

# Non-equilibrium Ionic Assemblies of Oppositely Charged Nanoparticles

## Supplementary Information

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**This material contains four kinetic Monte Carlo simulation movies. General system conditions include  $\eta=0.004$ ,  $\sigma_+=\sigma_-=\sigma$ , and  $Z_+=-Z_- = Z$  (except for Movie S4). Red and Blue spheres represent positively and negatively charged NPs, respectively. All movies start from a random distribution of particles ( $t=0$ ) and end at the time  $t_{\text{end}}$ . Additional details for each movie are described below.**

Movie S1.

A typical simulation run for the  $\kappa\sigma=1$ ,  $Z^2 l_B/\sigma=81$  system ( $N_+=N_-=500$ ,  $t_{\text{end}}=1100\tau_0$ ). In this case, NPs self-assemble into crystalline NaCl-type clusters.

Movie S2.

A typical simulation run for the  $\kappa\sigma=1$ ,  $Z^2 l_B/\sigma=300$  system ( $N_+=N_-=250$ ,  $t_{\text{end}}=200\tau_0$ ). In this case, NPs self-assemble into ionic chains and fibril-like clusters.

Movie S3.

A typical simulation run for the  $\kappa\sigma=5$ ,  $Z^2 l_B/\sigma=160$  system ( $N_+=N_-=250$ ,  $t_{\text{end}}=500\tau_0$ ). In this case, NPs self-assemble into disordered compact clusters.

Movie S4.

A typical simulation run for a “charge-asymmetric” system at the Yukawa conditions  $Z_+=-Z_-/2=Z$ ,  $\kappa\sigma=1$ ,  $Z^2 l_B/\sigma=50$  ( $N_+=N_-=250$ ,  $t_{\text{end}}=1000\tau_0$ ). In this case, average cluster size attains a steady value  $\sim 7.8$  around  $t=700\tau_0$  and some clusters are in stable chain configuration.