# Graphene Template Induced Growth of Single-crystalline Gold Nanobelts with High Structural Tunability

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**Figure S1.** (a) Representative TEM image of mlG used in this work. (b) High resolution TEM image showing 11-layered graphene edge with the interspace of 3.60 Å.



**Figure S2** Raman spectroscopic characterization of mlG before and after reaction with gold precursor. (a) Normalized Raman spectra of pristine mlG, mlG-Au nanobelts at 9 hr and mlG-Au nanobelts at 16 hr in the range of 1000 cm<sup>-1</sup> to 2800 cm<sup>-1</sup>. (b) Raman spectra from 1560 cm<sup>-1</sup> to 1600 cm<sup>-1</sup> (the green dotted box in (a)) showing slight redshift of G peak with the reaction time.



**Figure S3.** Morphologies of mlG-Au nanobelts. (a) Au nanobelts lying flat and parallel to the surface of multilayer graphene sheet. (b) An ultralong Au nanobelt isolated from the mlG template. (c) Au nanobelts fluctuating on graphene template. (d) Thickness of the nanobelts found to be sub-100 nm.



**Figure S4.** Elemental analysis of *ml*G-Au nanobelts. Only the elements of carbon, silicon and gold are recorded in the element spectrum. Note silicon signal originates from the substrate.



**Figure S5.** (a) TEM image of gold precursor reacting with graphene for 1hr. (b) and (c) Atomic structures and corresponding FFT patterns of two Au nanoplatelets in (a).



**Figure S6.** SEM image (a) and TEM image (b) of Au nanobelts grown on mlG template for 2 hr, forming Au nanoplates as the embryos of the nanobelts. (c) Selected Au nanoplates showing the favorable extending direction. (d) Atomic structure of selected area of (c). (e)The corresponding FFT of (d).



**Figure S7.** (a) Au nanobelts with two representative shapes. (b) Schematic image of two typical Au nanobelts: triangular and straight end.



**Figure S8.** Raman shift of G peak and the modification of  $I_D/I_G$  of *ml*G with the number of plasma scans.



**Figure S9.** Quantification analysis of hydrophilic functional groups on mlG before (a) and after (b) oxygen plasma treatment (30 scans) using X-ray photoelectron spectroscopy. (c) A selected area of Au clusters nucleated on plasma oxidized mlG (30 scans) at 1 hr and ambient temperature. Inset is the corresponding FFT of the entire area. (d) Atomic structure of Au clusters selected in (c).



Figure S10. The relation between width of Au nanobelts and gold precursor concentrations.



**Figure S11.** Au nanobelts synthesized on mlG by using 12.0 mM HAuCl<sub>4</sub> at the time of (a) 4 hr and (b) 16 hr.