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Counterion-Mediated Assembly of Spherical Nucleic Acid-Au Nanoparticle Conjugates (SNA-AuNPs) SUMIT KEWALRAMANI, LIANE MOREAU, GUILLERMO GUERRERO-GARCÍA, CHAD MIRKIN, MONICA OLVERA DE LA CRUZ, MICHAEL BEDZYK, Northwestern University, AFOSR MURI TEAM — Controlled crystallization of colloids from solution has been a goal of material scientists for decades. Recently, nucleic acid functionalized spherical Au nanoparticles (SNA-AuNPs) have been programmed to assemble in a wide variety of crystal structures. In this approach, the assembly is driven by Watson-Crick hybridization between DNAs coating the AuNPs. Here, we show that counterions can induce ordered assembly of SNA-AuNPs in bulk solutions, even in the absence of base pairing interactions. The electrostatics-driven assembly of spherical nucleic acid-Au nanoparticle conjugates (SNA-AuNPs) is probed as a function of counterion concentration and counterion valency [+1 (Na^+) or +2 (Ca^{2+})] by *in situ* solution X-ray scattering. Assemblies of AuNPs capped with single-stranded (ss-) or double-stranded (ds-) DNA are examined. SAXS reveals disordered (gas-like) $\rightarrow$ face-centered-cubic (FCC) $\rightarrow$ glass-like phase transitions with increasing solution ionic strength. These studies demonstrate how non-base-pairing interactions can be tuned to create crystalline assemblies of SNA-AuNPs. The dependence of the inter-SNA-AuNP interactions on counterion valency and stiffness of the DNA corona will be discussed.

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