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Quantitative Study of Temperature Effects on The Nucleation and Growth of Gold Nanocrystals in Water

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Temperature is a crucial parameter in the liquid-phase synthesis of metal nanoparticles (NPs) that directly impacts all the atomic-scale processes that drive the size dispersion of colloidal assemblies and the shape of nanostructures [1-2]. As the temperature concomitantly affects the kinetics of chemical reactions and the thermodynamic equilibrium of nanomaterials in solution, the full understanding of thermal effects on the nucleation and growth processes requires direct in situ observations at the nanoscale.

Here, we exploit for the first time temperature controlled liquid-cell TEM to study thermal effects on the radiolysis-driven formation of gold nanocrystals in water between 25 °C and 85 °C (Figure 1) [3]. The huge impacts of temperature on the nucleation and growth rates of nanostructures measured using automated video processing are quantitatively explained in the framework of the classical theories. Thus, we show that the increase of molecular diffusion and nanoparticle solubility governs the drastic changes in the formation dynamics of nanostructures in solution with temperature. In contradiction with the common view of coarsening processes in solution, we also demonstrate that the dissolution of nanoparticles and thus the Ostwald ripening are not only driven by size effects (Figure 2). Furthermore, visualizing thermal effects on faceting processes at the single nanoparticle level reveals how the competition between the growth speed and the surface diffusion dictates the final shape of nanocrystals. Our method and data-analysis workflow can be applied to other dynamical processes in nanochemistry where temperature plays an important role [5].

References:

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[5] We gratefully acknowledge the help of Maxime Moreaud in the processing of our videos using the PlugIM plateform developed at IFPEN.

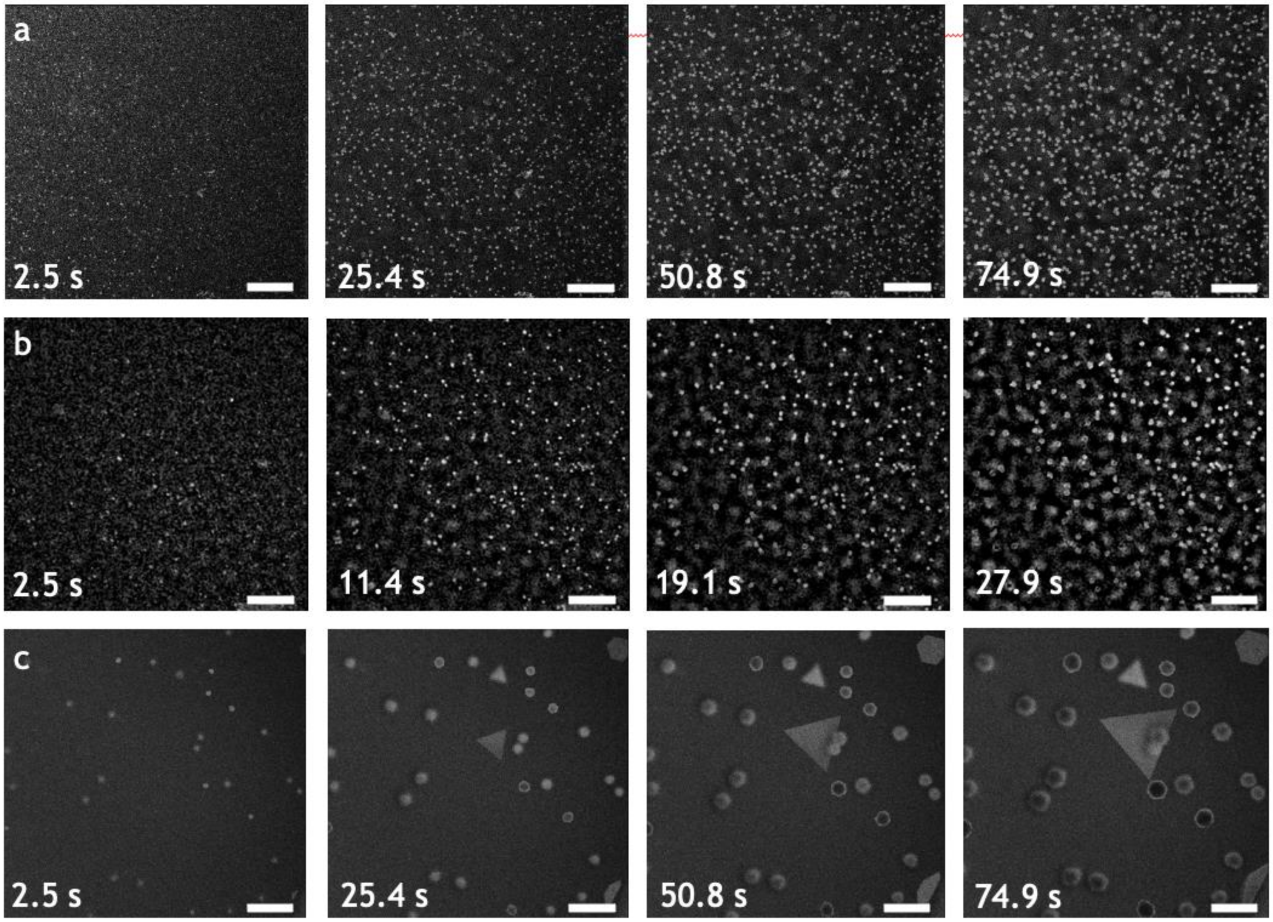

Figure 1. Low magnification STEM HAADF image series of the radiolysis-driven formation of gold nanoparticles in water at (a) 25 °C, (b) 50 °C and (c) 85 °C. The acquisition time is indicated in the bottom left corner of each image. All scale bars correspond to 1 µm. The three image series were acquired with continuous electron dose rate of 3.4 electrons.nm^{-2}.s^{-1} (adapted with permission from ref [4], copyright 2021 Wiley).

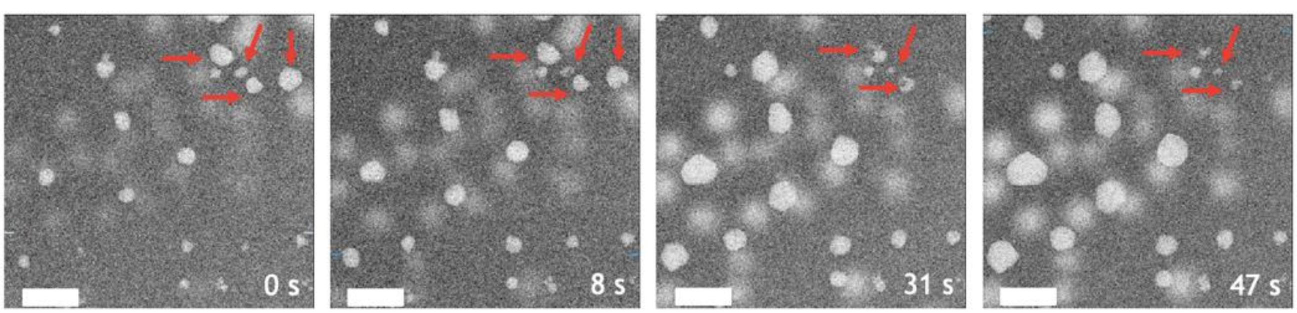

Figure 2. High-magnification STEM HAADF image series at 85 °C showing Ostwald ripening process during the growth of gold nanoparticles. The red arrows indicate the nanoparticles that shrink. The acquisition time is indicated in the bottom right corner of each image. All scale bars correspond to 250 nm. Image series acquired with continuous electron dose rate of 20.9 electrons.nm^{-2}.s^{-1} (adapted with permission from ref [4], copyright 2021 Wiley).