## **Supporting Information**

## An experimental and theoretical investigation of the anisotropic branching in gold nanocrosses

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Fig. S1 XRD pattern of the Cu nanostructures that formed from the disproportionation of  $Cu^+$  when CuCl was heated in oleylamine at 200 °C for 10 min.



**Fig. S2** Typical morphologies of the Au nanocrosses obtained using Method 1. HRTEM and TEM (inset) images of (A) doubly-twinned Au nanocross with  $D_{2h}$  symmetry and (B) singly-twinned Au nanocross with  $C_{2v}$  symmetry. The XRD pattern of the Au nanocrosses is presented in the lower inset of (B).



**Fig. S3** TEM image of the Au nanostructures obtained using Method 2 when the pre-formed anisotropic Au seeds were allowed to grow at 100 °C for 10 min.



Fig. S4 TEM image and HRTEM image (inset) of  $\sim$ 12 nm multiply-twinned Au nanocrystals prepared by reduction of AuCl<sub>3</sub> by oleylamine at 100 °C.