Coalescence between Au nanoparticles as-induced by nanocurvature effect and electron beam athermal activation effect

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Supporting Information

The beam energy deposition rate on specimen as used herein is an independent parameter to account for energetic beam athermal activation effect which is related to the beam irradiation current and is defined [24] as the beam energy deposited per volume of specimen per duration of irradiation. After a simple calculation, we could easily get the beam energy deposition rate v_E

$$v_E = \lim_{\Delta V \to 0} \left| \Delta E / (\Delta t \cdot \Delta V) \right| = j\, U / h$$

where ΔE is the beam energy deposited on specimen, ΔV is the specimen volume irradiated, Δt is the duration of irradiation, j is the beam irradiation current density, U is the accelerating voltage and h is the local thickness of specimen along the direction of incident beam.

Figure S1 shows the *in situ* structural evolution of coalescence between two adjacent Au NPs on the surface of an a-SiO_x NW during another uniform irradiation of e-beam with a beam current density of 50.2 A/cm^2, in contrast to the lower current density of 15.8 A/cm^2 in Figure 1 in the manuscript. Similarly, before the irradiation, as shown in Figure S1(a), the two Au NPs have nearly the same size (~7.5 nm in diameter) with a separation distance about 1 nm. Furthermore, both of the NPs appear to be face-centered cubic (or fcc) single crystal structure. The measured d-spacing values of the two Au NPs are both around 2.4 Å, which are well attributed to (111) planes of fcc Au (JCPDF # 040784) [23].It is also worth noting that their relative crystallographic orientations do not match in which there is a deviation of 88° from each other initially. Comparing with Figure 1, under the irradiation with a higher current density of 50.2 A/cm^2 (see Figure S1), the two contacted Au NPs coalesce quickly within 240 s whereas under the irradiation with a lower current density of 15.8 A/cm^2 (see Figure 1 in the manuscript), the two contacted Au NPs coalesce slowly within 380 s. In addition, we also note that the rotation rate of the NPs in Figure S1 is also obviously much faster than that in Figure 1 with the increased beam current density. According to the definition of beam energy deposition rate on specimen [24], the beam energy deposition rate is proportional to the irradiation current density. So in this work, a higher current density in FigureS1 would result in a higher beam energy deposition rate and thus cause a more pronounced structural change or instability. That is, with the current density or energy deposition rate being raised, the coalescence accelerated and vice versa. This indicates the beam activation

effect is athermal and has a crucial effect on the structural change or the coalescence.

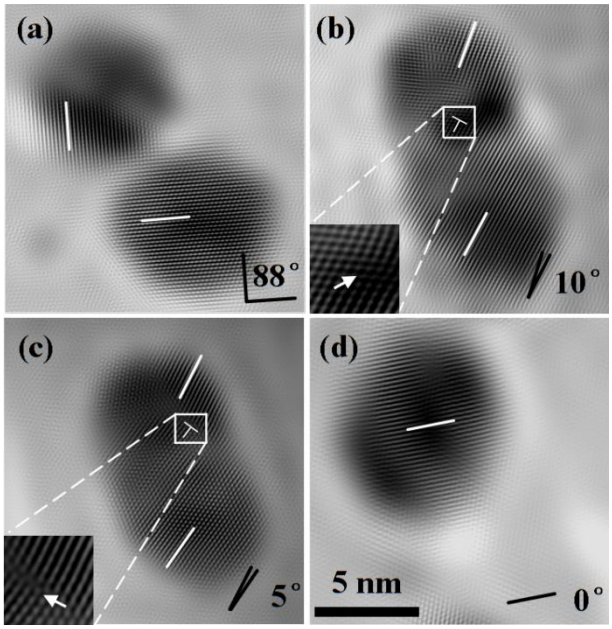

Figure S1. Sequential HRTEM images showing the coalescence between two adjacent Au NPs as induced by irradiation of uniform e-beam with a current density of 50.2 A/cm^2 to different irradiation times (s): (a) 0; (b) 20; (c) 100; (d) 240. The white lines indicates the (111) atomic plane orientations.

With the same practice to our previous experiments on the e-beam irradiation of the other low dimensional nanostructures, we also observed many times that during the irradiation, the structure change would always stop immediately once the beam irradiation is suspended (that is, the beam intensity was spread to an around 100 times weaker intensity). This indicates the beam activation effect is instant and athermal which is crucial or indispensable to the structural change or the coalescence.