Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Electronic Supporting Information for "Interplay between size, shape, and surface segregation in high-entropy nanoalloys"

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1 Pair correlation entropy versus inverse radius

Figs. S1 and S2 show the variations of the average pair correlation entropy $S_{\rm pc}$ obtained from the samples at 300 K of truncated octahedral, icosahedral, and decahedral nanoparticles from the AlCuFeCrNi and AuCuPdNiCo alloys, respectively, both at equiconcentration.

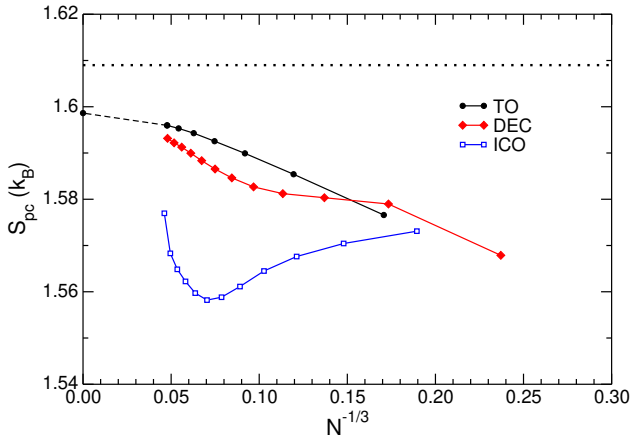

Figure S1: Pair correlation entropy $S_{\rm pc}$ of AlCuFeCrNi nanoalloys at equiconcentration and at 300 K, as a function of $N^{-1/3}$ and for truncated octahedral (TO), Marks' decahedral (DEC), and Mackay icosahedral (ICO) shapes.

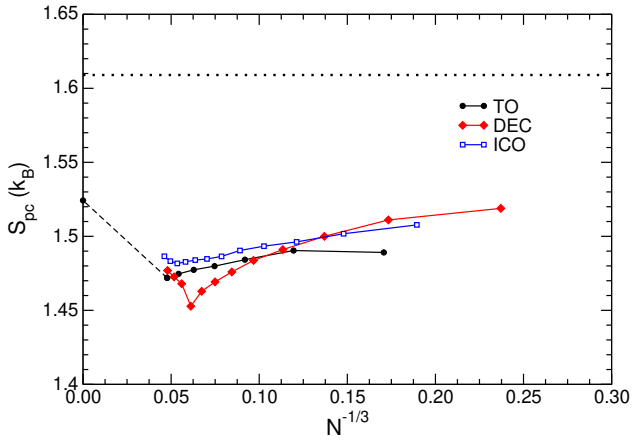

Figure S2: Pair correlation entropy $S_{\rm pc}$ of AuCuPdNiCo nanoalloys at equiconcentration and at 300 K, as a function of $N^{-1/3}$ and for truncated octahedral (TO), Marks' decahedral (DEC), and Mackay icosahedral (ICO) shapes.

2 Mixing energy versus pair correlation entropy

Figs. S3 shows the correlation between the mixing energy $U_{\rm mix}$ and the pair correlation entropy $S_{\rm pc}$ for the 2406-atom truncated octahedral AlCuFeCrNi nanoalloys at various concentrations varying by steps of 20%, and the predictions of a simple Gaussian regression model of concentrations that maximize $S_{\rm pc}$ or minimize $U_{\rm mix}$.

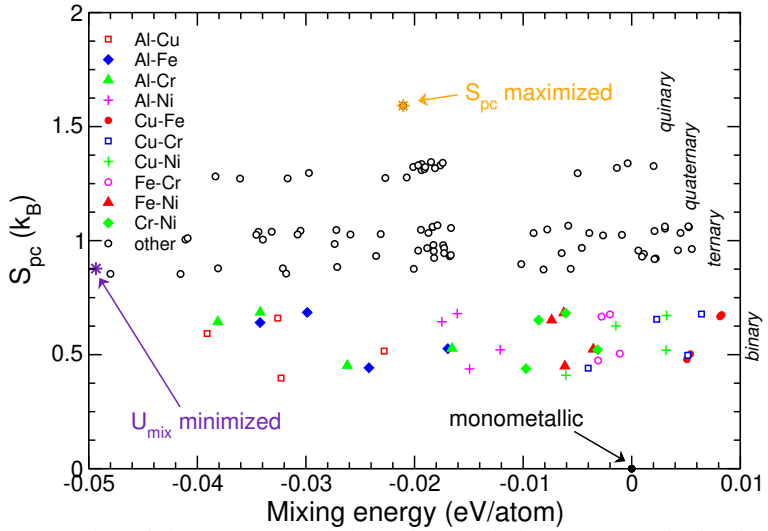

Figure S3: Correlation between the mixing energy and the pair correlation entropy obtained at 300 K for 2406-atom truncated octahedral AlCuFeCrNi nanoalloys with compositions varying by steps of 20%. The predictions of a simple Gaussian regression model maximizing the pair correlation entropy or minimizing the energy of mixing are highlighted.

Fig. S4 shows the corresponding data for the 2057-atom icosahedral AuCuPdNiCo nanoalloys.

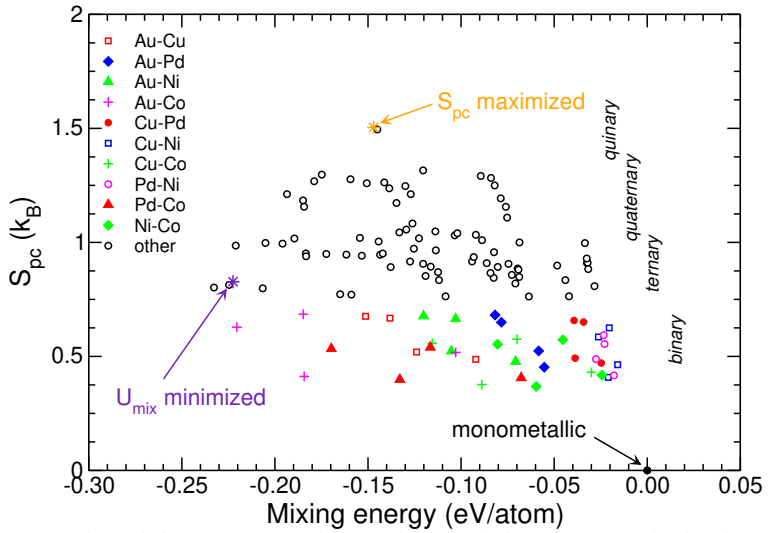

Figure S4: Correlation between the mixing energy and the pair correlation entropy obtained at 300 K for 2057-atom icosahedral AuCuPdNiCo nanoalloys with compositions varying by steps of 20%. The predictions of a simple Gaussian regression model maximizing the pair correlation entropy or minimizing the energy of mixing are highlighted.

For both systems, the pair correlation entropy is particularly sensitive to the number of chemical species present in the nanoparticle, and the values obtained for $S_{\rm pc}$ for binary, ternary, quaternary and quinary mixtures appear separated, especially for the AlCuFeCrNi system.