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Understanding the strain-dependent structure of Cu nanocrystals in Ag-Cu nanoalloys

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Electronic Supplementary Information

Table S 1. The energy of a nanoalloy (ΔE) for the EAM potential used in the current work (ΔE_{EAM}), the Gupta potential (ΔE_{Gupta}), and based on the density functional theory (DFT) calculations (ΔE_{DFT}). (ΔE is measured with respect to the unreconstructed structures corresponding to each nanoalloy. The unreconstructed structure at the sizes 147, 192, and 201 are Ino decahedron, Marks decahedron, and truncated octahedron respectively. The values corresponding to the Gupta potential and DFT are taken from the ref. 44 of the main manuscript.

Nanoalloy	**Structure**	ΔE_{EAM} **(eV)**	ΔE_{Gupta} **(eV)**	ΔE_{DFT} **(eV)**
$Ag_{92}Cu_{55}$	reconstructed decahedron	-5.79	-3.87	-5.60
$Ag_{92}Cu_{55}$	Mackay icosahedron	-7.15	-5.30	-6.83
$Ag_{117}Cu_{75}$	reconstructed decahedron	-2.67	-1.73	-2.64
$Ag_{122}Cu_{79}$	pyritohedron	-3.58	-2.80	-3.48

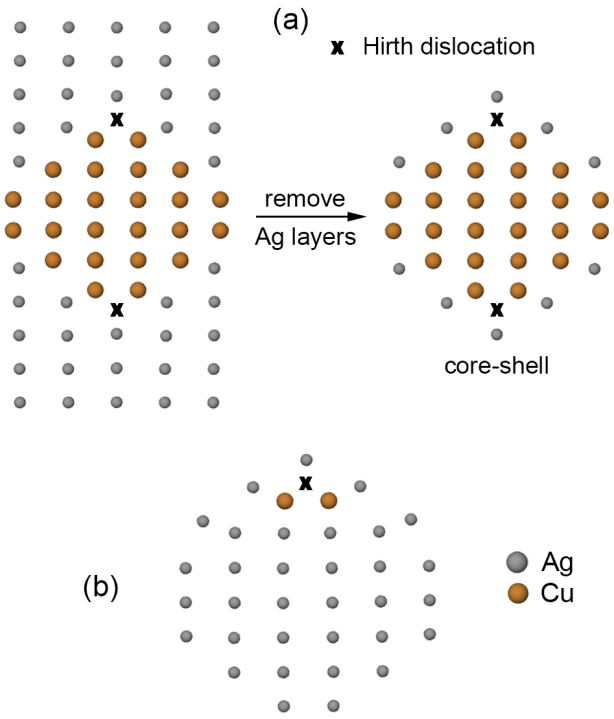

Fig. S 1. (a) By removing the Ag layers and retaining only one layer of Ag results in a core-shell structure. (b) A truncated octahedron Ag-Cu nanoalloy having the same dislocation structure as the bulk Ag-Cu shown in (a).

In the Fig. S1, we demonstrate the equivalence of the atomic arrangement of the surface dislocation and the bulk dislocation. In the Fig S1a, a single layer of $\{100\}$ is shown where the Cu phase is covered by Ag layers (only the atomic columns corresponding to the Cu atoms are shown). The Hirth partial dislocations are indicated by the symbol *X*. Removing all the Ag layers except the immediate layer surrounding the Cu phase results in a perfect core-shell structure. We now notice that the dislocation arrangement remains the same with difference being that there is only a monolayer of Ag above the dislocation. In the Fig. S2b,

we a Ag-Cu nanoalloy is shown. Clearly, the arrangement close the symbol X resembles the arrangement of the Hirth dislocations shown in the Fig. S1a.

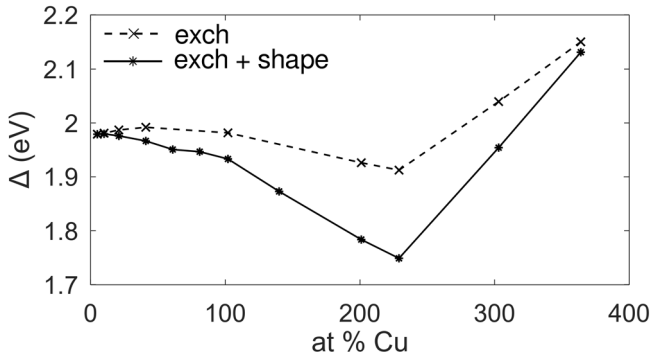

Fig. S 2. Comparison of the excess energy (Δ) of the lowest energy structures obtained from *exch* only BH and *exch + shape* BH searches at the size 405.

In the Fig. S2, a comparison of the excess energy obtained from the two methods, optimization of only the chemical ordering (*exch* only BH) and complete structural optimization (*exch + shape* BH), at the size 405 is shown. At all the compositions, the structures obtained from *exch + shape* BH have lower excess energy. The difference is maximum in the central region of the composition close to the perfect core-shell structure (with monolayer of Ag) and diminishes towards the Ag-rich and Cu-rich ends. This highlights the importance of complete structural optimization to capture the structural features.