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Coulombic Interactions in Nanoparticle Superlattices

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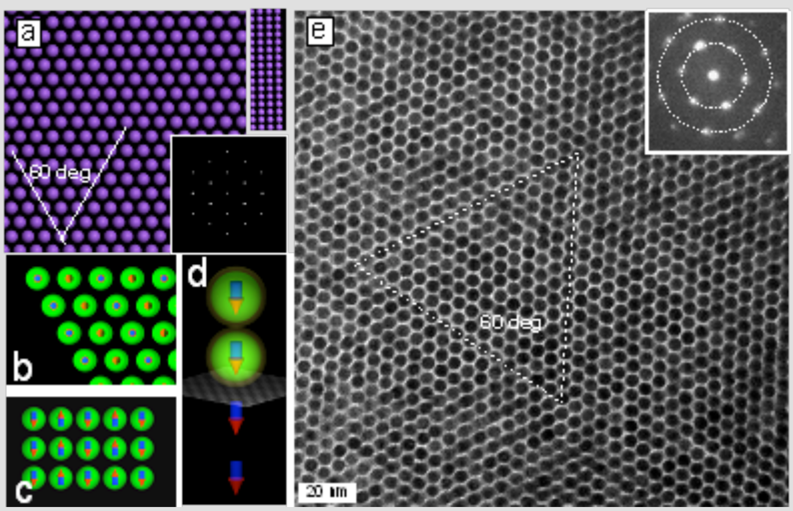

Figure. (a) Modelled [001] projection of *simple hexagonal* (*sh*) lattice. Right top inset shows side view of the superlattice, right bottom inset shows FFT of the lattice projection. **(b,c)** Calculations predict antiferroelectric dipole ordering in *sh* nanoparticle superlattices. **(d)** Interaction of dipolar nanoparticles with conductive substrate occurs through the image charges. **(e)** TEM image of 7.2 nm PbSe self-assembled nanocrystal superlattice. The inset shows small angle electron diffraction pattern collected from 6 μm^2 area of PbSe nanocrystal superlattice.

Self-assembly is the fundamental phenomenon which generates structural organization on all length scales. We studied self-assembly of monodisperse nanocrystals into long-range ordered superlattices with examples drawn from both semiconductor (CdSe, PbSe, PbS) and magnetic ($CoPt_3$, γ-Fe_2O_3) nanoparticles. The goal of the project was to provide a better understanding of parameters which govern self-assembly of nanoscale objects and could be used to engineer nanoparticle superlattices structure and properties. We found that sub-10nm nanoparticles often self-assemble into hexagonal-close-packed (*hcp*) structures predicted to be less stable than *fcc* packing in hard sphere models. In addition to close-packed *fcc* and *hcp* superlattices, we observed formation of non-close packed *simple-hexagonal* (*sh*) superlattices of nearly spherical PbS, PbSe and Fe_2O_3 nanocrystals. This surprisingly rich phase diagram of monodisperse semiconducting nanoparticles was explained by considering the interactions between non-local dipole moments of individual nanoparticles. By calculating the total electrostatic and dispersive energies for various superlattice structures, we explained the stability of *fcc*, *hcp* and *sh* nanoparticle superlattices, introduced the superlattice phase diagram and predict antiferroelectric ordering in dipolar nanoparticle superlattices.