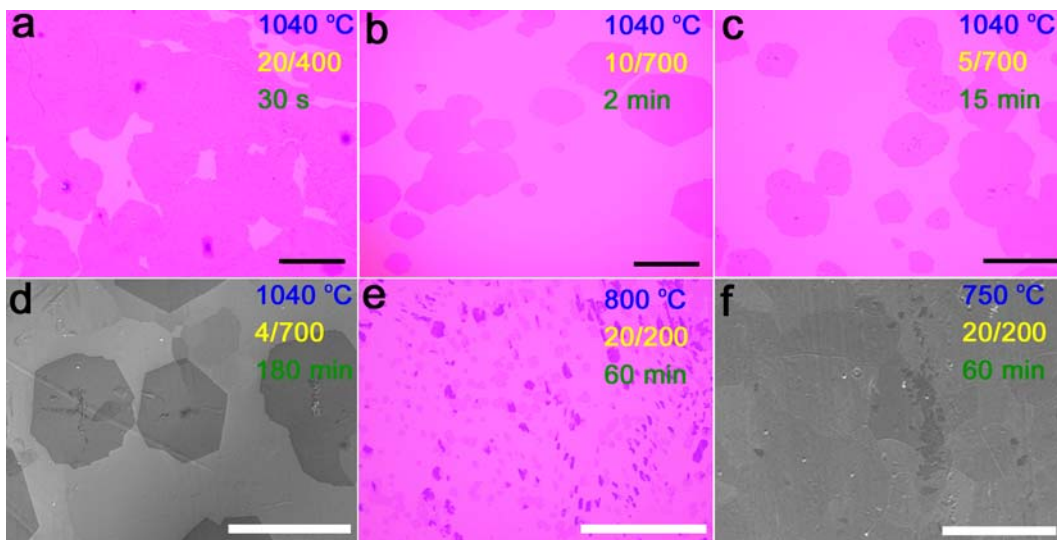


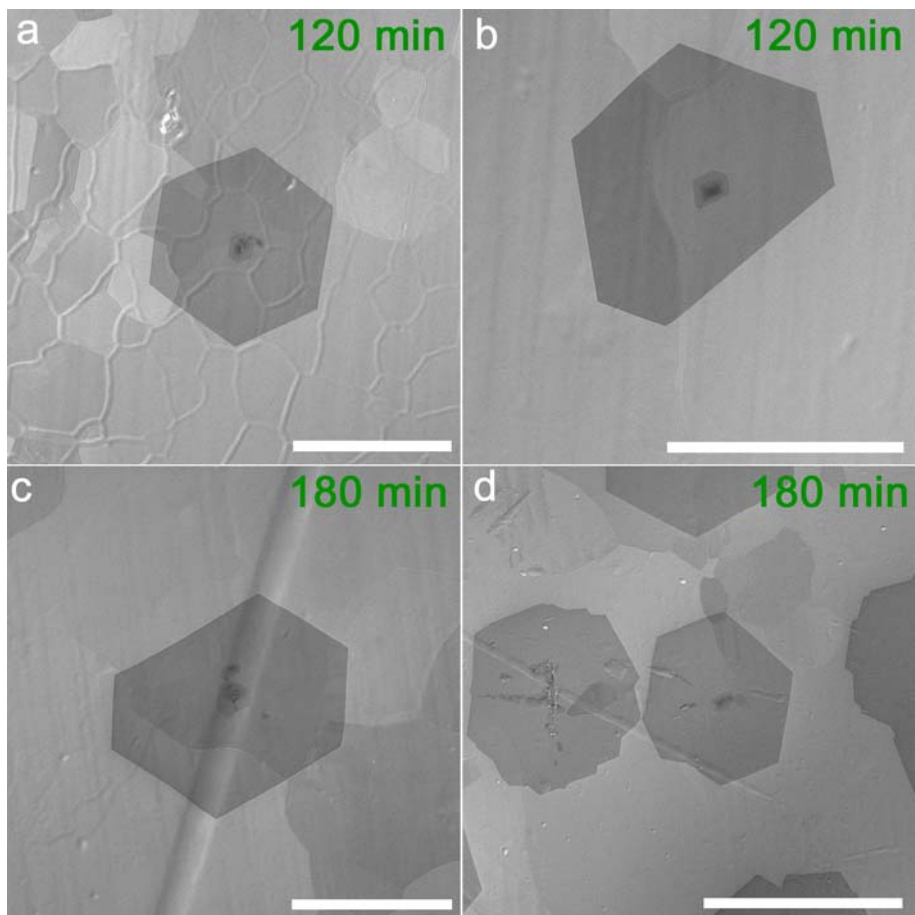
SUPPLEMENTARY INFORMATION

Supplementary Table S1. Summary of the growth conditions and corresponding grain sizes of graphene on polycrystalline Pt foils.

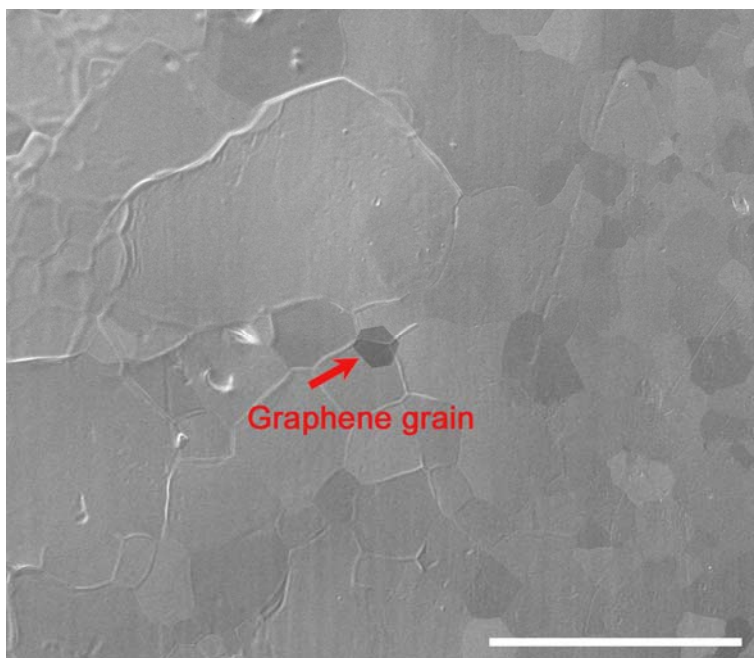
Temperature/°C	Methane/sccm	Hydrogen/sccm	Growth time/min	Grain size/ μm
1040	20	400	<1	>50
1040	10	700	<5	>100
1040	5	700	<30	>500
1040	4	700	>240	>1000
800	20	200	>240	>30
750	20	200	>240	no data



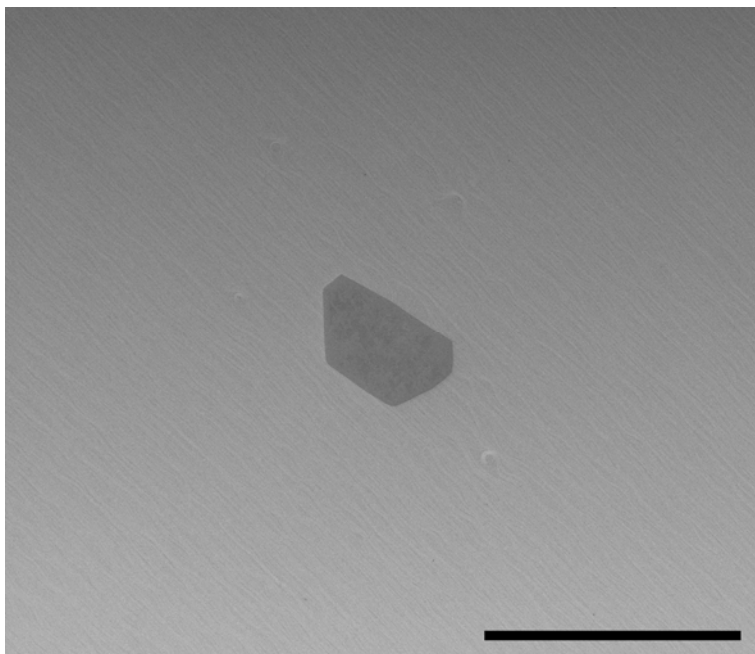
Supplementary Figure S1. Graphene grains obtained under different growth conditions. a-c, e, Optical images of transferred graphene grains on Si/SiO₂. **d, f,** SEM images of graphene grains grown on polycrystalline Pt foils. The blue numbers are the growth temperature, yellow numbers the flow rates (sccm) of CH₄ and H₂, and green numbers the growth time. The scale bar in **a** is 50 μm, in **b** 100 μm, in **c** 500 μm, in **d** 1 mm, in **e** 200 μm, and in **f** 100 μm.



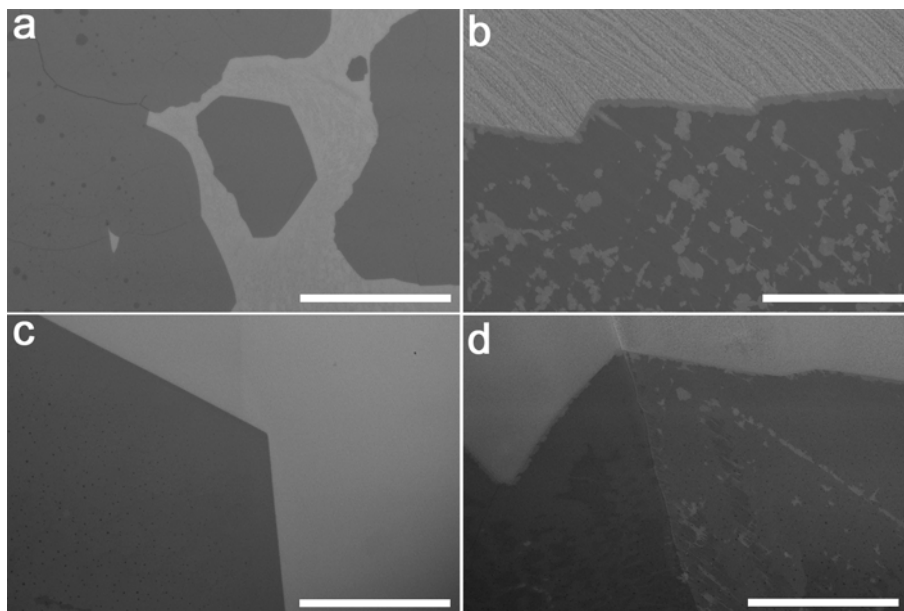
Supplementary Figure S2. SEM images of the as-grown graphene grains on the same polycrystalline Pt foils after repeated use more than 500 times. They show no obvious difference in graphene growth. In all cases, the growth temperature was 1040 °C, and the flow rates of CH₄ and H₂ were 4 sccm and 700 sccm, respectively. The green numbers are the growth time. Note that large graphene grains contain some reflex angles at edges in **d**, but smaller ones contain no visible reflex angles in **a-c**. The scale bar in **a-c** is 400 μm, and in **d** 1 mm.



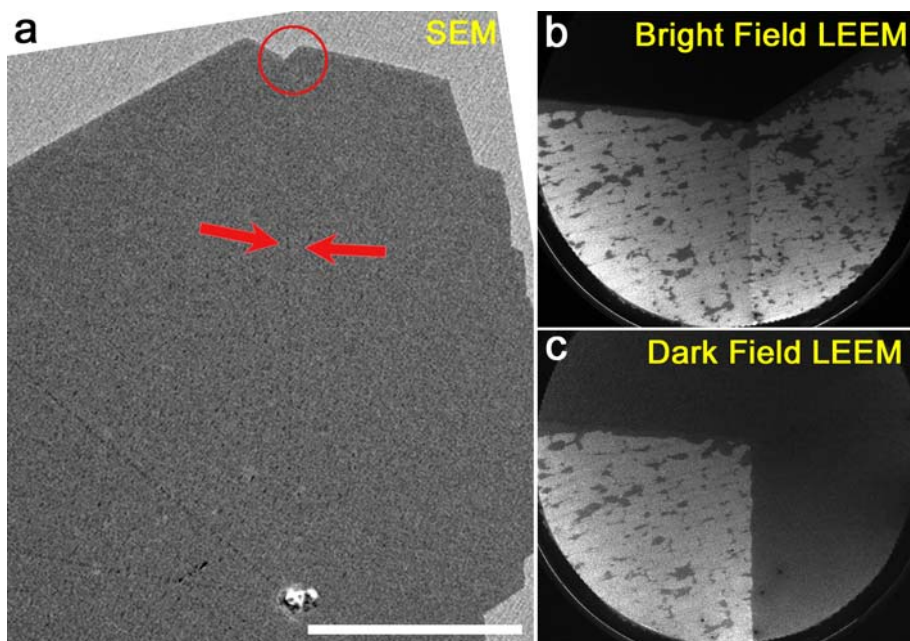
Supplementary Figure S3. SEM image of a single hexagonal single crystal graphene grain grown on a Pt foil in a region of 2.5 mm × 2.5 mm. The scale bar is 1 mm.



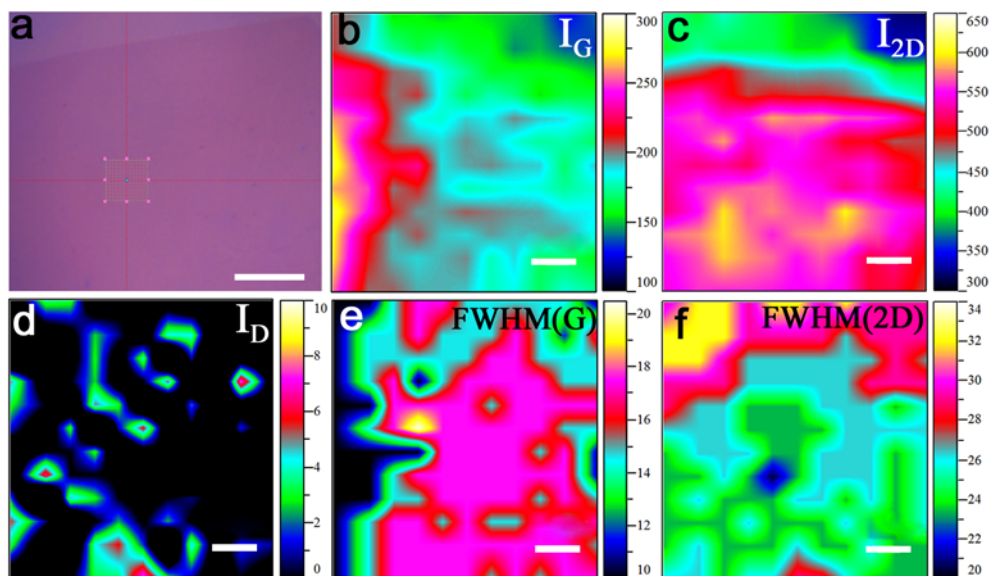
Supplementary Figure S4. SEM image of a hexagonal single crystal graphene on Pt(111). Graphene was grown with 4 sccm CH₄ and 700 sccm H₂ at 1040 °C for 45 min. The scale bar is 200 μm.



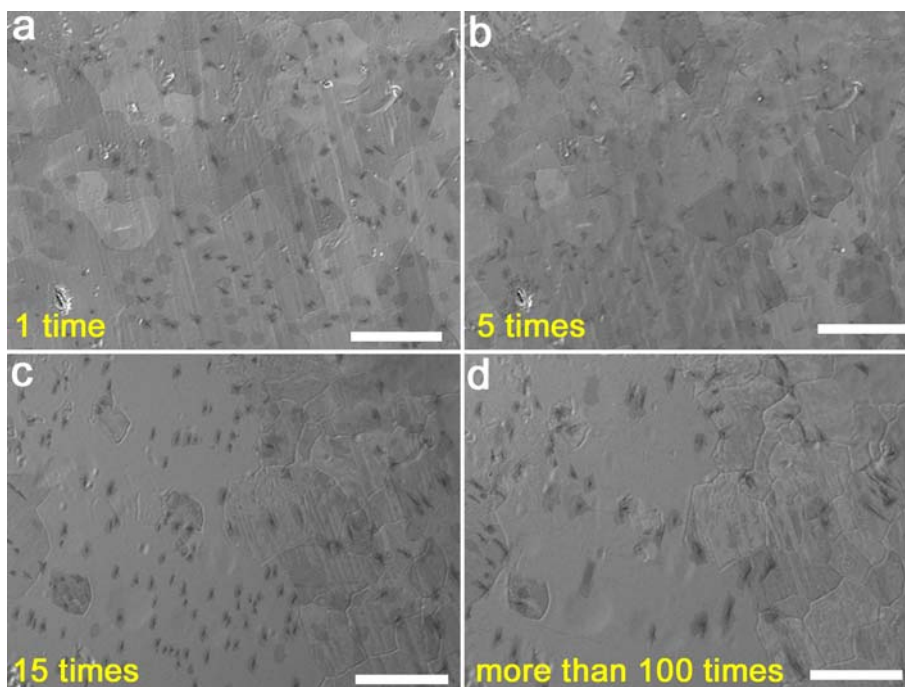
Supplementary Figure S5. Surface change of graphene on a Pt substrate under an ultra-high vacuum (UHV) system. **a, b**, SEM images of as-grown graphene grains on single crystal Pt(111) and those after being held in the UHV system for ~10 hrs. The scale bar in **a** is 10 μm , in **b** 20 μm . **c, d**, SEM images of as-grown graphene grains on a polycrystalline Pt foil before and after being held for ~10 hrs in the UHV system. The scale bars in **c-d** are 20 μm . It can be found that some bubble-like regions appear in these graphene grains after being held in the UHV system (also see Figure 1d, e, g, h in the manuscript). They show black areas in LEEM images and white areas in SEM images. These surface changes are similar to those observed in decoupled epitaxial graphene produced on SiC by hydrogen intercalation³⁹. We suggest that this change comes from the decoupling of graphene caused by H₂ degassing from the Pt substrate under UHV¹⁵ due to the weak interaction between the graphene and the Pt substrate⁹.



Supplementary Figure S6. Analysis of grain boundaries among graphene grains by SEM and LEEM. a, High-contrast SEM image of a graphene grain on Pt(111), the same one shown in Figure 1c in the manuscript. The scale bar is 100 μm . **b, c,** Bright field (**b**) and dark field (**c**) LEEM images of the red circle region indicated in **a** with a reflex angle at the edge, the same ones shown in Figure 3d and 3e in the manuscript. Note that most of the grain boundaries are visible in SEM images owing to the preferential decoration of grain boundaries with surface contamination²¹. The black boundary denoted by red arrows in **a** is the grain boundary determined by LEEM in **b** and **c**.



Supplementary Figure S7. Optical and Raman mapping images of a transferred graphene on a Si/SiO₂ substrate. **a**, Optical image of a transferred graphene on Si/SiO₂, with the Raman mapping measurement region indicated by a square box (6 μm × 6 μm). The scale bar is 10 μm. **b-d**, Raman mapping images of the intensity of G band (1560 to 1620 cm⁻¹), 2D band (2640 to 2720 cm⁻¹), and D band (1300 to 1400 cm⁻¹). Raman mapping images of the FWHM of **(e)** G band and **(f)** 2D band. The scale bars in **b-f** are 1 μm.



Supplementary Figure S8. SEM images of the as-grown graphene films on the same polycrystalline Pt foils after repeated uses. a, the first time, b, the 5th time, c, the 15th time, d, after more than 100 times, showing no obvious difference in the films. The scale bars in a-d are 100 μm .

Supplementary References

39. Riedl, C., Coletti, C., Iwasaki, T., Zakharov, A. A. & Starke, U. Quasi-Free-Standing Epitaxial Graphene on SiC Obtained by Hydrogen Intercalation. *Phys. Rev. Lett.* **103**, 246804 (2009).