Supporting information

Oriented-attachment dimensionality build-up *via* van der Waals interaction

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1. Facet effect on the vdW of OA assembly of Ag nanorods

OA assembly of Ag nanorods along [111] crystalline orientation experiences smaller vdW compared with [100] and [110] directions. The vdW of OA assembly along [100] and [110] shows a rather small difference as evidenced by the nearly overlapped red curve and black curve. Therefore, NRs growth along [111] is thermodynamically unfavorable.

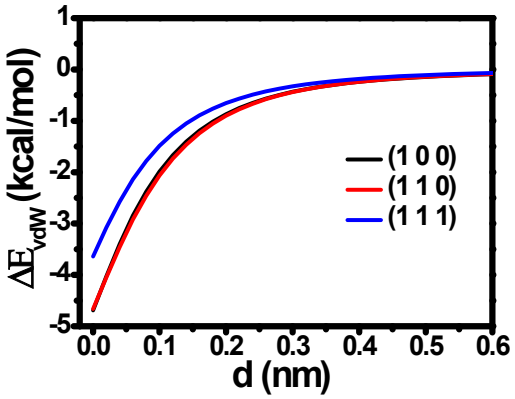

Figure S1. Calculated vdW interaction *versus* separation between NR and NP of different crystal faces along the axes of NRs with the diameters of 1.2 nm. The AR of Ag NR is fixed at 10.

2. The effect of diameter on the attaching energy of axial growth and off-axial growth of nanorods

Figure S2 shows $\Delta E_{vdW}(0)$ *versus* the size of NRs and NPs in the on axial OA assembly and the off-axial assembly (0.5*l*) between a NR and a NP. As the size of nanoparticles and nanorods increases, the difference of attachment energy between axial growth and 0.5*l* off-axial growth increases significantly.

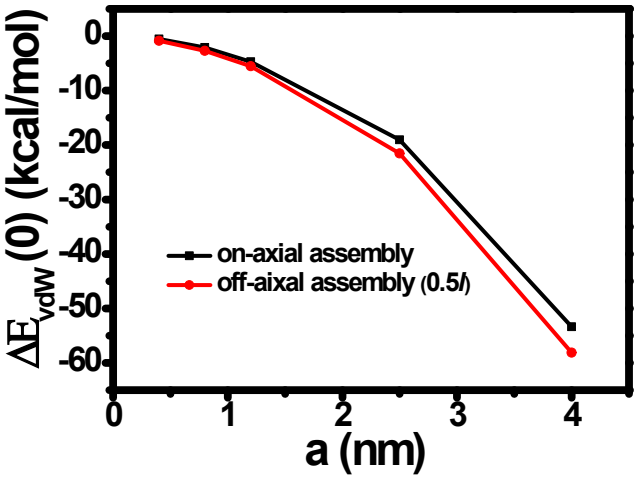

Figure S2 $\Delta E_{vdW}(0)$ *versus* the size of NRs and NPs in the on axial OA assembly and the off-axial assembly (0.5*l*) between a NR and a NP.