Potential of Mean Force for (4.5nm+chain) NCs

Figure S1: Potential of Mean Force plots for the larger NCs(4.5 nm), see Table 1. The minimum as predicted by OPM The temperature in units of energy is $T = T_m$. 

S1
Cascade Effect for Other Chain Lengths

Symmetric Nanocrystals

Figure S2: The internal energy of NC and different chain regions for the 3.8 nm (1.1k), (3k), (5k), (11.5k) and (20k) NCs at different distances.
Figure S3: Average configuration for the 3.8 nm (3k), (5k) and (11.5k) during the pre-cascade and post-cascade process. The APS ligand chains are divided into three parts, that is the inner one (green) closer to the core, the middle one (red) and the outer one (purple).
Asymmetric Nanocrystals

Figure S4: Average configuration for the 4.5 nm (1.1k)-3.8 nm (3k) and 4.5 nm (3k)-3.8 nm (1.1k) during the pre-cascade post-cascade process. The APS ligand chains are divided into three parts, that is the inner one (green) closer to the core, the middle one (red) and the outer one (purple)
Cascade Effect for SL Systems

Figure S5: The internal energy of NC and different chain regions for the 3.8 nm (1.1k), (3k), and (5k) superlattices at some lattice constants.
Figure S6: The internal energy of NC and different chain regions for the 3.8 nm (11.5k) and (100k) superlattices at some lattice constants.
Figure S7: Averaged snapshots at different lattice constant for bcc/fcc SLs. cont.
Figure S8: Averaged snapshots at different lattice constant for bcc/fcc SLs.
Lattice Free Energy for (4.5nm+chain) NCs

Figure S9: Free energy for bcc and fcc for 4.5nm(jk), j=1.1, 3.
The local density for the fcc SCSL

Figure S10: The local number density and the corresponding histogram along the nearest neighbor (1) and next nearest neighbor (2) in a radial cone with aperture angle (a) $\pi/2$ and (b) $\pi/4$
Additional Thermodynamic Functions in SLs

Figure S11: Free Energy, Internal Energy and Entropy comparison for bcc/fcc SLs. cont.
Figure S12: Free Energy, Internal Energy and Entropy comparison for bcc/fcc SLs.
Figure S13: Gibbs free energy and their difference for bcc/fcc SLs.