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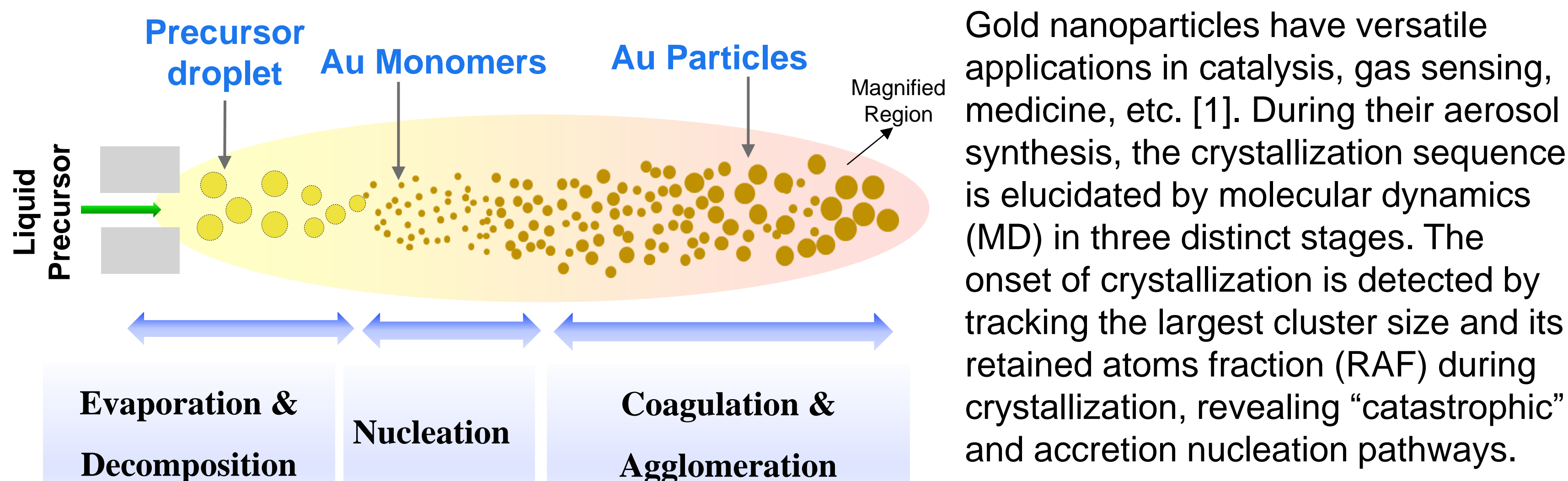
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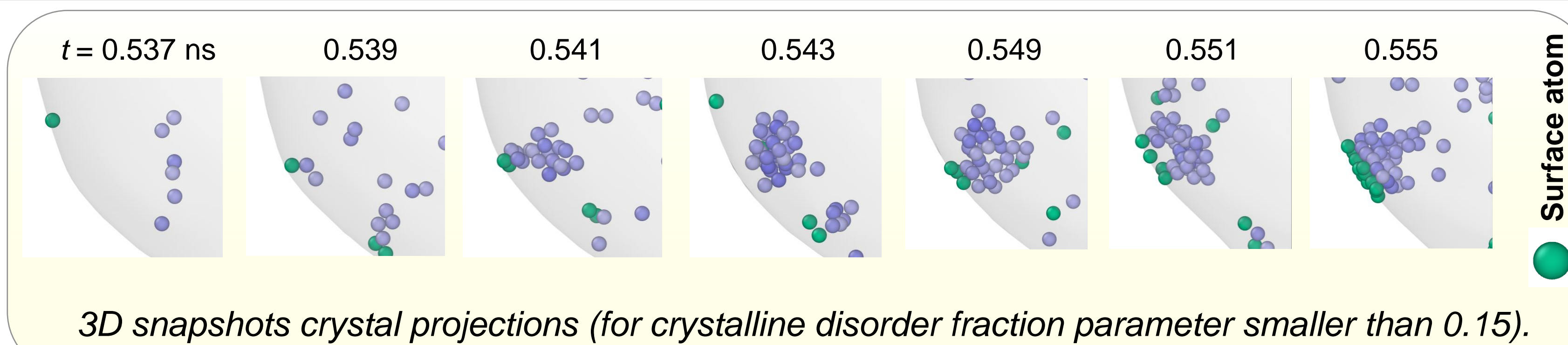
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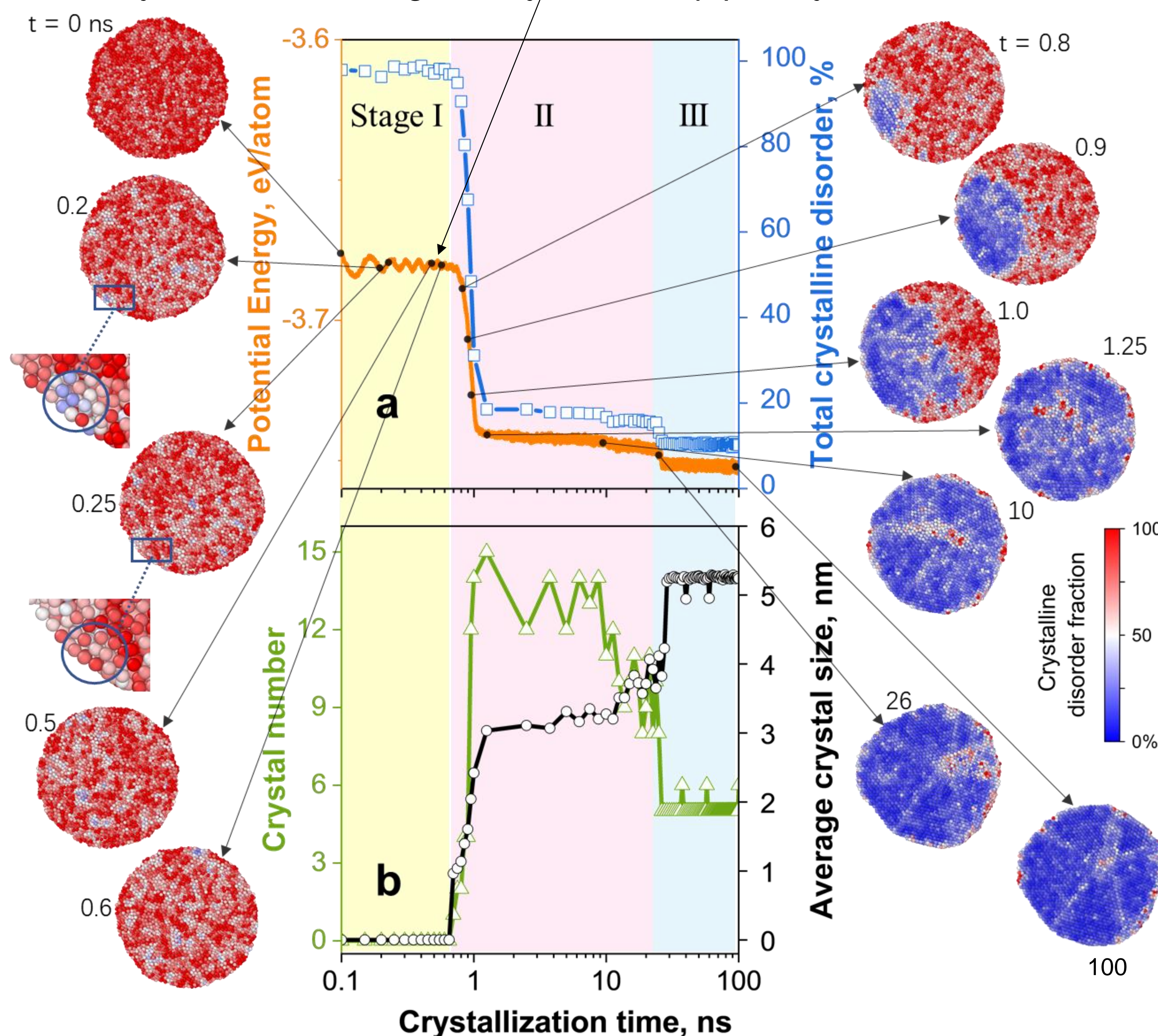
## Motivation



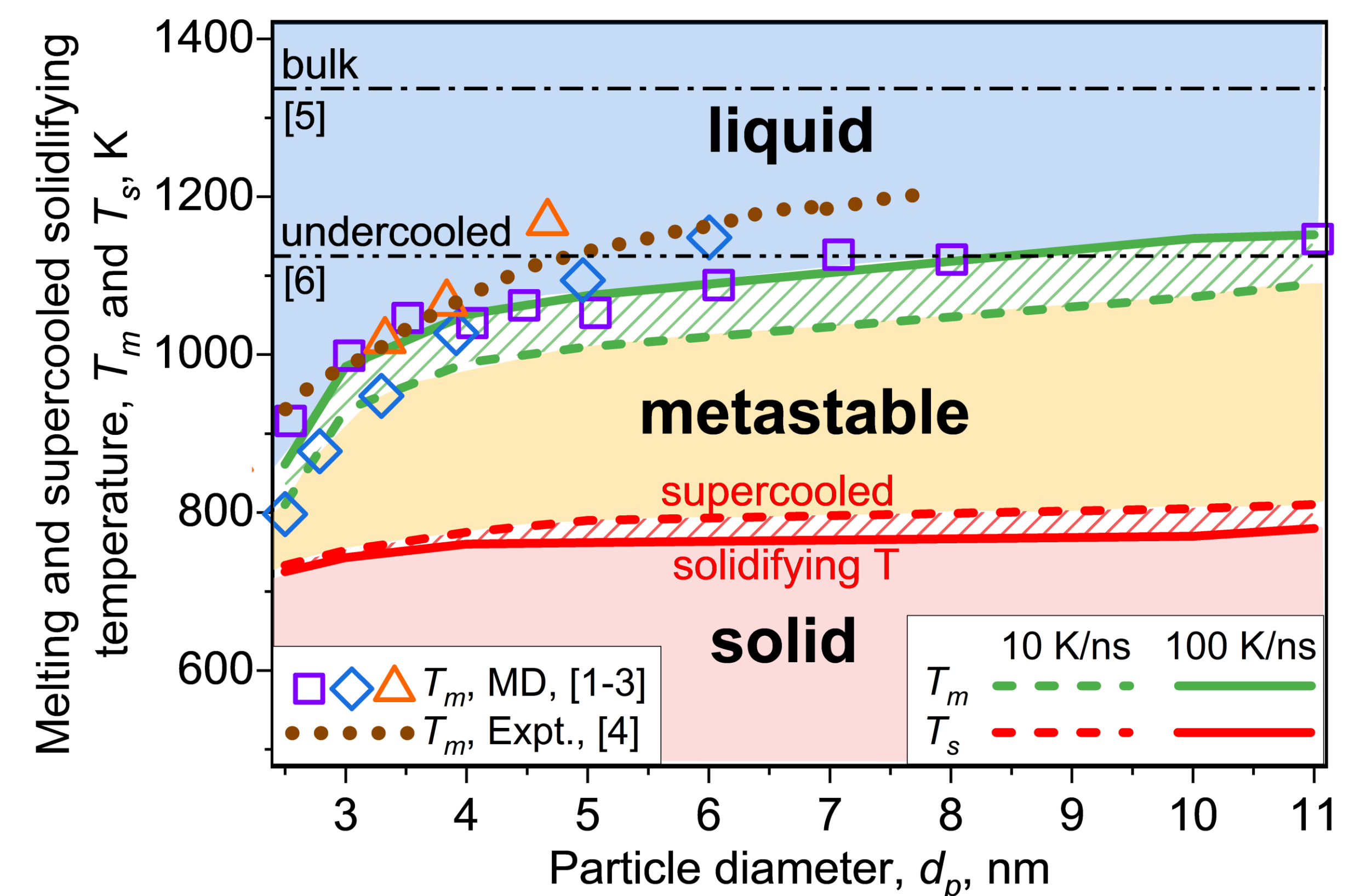
## Sequence of Au crystallization



Crystallization proceeds in three stages: (I) subcritical Au nuclei formation; (II) super-critical Au crystal formation and growth by accretion; (III) Au crystal domain formation.

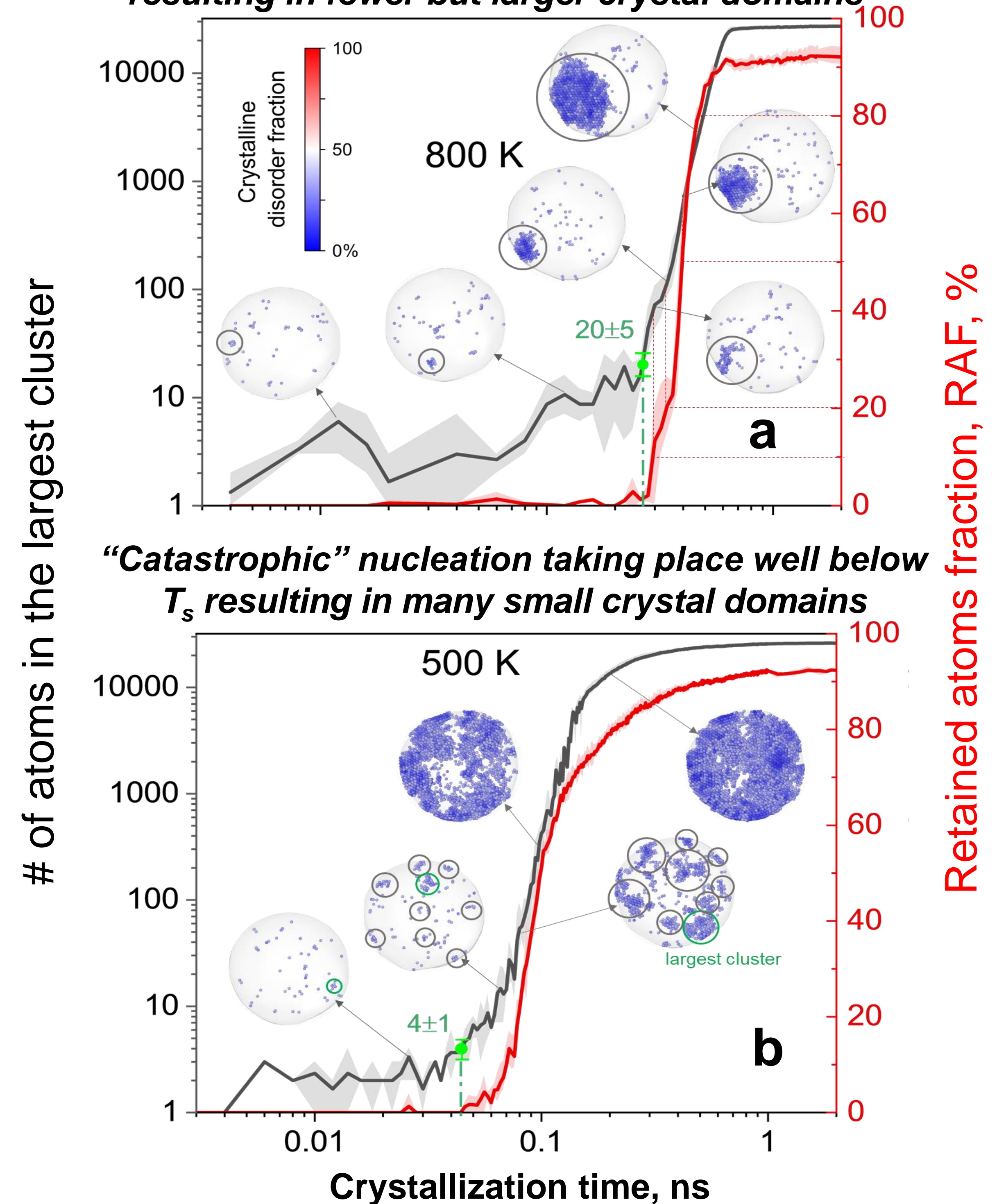


## Method validation

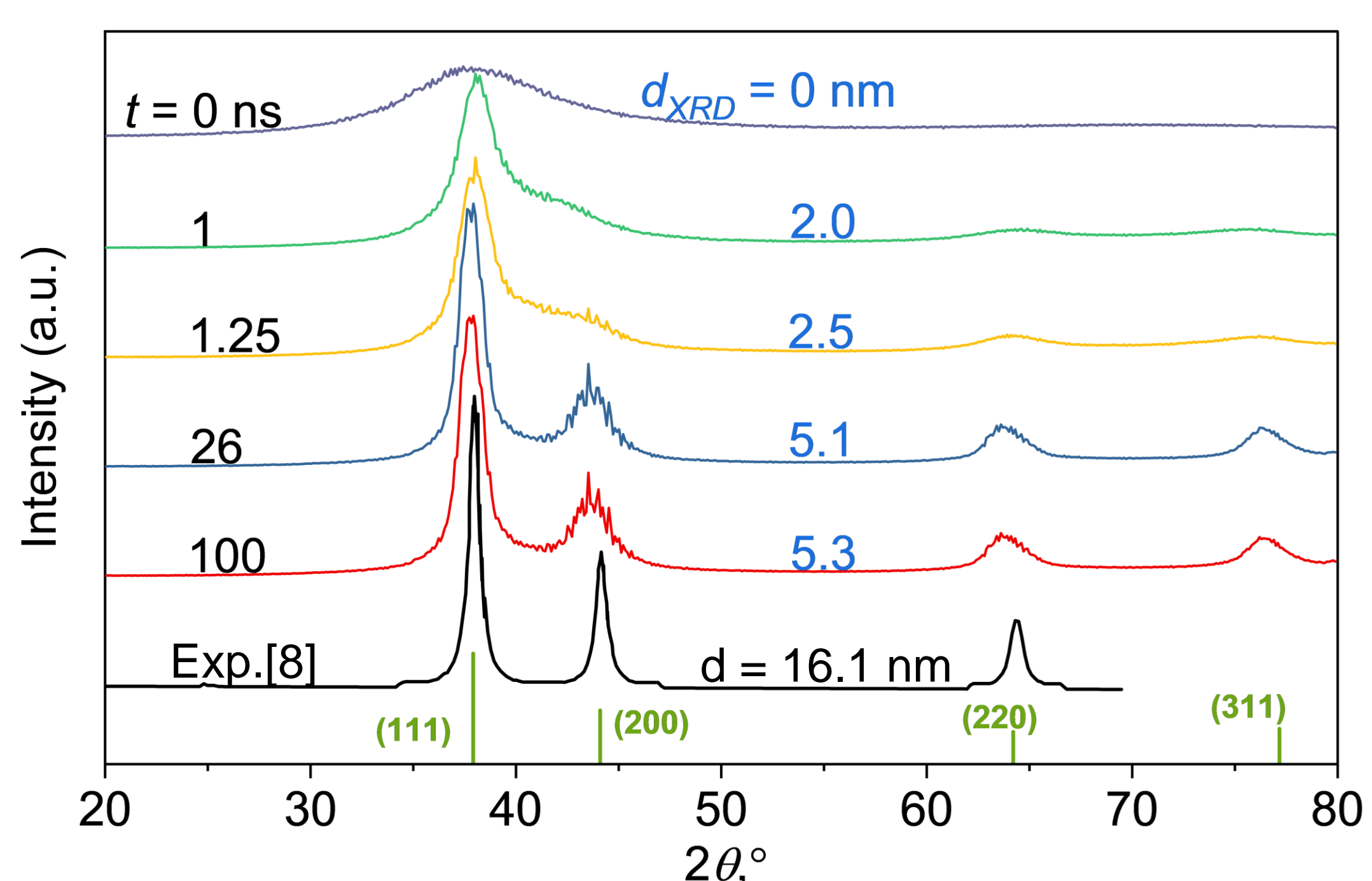


## Crystallization nucleation pathways

Accretion nucleation taking place near  $T_s$  resulting in fewer but larger crystal domains



## Crystal size evolution



## Conclusions

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.
2. 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.

## References

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