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► To cite this version:

Jens Baumgartner, Alexy Freitas, Blaise Fleury, Raj-Kumar Ramamoorthy, Marie-Alexandra Neouze, et al.. Nucleation of nanocrystals in solution: confinement by the amorphous networks. Nano2018, Jun 2018, Hong Kong, China. cea-02340795

HAL Id: cea-02340795

<https://hal-cea.archives-ouvertes.fr/cea-02340795>

Submitted on 31 Oct 2019

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Nucleation of nanocrystals in solution: confinement by the amorphous networks

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Key Words: Suprananostructure, Nanotwins, Nano-Precipitation, Energy

Recent advances show that nucleation of crystalline phases from dilute solutions often involves the formation of intermediate disordered states: clusters, droplets, amorphous nanoparticles, polymorphs etc [1]. It remains unclear how the intermediate states affect the predictions of the classical single-step nucleation theory, in terms of activation barrier, rate of nucleation, or critical cluster sizes. Progress is hampered by the difficulty to reach sufficient resolutions ($<1\text{nm}$, $\ll 1\text{s}$), without perturbing the metastable process.

Here, we overcome this issue using in situ x-ray scattering, and report nucleation rates upon formation of different nanocrystals in solution (luminescent $\text{YVO}_4\text{:Eu}$, magnetite Fe_3O_4), where an intermediate nanostructured amorphous network is evidenced [2,3]. We find how the amorphous precursor impedes nucleation at the nanometer scale, and limits the size of the nanocrystals. The clarification of the role of the intermediate nanostructure clarifies why the predicted nanocrystal sizes are overestimated by several orders of magnitudes by classical theories.

Reference:

- [1] J. J. D. Yoreo *et al.*, Crystallization by particle attachment in synthetic, biogenic, and geologic environments. *Science*. **349**, aaa6760 (2015).
- [2] B. Fleury *et al.*, Amorphous to Crystal Conversion as a Mechanism Governing the Structure of Luminescent $\text{YVO}_4\text{:Eu}$ Nanoparticles. *ACS Nano*. **8**, 2602–2608 (2014).
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