Supporting Information

## Geometry-Tailored Freestanding Epitaxial Pd, AuPd, and Au

## **Nanoplates Driven by Surface Interactions**

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**Fig. S1.** Schematic illustration showing the experimental setup for the epitaxial growth of single-crystalline Pd, AuPd, and Au nanoplates. While a Pd lump was used as the only precursor to synthesize Pd nanoplates, an Au lump was employed as the only precursor to obtain Au nanoplates. Both Pd and Au lumps were used as precursors to prepare AuPd nanoplates.



Fig. S2. TEM data of a single AuPd alloy nanoplate. (a) TEM image of the nanoplate. (b)HRTEM image and its FFT pattern of the nanoplate. (c,d) EDS mapping of the nanoplate.(e) EDS spectrum of the nanoplate, confirming that the nanoplate has approximately 1:1atomic ratio of Au and Pd.



**Fig. S3.** XRD patterns of Pd, AuPd, and Au nanoplates. The peaks correspond to the (111) and (200) peaks of face-centered cubic Pd, AuPd, Au, which are shifted to a higher d-value (lower 20 value) as we move from Pd to AuPd and Au. XRD patterns were taken on a Rigaku D/max-RC (12 kW) diffractometer using Cu K $\alpha$  radiation.



**Fig. S4.** Au nanostructures obtained at different Au precursor temperatures.  $45^{\circ}$  tiltedview SEM images of (a) Au nanowires, (b) Au nanobelts, and (c) Au nanoplates grown on a *m*-cut substrate at a precursor temperature of 1100 °C, 1150 °C, and 1200 °C, respectively. As the precursor temperature increases, the Au vapor pressure increases. The higher Au vapor pressures favorably promote horizontal growth of nanostructures and lead to the growth of nanobelts and nanoplates.



**Fig. S5.** Additional SEM images of (a) Pd, (b) AuPd, and (c) Au nanostructures grown on *a*-cut sapphire, showing intermediate growth states including square pyramidal seeds and slightly grown seeds of Pd, AuPd, and Au nanoplates.



**Fig. S6.** Structural model of a square pyramidal nanocrystal and a triangular nanocrystal. (a) Structural model of the square pyramidal nanocrystals enclosed by four {111} side planes and a {100} bottom plane. (b) Structural model of the triangular nanocrystals enclosed by {111} top and side planes and a {111} bottom plane. These square pyramidal and triangular nanocrystals are twin-free single crystalline, because they can be obtained by cutting twin-free single-crystalline octahedral nanocrystals.



**Fig. S7.** Classification of (a) Pd, (b) AuPd, and (c) Au nanostructures grown on *a*-cut sapphire. Nanostructures in the sky-blue circles are the nanocrystals derived from square pyramidal seeds, which possess  $\{111\}$  side planes and a  $\{100\}$  bottom plane. Nanostructures in the red circles are the nanocrystals derived from triangular seeds, which have  $\{111\}$  top and side planes and a  $\{111\}$  bottom plane.



**Fig. S8.** Monolayer graphene grown on single-crystalline Pd nanoplates. (a) Schematic illustration showing the experimental procedure. After a poly(methyl methacrylate) (PMMA) thin film (100 nm) was spin-coated on single-crystalline Pd nanoplates, reductive gases (H<sub>2</sub>/Ar 3 %) flowed at a rate of 500 sccm, maintaining the chamber pressure at 50 Torr. A monolayer graphene was grown on single-crystalline Pd nanoplates at a temperature of 970 °C for 10 min. (b,c) Optical images and Raman spectra of monolayer graphene grown on single-crystalline Pd nanoplates. The graphene exhibits the G peak and 2D peak without showing the D peak. The I<sub>2D</sub>/I<sub>G</sub> intensity ratio is about 2, suggesting that the graphene is a monolayer.