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Nanostructured Gold Architectures Formed through High Pressure-Driven Sintering of Spherical Nanoparticle Arrays

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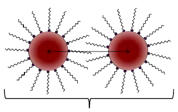
Introduction

Due to the size- and shape-dependent properties, nanoparticles have been successfully used as functional building blocks to fabricate multi-dimensional (D) ordered assemblies for the development of 'artificial solids' with potential applications in nanoelectronic and optic devices.

•Nanoparticle assemblies have been limited to entropy (and/or enthalpy) driven processes in which specific interparticle chemical or physical interactions such as van der Waals interaction, dipole-dipole interaction, chemical reactions, and DNA-templating are required

•We developed the Pressure-Directed Assembly (PDA) method, as an artificial tool, to emulate natural folding and unfolding processes in self-assembled DNA systems to explore energy landscaping that govern local interactions (hydrogen bonding, particle-particle interaction, etc), to design new classes of active materials with structure and function that are not attainable for current materials, and to investigate new property resulted from the folding and unfolding processes.

Nanoparticle Assembly at Ambient Pressure



Balanced interparticle interactions:
•Charge interactions
• Dipole-dipole interactions
• Van der Waals interactions
• Interparticle attractions
• chain-chain interactions
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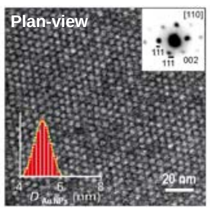
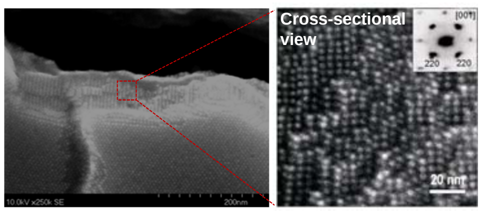
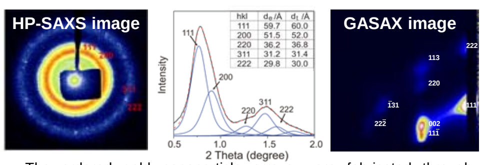
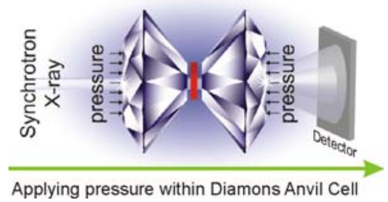




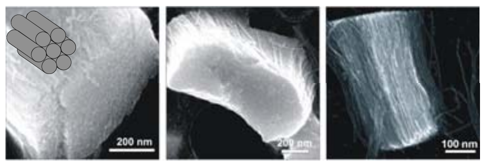


- The ordered gold nanoparticle arrays were fabricated through solvent evaporation.
- Results from SEM images, XRD spectrum and GISAX patterns indicate the nanoparticle films have standard face-centered cubic (*fcc*) structure (*Fm-3m*) with the unit cell parameter a_{fcc} = 10.4 nm.

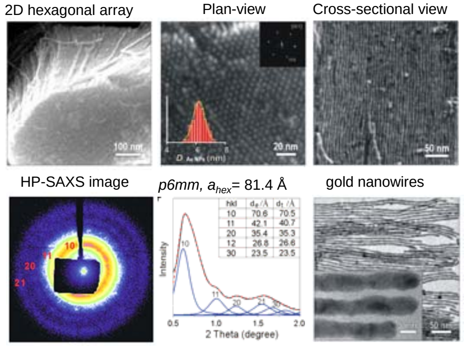
Nanoparticle Assembly under Pressure



Pressure-driven formation of gold nanowire arrays

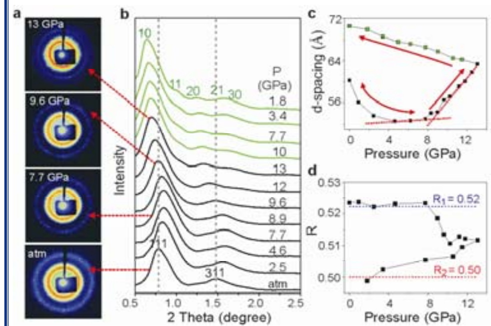


Characterization of gold nanowire arrays

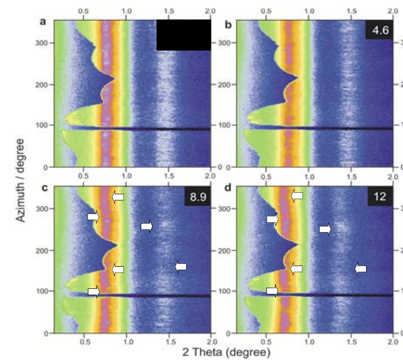


- After the pressure is released from 13 GPa to ambient pressure at room temperature, the ordered *fcc* spherical nanoparticle arrays ⇒ ordered 2D nanowire arrays. The gold nanowire array consists of individual gold nanowires with hexagonal close packing with a unit cell with lattice parameter a_{hex} = 81.4 Å.
- Single nanowires can be obtained by dispersion of the nanowire arrays in organic solvents. These nanowires are uniform in diameter and length. The length depends on the initial nanoparticle film thickness, ranging from 400 nm to 1.5 µm.
- The High-resolution TEM image reveals that the gold nanowires are polycrystalline. Along the *c*-axis, each nanowire consists of crystalline nanodomains whose size is close to that of original spherical gold nanoparticles. This indicates that the formation of gold nanowires occurs through sintering of spherical gold nanoparticles to form nanowire *c*-axis.

Pressure-driven nanoparticle assembly



Structural evolution of gold nanoparticle assemblies during compression and release. (a) Representative HP-SAXS patterns of gold nanoparticle assemblies collected at ambient pressure, 7.7 GPa, 9.6 GPa, and 13 GPa. (b) The integrated spectra from the HP-SAXS patterns at varied pressures during compression and release. For convenient comparison, the SAXS pattern at ambient pressure is shown again. (c) The d-spacing changes along with the pressure during compression and release. The d-spacing of the first Bragg reflection in each HP-SAXS spectrum is used. (d) d-spacing ratios (R) at different pressures.

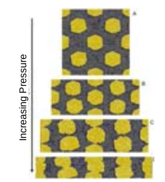


Cartesian (cake) plots of azimuth angle versus 2-theta angle for the gold nanoparticle assembly at different pressures: (a) ambient pressure, (b) 4.6 GPa, (c) 8.9 GPa, and (d) 12 GPa. For the pressures < 8.9 GPa, there is no waviness, but > 8.9 GPa, waviness appears.

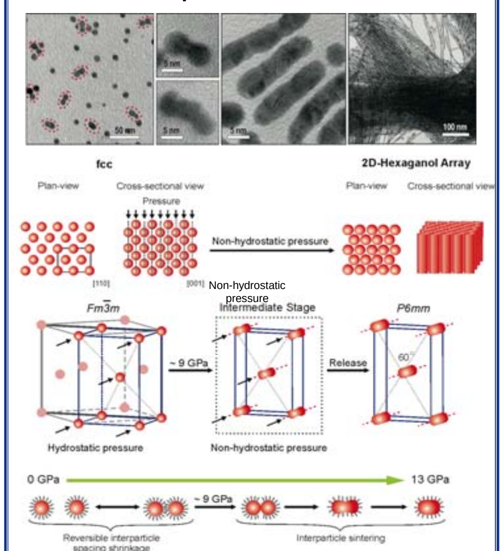
- Phase transformation of the nanoparticle assembly starts at 8.9 GPa via an intermediate stage in the pressure course of 8.9 GPa → 13.0 Gpa → 7.7 GPa.
- P < 8.9 GPa, the nanoparticle assembly has *fcc* structure and *d*-spacing ratios $R = d_{311}/d_{111}$ =0.52; release P < 7.7 GPa, *R* becomes ~0.5, the theoretical ratio d_{200}/d_{100} for a 2D hexagonal mesophase.

Proposed Mechanism

Initial simulations indicate that the existence of a high pressure gradient, combined with uniaxial elastic deformation, makes the folding/unfolding processes both thermodynamically and kinetically possible.



Proposed Mechanism



A schematic model shows the orientated sintering of gold NPs into NW arrays along [110]-direction under compression.

Conclusions

We demonstrate that the mechanical compression process under pressure provides a new route to engineer nanoparticle architectures and to fabricate new nanostructured materials. The reversible changes of the nanoparticle unit cell dimension under pressure allow precise control over interparticle separation in 2D or 3D nanoparticle assemblies, offering unique robustness for interrogation of both quantum and classic coupling interaction2. The fundamental understanding of nanoparticle assembly under pressure provides useful insight for material integration through pressure-driven nanofabrication processes, such as the embossing process - a key process for fabrication of micro/nano- optical and electronic devices.

Acknowledgement

Sandia National Laboratories

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