

# Supporting Information

## Metal-Metal Binary Nanoparticle Superlattices: A Case Study of Mixing Co and Ag Nanoparticles

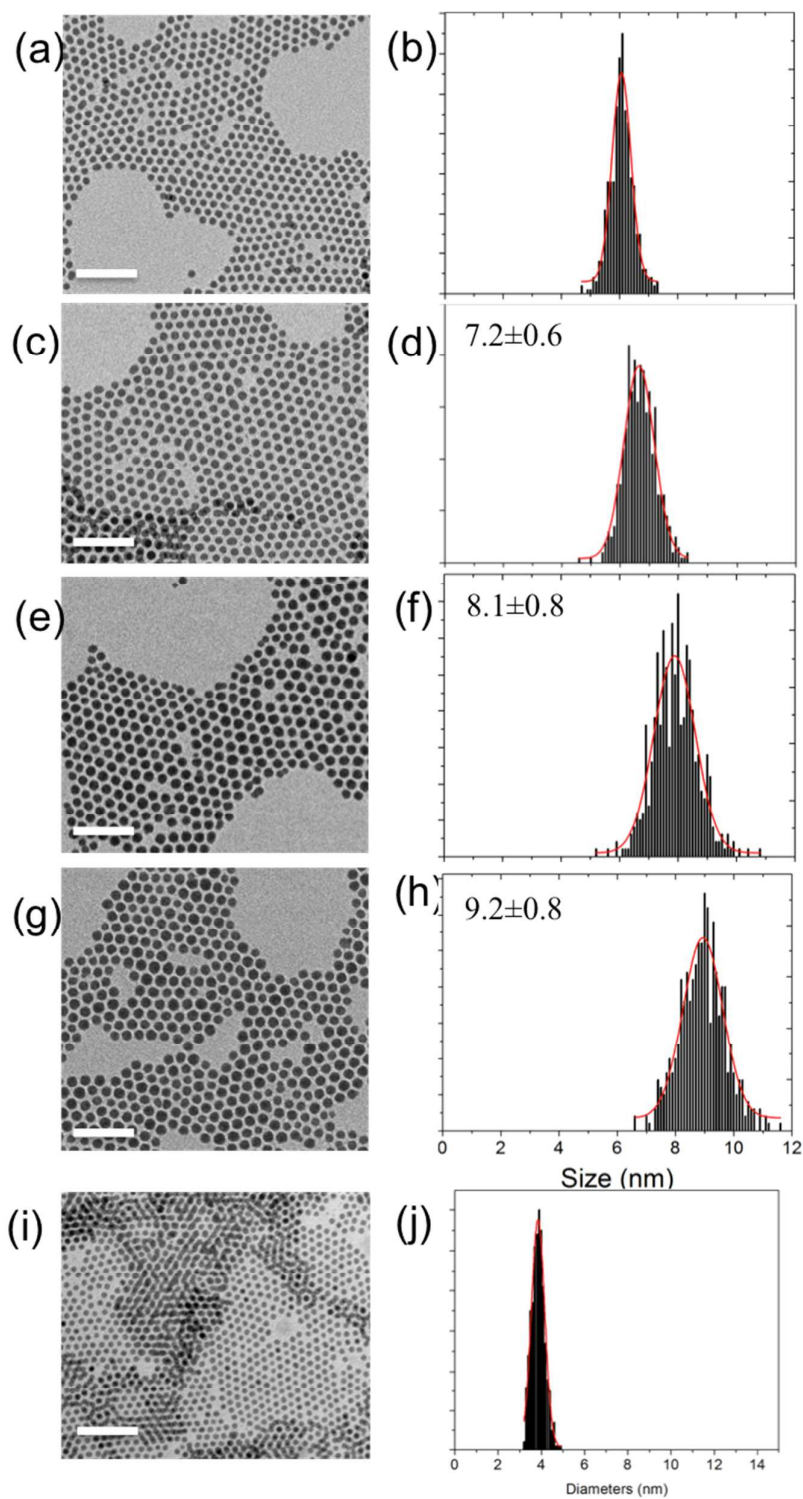
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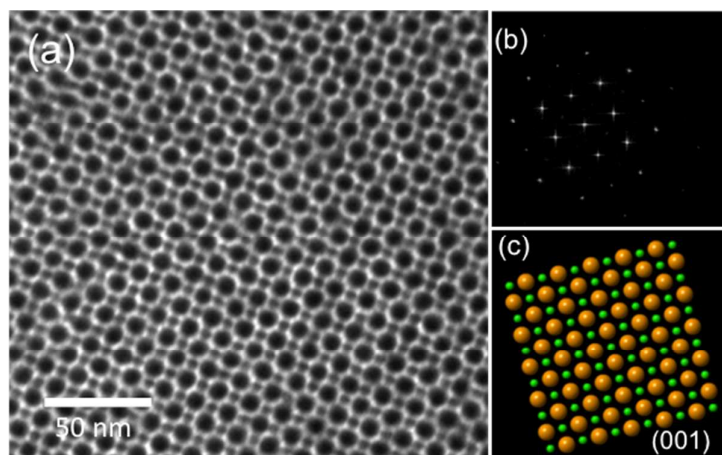
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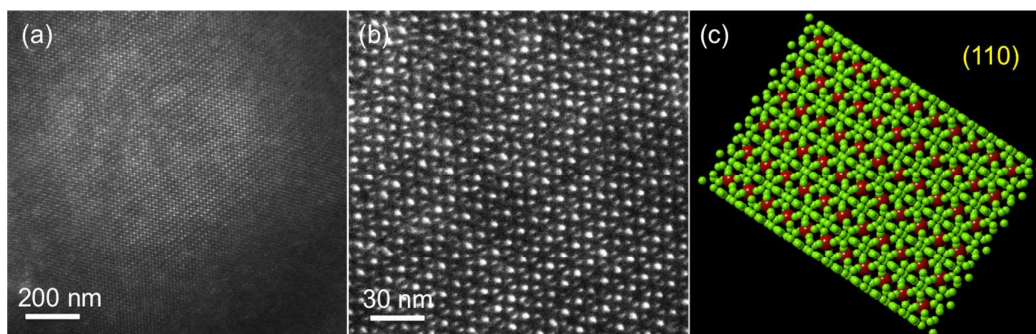
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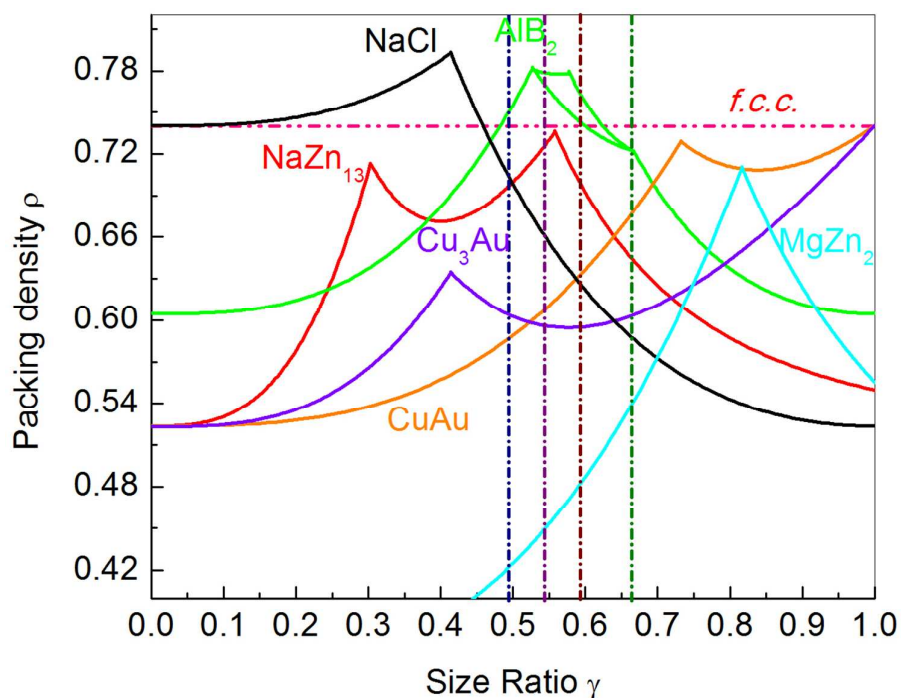
**Figure S1.** Tem images of the nanoparticles that were used as building blocks for the growth of binary nanoparticle superlattices: (a) 6.0-nm Co; (c) 7.2-nm Co; (e) 8.1-nm Co; (g) 9.2-nm Co; (i) 4.0-nm Ag; (b), (d), (f), (h) and (j) are the corresponding size histogram.



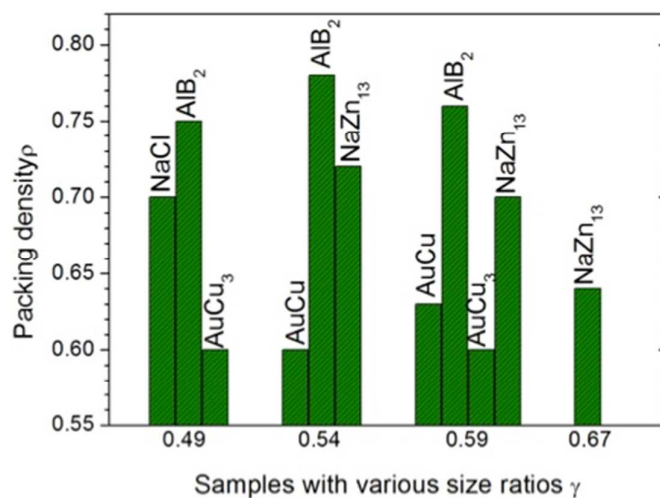
**Figure S2.** AuCu-type CoAg binary nanoparticle superlattices with (001) planes parallel to the substrate: (a) TEM image; (b) corresponding FFT pattern; (c) crystal model.



**Figure S3.** TEM images of (110) planes of NaZn<sub>13</sub>-type structure (a,b). Panel (c) is the model of (110) plane of NaZn<sub>13</sub>.



**Figure S4.** Calculated space-filling curves for NaCl, AuCu, AlB<sub>2</sub>, MgZn<sub>2</sub>, AuCu<sub>3</sub> and NaZn<sub>13</sub> binary structures. Dashed red line is the space-filling of single-component fcc structure. The Dashed vertical lines show  $\gamma$  for nanocrystals studied in this work.



**Figure S5.** The space filling of the all the binary structures observed at all experimental  $\gamma$  ratios.