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Electronic Supplementary Information

In-Situ Atomic Imaging of Coalescence of Au Nanoparticles on Graphene: Rotation and Grain Boundary Migration

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S1. Methods

Synthesis of Au nanoparticles. 10 mg of gold (III) chloride hydrate ($HAuCl_4 \cdot xH_2O$, 99.999 %, Aldrich, 1.5 mmol) were dissolved in 10 ml of oleylamine (70 %, Aldrich) at 40 °C with vigorous stirring. The solution was continually heated to 160 °C and then maintained for 2 hours. The solution was cooled down to room temperature, and Au nanoparticles were precipitated in ethanol by centrifugation at 10000 rpm for 10 min.

Graphene preparation. Graphene were synthesized by using chemical vapor deposition (CVD) on 5-μm-thick copper foil (99.8 % Alfa Aesar, Ward Hill, MA), following a previous report [S1]. The CVD-grown graphene are predominantly single-layer graphene confirmed by the Raman spectroscopy and electron diffraction pattern [S2, S3].

Direct graphene transfer onto TEM grid. The synthesized graphene on Cu foil was directly transfered onto TEM grid without other supporting materials, such as poly(methyl methacrylate) (PMMA) or polydimethylsiloxane (PDMS), following a previous report [S4]. The CVD grown graphene on Cu foil is attached to a Quatifoil TEM grid with an array of holes 2 μm in diameter. The supporting Cu foil was etched with $Na_2S_2O_8$ solution (concentration of 0.1 mg $Na_2S_2O_8$ / 1 mL DI water) overnight.

Characterization. The *in-situ* TEM investigation in the coalescence of Au nanoparticles was conducted using aberration-corrected atomic-resolution electron microscopy (JEM-ARM200F operated at 200kV). We set 0.1 seconds of exposure time and the time interval between acquisitions of two images is 1 sec. Coalescence of the Au nanoparticles was initiated by the electron beam irradiation in the TEM rather than the thermal annealing. Raman spectroscopy measurement of graphene was performed using high resolution dispersive Raman microscope (ARAMIS, Horiba Jobin Yvon, 514 nm Ar ion laser).

HRTEM image simulation. We simulated HRTEM image of face-centered cubic Au along [110] direction with under-coordinated atoms on the edges, via multislice method. We used Mac Tempas

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program for HRTEM image simulation and Crystal Kit program to bulid an Au structure with surface atoms. We set the particle thickness of 6.3 nm, the imaging focus of 10 nm and the spherical abberation of 0.5 mm, which correspond with experimental condition.

S2. Supplementary figures

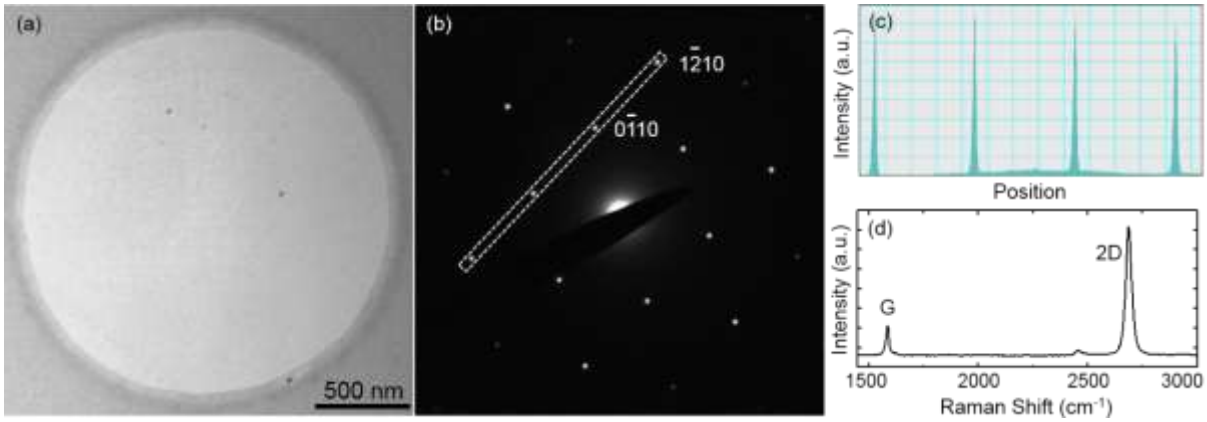

Fig. S1 TEM and Raman spectroscopy images of graphene. (a) Bright-field TEM image of transferred graphene onto quantifoil holey carbon grid. (b) Diffraction pattern of monolayer graphene. (c) Histogram of diffraction spots from dashed rectangle in (b) shows that the intensity ratio of $0\bar{1}10$ and $1\bar{2}10$ is about 1, which indicates that synthesized graphene is monolayer [S5]. (d) Raman spectrum of graphene.

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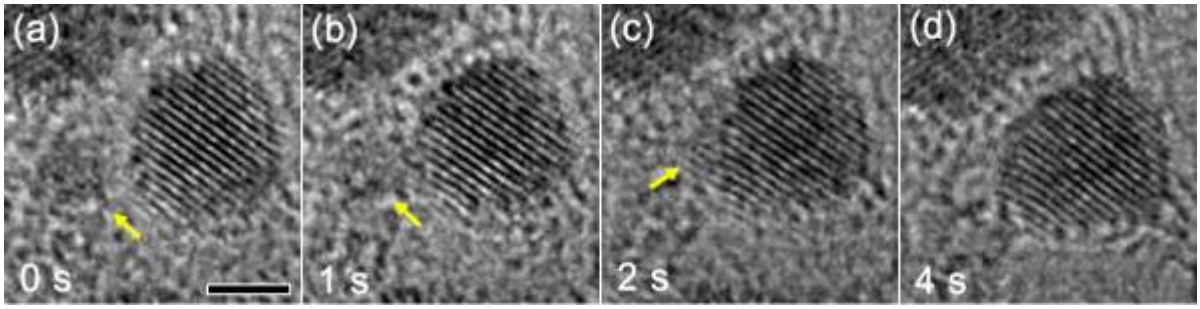

Fig. S2 Real-time HRTEM images of small nanoparticles fused into relatively large nanoparticle. The yellow arrows indicate incoming small particle. Scale bar is 2 nm.

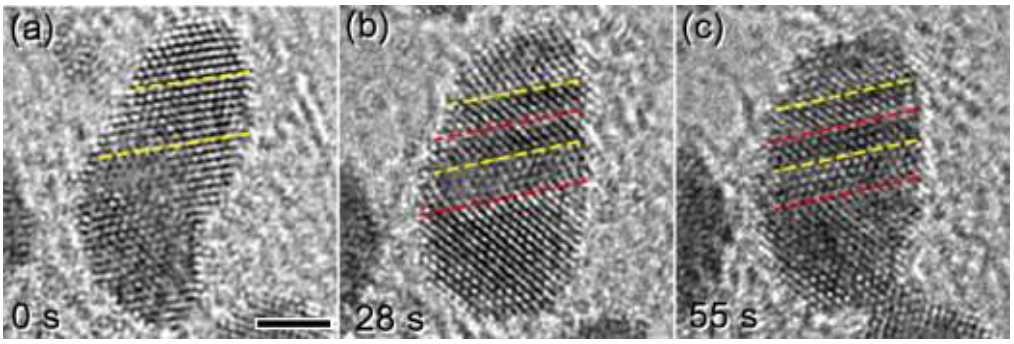

Fig. S3 The sequential HRTEM images of twin boundary formation due to the electron bombardment in the TEM. (a) Combined particle has twin boundaries, indicated by yellow dashed lines. (b and c) Other twin bounaries are formed during coalescence by the electron bombardment in the TEM. The created twin boundries are indicated by red dashed lines. Scale bar is 2 nm.

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