Sintering Rate and Crystal Structure of Gold Nanoparticles by Molecular Dynamics

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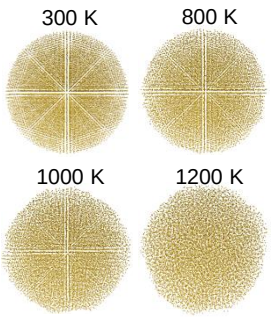
Motivation

Gold nanoparticles find applications in catalysis, target-specific drug delivery, nanolithography and electronics[1]. The rates of sintering and crystalization of small nanoparticles (<10 nm) are significantly different from those of the corresponding bulk materials.

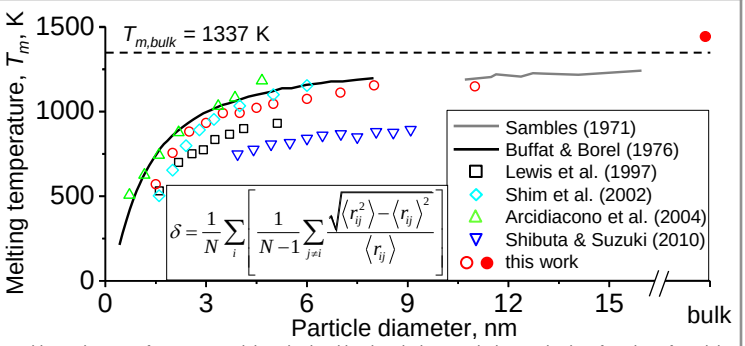
Here, detailed atomistic Molecular Dynamics (MD) simulations are used to systematically investigate the sintering up to full coalescence of gold nanoparticles of 2 – 4 nm initial diameter at 227 – 727 °C.

The crystallinity dynamics during sintering are investigated, as along with particle size affect the final product properties (e.g. morphology and structure) and eventually nano-Au performance.

Size-dependent Melting Temperature, T_m

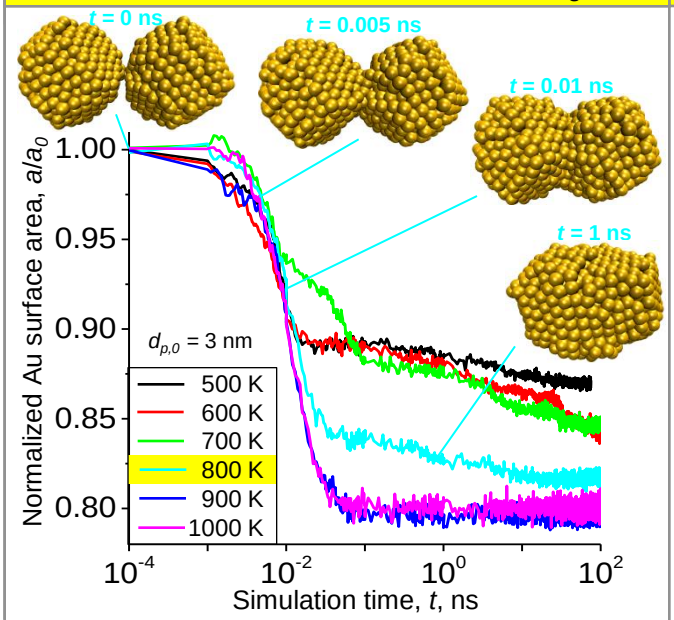


Increasing T causes gradual loss of the Au fcc crystal structure from the surface moving inwards.

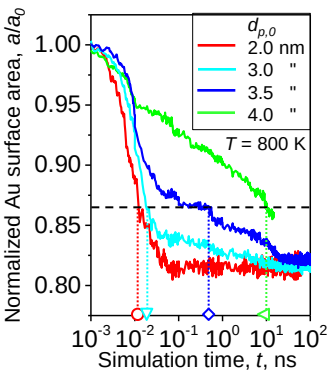


Melting point, T_m, of Au nanoparticles obtained by the Lindemann index method as function of particle diameter, d_p. This work (circles) is compared to MD simulations (open symbols) and experiments (lines).

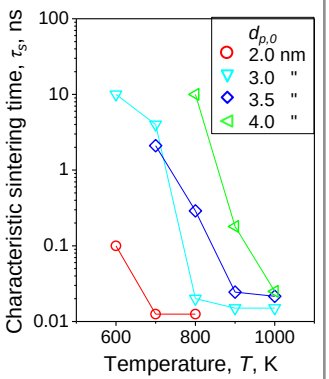
Characteristic Sintering Time, τ_s



Normalized surface area by sintering of two gold nanoparticles with initial diameter, $d_{p,0}$, of 3 nm. Snapshots of the coalescing particles are shown at t = 0, 0.01 and 1 ns for T = 800 K.

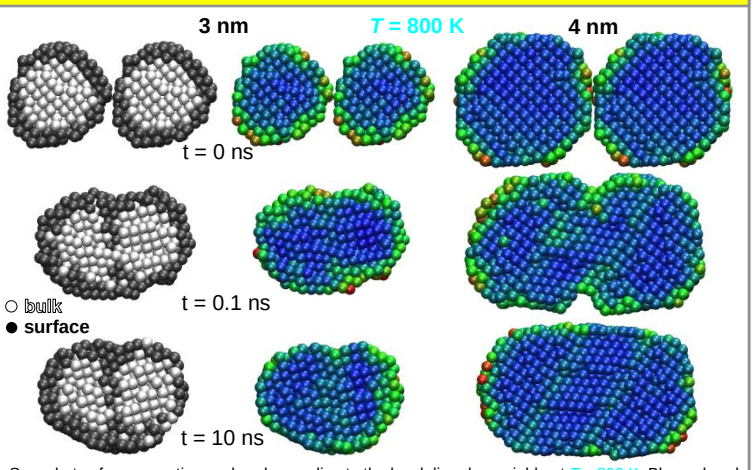


Normalized surface area by sintering of two Au nanoparticles at T = 800 K.

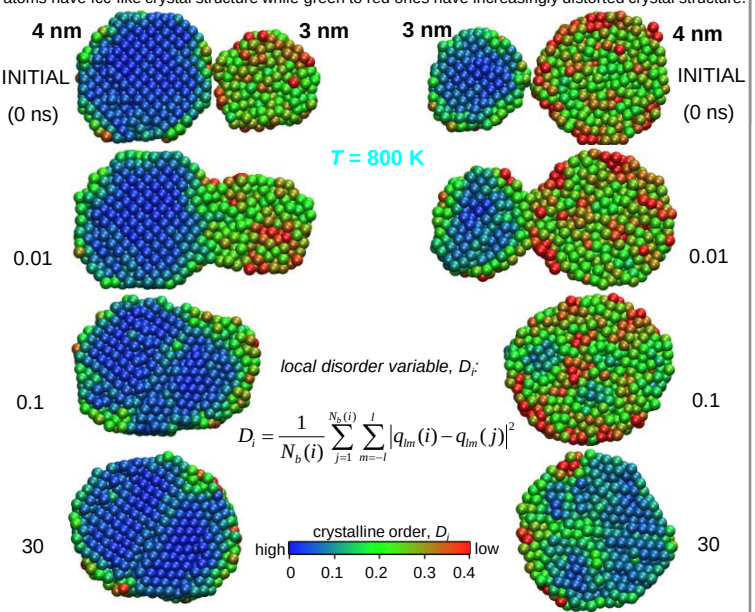


Characteristic sintering time by MD simulations as a function of temperature.

Crystallinity Dynamics during Sintering



Snapshots of cross-sections colored according to the local disorder variable at T = 800 K. Blue-colored atoms have fcc-like crystal structure while green to red ones have increasingly distorted crystal structure.



Snapshots of cross-sections of Au nanoparticles with $d_{p,0}$ = 3 and 4 nm of different initial crystallinity.

References

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Conclusions

1. The MD-obtained melting point of nano-sized Au particles is in excellent agreement with experiments and theory validating the present analysis.
2. The characteristic sintering time, τ_s, is quantified for particle diameters, $d_{p,0}$ = 2 – 4 nm by tracing the evolution of the particles' surface area. Increasing temperature and decreasing particle size results in faster particle coalescence.
3. The degree of disorder is quantified by the disorder variable. Two-equal-particle sintering leads to enhanced disorder of the surface atoms during particle adhesion and finally to formation of polycrystalline grains.
4. Initial crystalline structure affects crystallinity dynamics and grain formation of final product particles.