Controlling the Reaction Process in *Operando* STEM by Pixel Sub-Sampling

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Recently there has been an increase in the number of experiments making use of either in-situ gas or liquid stages, or using dedicated environmental (scanning) transmission electron microscopes (S/TEM) to study dynamic materials processes. While in-situ observations have traditionally been performed in TEM mode, allowing the intrinsic increase in temporal resolution of the projection image to be utilized, there are a number of key advantages of using the STEM imaging mode for these experiments. Namely, the same physics that makes high angle annular dark field (HAADF) or Z-contrast imaging optimum for quantifying small metal/oxide catalyst particles on a support also makes it optimum for imaging particle dynamics in liquids (now the liquid is the background in the image rather than the support). In addition, the incoherence of the Z-contrast image (i.e. decreased sensitivity to thickness effects) makes it the ideal method to image through the ~100-500 nm thick in-situ liquid cells that are typically used. Somewhat counter intuitively, the STEM imaging process is also optimized for controlling and reducing beam damage – the dose is controlled by the beam size and dwell time of the scan, while the probe only illuminates a small area, thereby reducing heating and depletion effects [1].

The use of the scanned beam to form images has another advantage in that it can readily make use of compressive sensing/in-painting approaches [2,3] to reduce the overall dose during the experiment and increase the acquisition speed [4,5]. The in-painting method is a mechanism by which a small sub-set of pixels can be acquired experimentally and then mathematical processes used to in-paint the missing information. Demonstration of this approach in practical acquisition of images [5] indicates that full reconstruction can be obtained for sub-sampling down to the ~10% level. In addition to reconstructing the images, analytics to quantify the content of the images has also been shown to work for extremely low sampling rates [6]. Such results suggest that an extra degree of flexibility in performing in-situ liquid or gas experiments can be built in by changing the level of sampling, i.e changing the density of dosed pixels used to acquire the images.

Figure 1 shows the final fully sampled images obtained after in-situ liquid nucleation and growth experiments were performed on a 10 mM AgNO\(_3\) solution sampled in full mode (Fig 1A) and 12% sub-sampled mode (Fig. 1B). What can be clearly seen from these results that the sub-pixel sampling leads to different growth morphology of silver nanoparticles. In this presentation, a discussion on how sub-sampling affects the growth mode for this and other in-situ liquid experiments will be discussed in detail [7].
References:
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Figure 1. HAADF STEM image of Ag nanoparticles acquired in (A) full mode and (B) 12% sub-sampled mode.