

Crystallization Onset of Aerosol Au Nanoparticles

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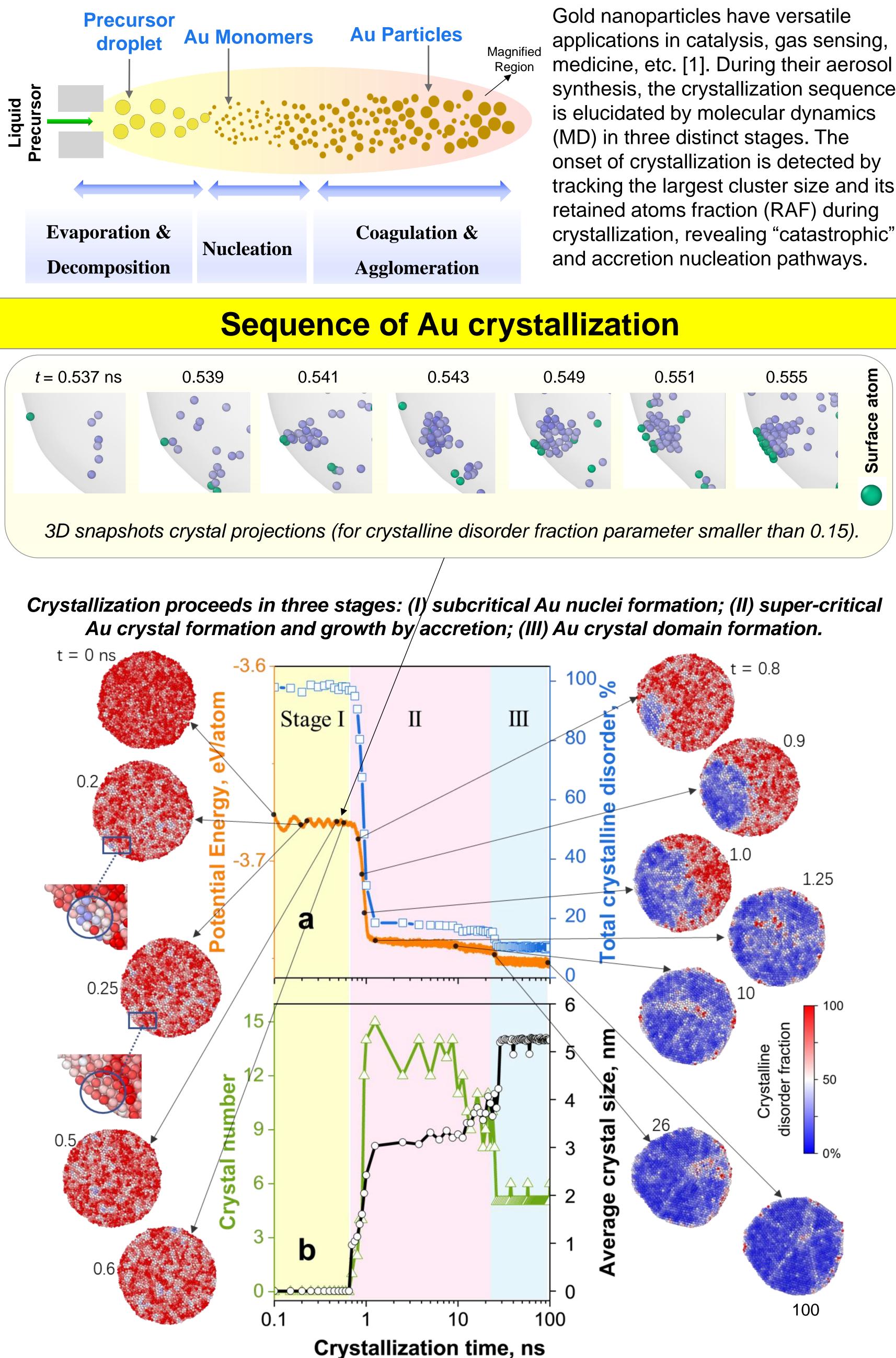
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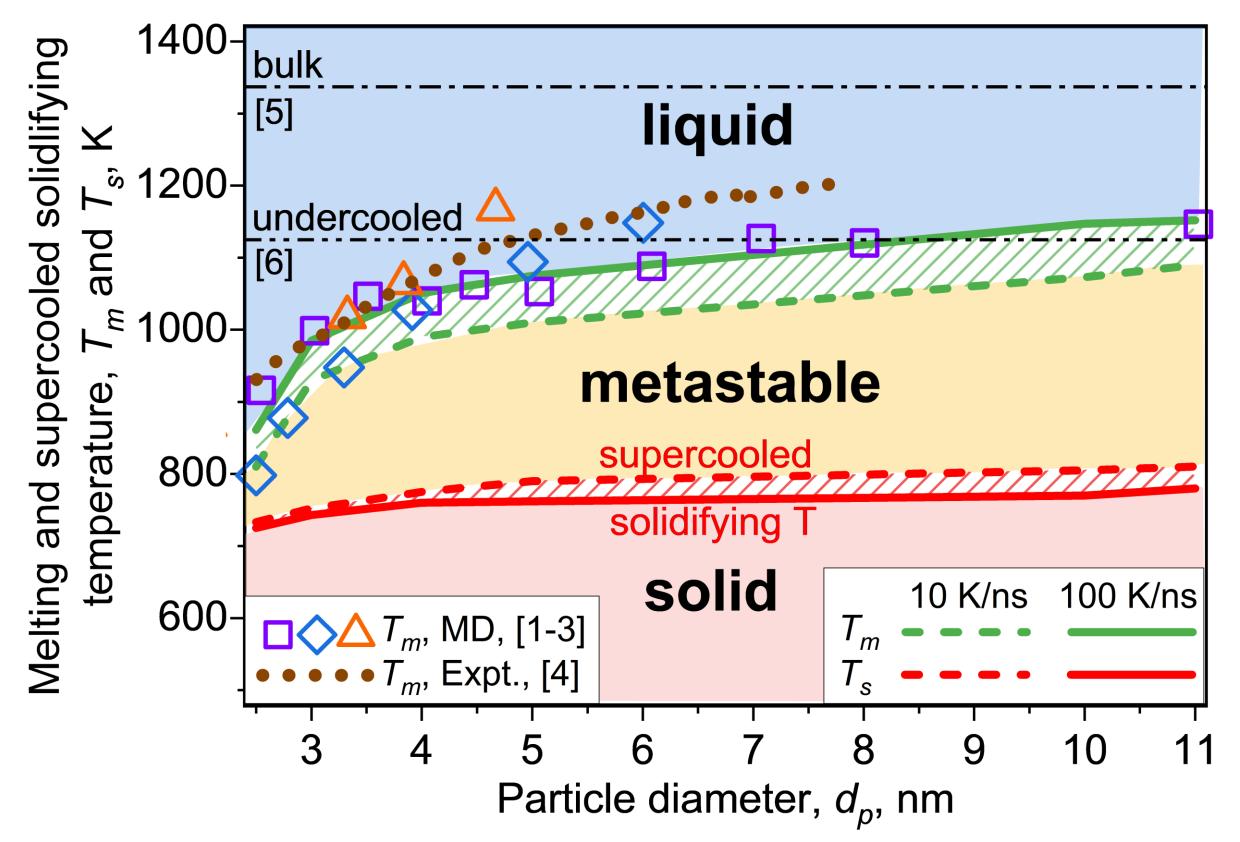


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Motivation



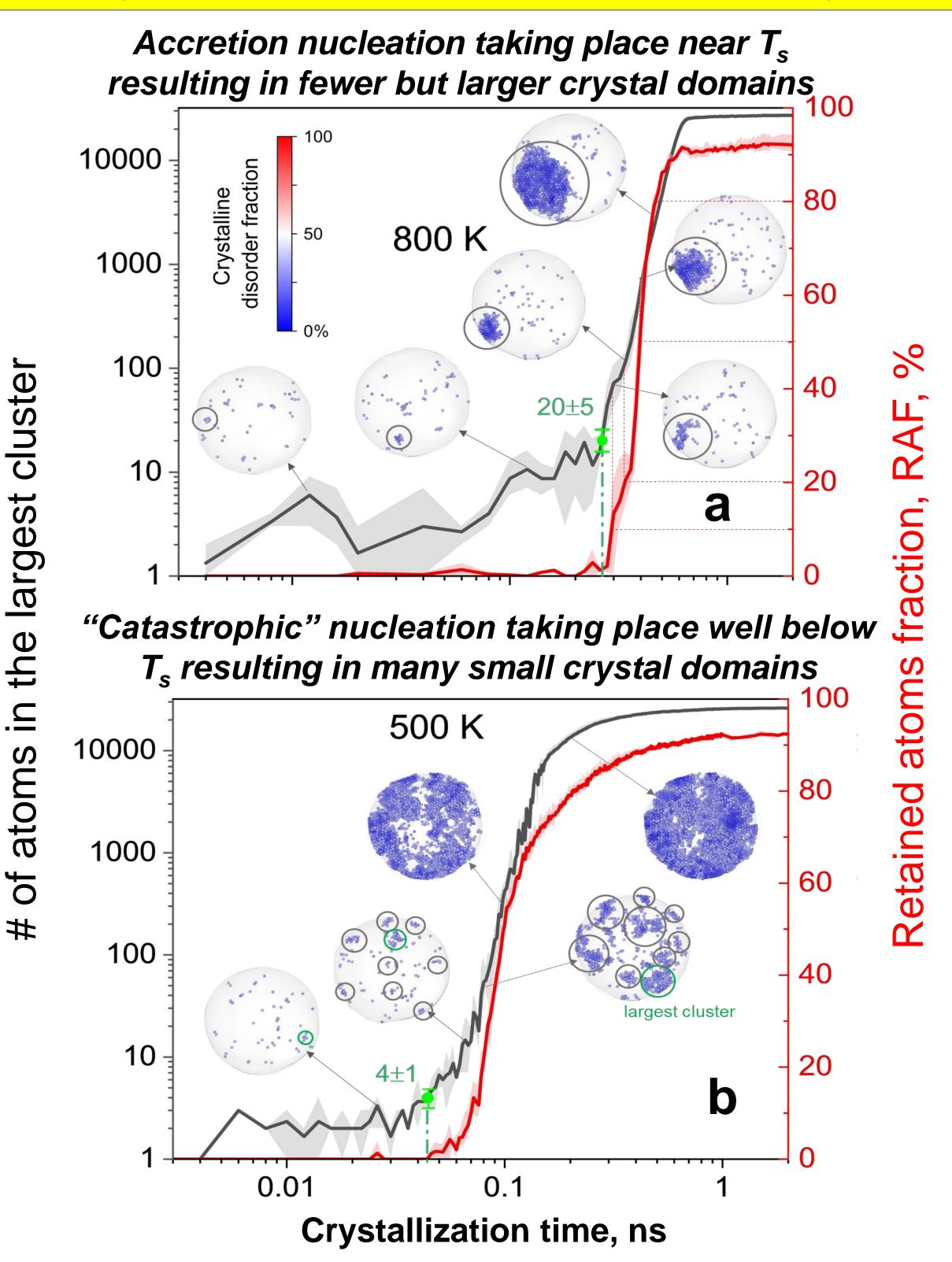
applications in catalysis, gas sensing, medicine, etc. [1]. During their aerosol synthesis, the crystallization sequence tracking the largest cluster size and its



Method validation

Melting point (T_m) & supercooled solidifying temperature (T_s) of Au as a function of particle diameter with the regions for liquid (blue), metastable (yellow) & solid (red) phases.

Crystallization nucleation pathways



Evolution of (a) average atomic potential energy and total crystalline disorder fraction as well as its (b) number of crystals and average crystal size along with cross-section snapshots colored by the local crystalline disorder fraction [7] during crystallization of a 10 nm Au particle at 800 K for 100 ns.

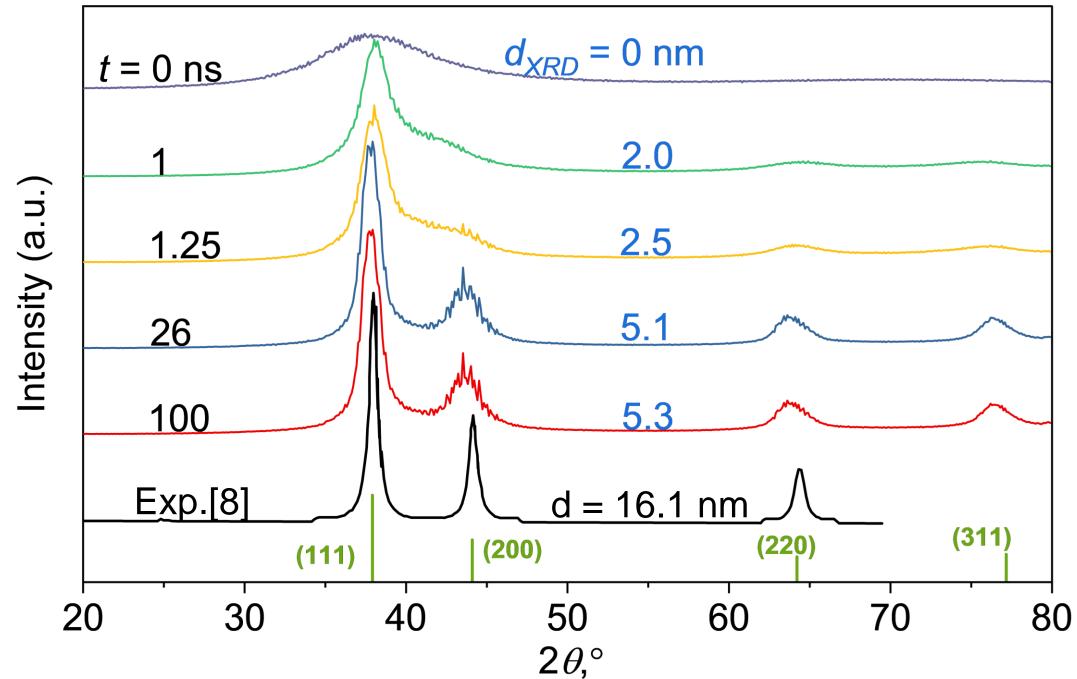
Number of atoms in the largest cluster and its retained atoms fraction of a 10 nm Au particle with 3D crystal projection at (a) 800 and (b) 500 K.

Crystal size evolution

Conclusions







Evolution of XRD patterns produced by MD of a 10 nm Au nanoparticle during crystallization at T = 800 K and crystal size, d_{XRD} , compared with the experimental XRD pattern for Au [8] (black line) with crystal size of 16.1 nm.

1. A size-dependent metastable region is revealed spanning between the melting point and supercooled solidifying temperature, T_s , which contracts as the cooling/heating rate decreases.

Isothermal crystallization progresses through (I) subcritical Au nuclei formation, (II) super-critical Au crystal formation and growth by accretion and (III) Au crystal domain formation.

- 3. Crystallization onset takes place by (A) "catastrophic" nucleation well below T_s and (B) accretion nucleation near T_s .
- 4. The initiation of nucleation is through critical crystal developing within at least one layer below the surface and then quickly expanding to the rest of the particle.

- Goudeli, E., Pratsinis, S. E. (2016) [1] AIChE J. 62, 589.
- Nakaso, K., Shimada, M., Okuyama, K., [2] Deppert, K. (2002) J. Aerosol Sci. 33, 1061
- Arcidiacono S., Bieri N.R., Poulikakos, [3] D., Grigoropoulos, C. P. (2004) Int. J. Multiph. Flow. **30**, 979.
- Buffat, P., and Borel, J. P. (1976) Phys. [4] *Rev. A.* **13**, 2287.
- Font, F., Myers, T. G. (2013) *J. Nanopart.* [5] *Res.* **15**, 2086.
- Wilde G, Sebright JL, Perepezko, JH [6] (2006) Acta. Mater. 54, 4759.
- Kawasaki, T., Onuki, A. (2011) J. Chem. [7] *Phys.* **135**, 174109.
- Mädler, L., Stark, W.J., Pratsinis, S.E. [8] (2003) J. Mater. Res. 18, 115.