Supplementary Material – Simultaneous reciprocal and real space X-ray imaging of time evolving systems

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FABRICATION OF X-RAY OPTICS AND SETUP CHARACTERISATION

In order to obtain the theoretically expected behaviour the zone plates have to be purely phase shifting with a phase shift of π, thus eliminating the zeroth order diffraction component. We utilised deep reactive ion etching (DRIE) of Si to fabricate the optical element. A 250 μm Si wafer was covered with a 50 nm Cr layer and a 200 nm layer of PMMA. The pattern of the optical element was written with an electron beam, developed and transferred into the Cr layer by reactive ion etching in Cl/CO_2 plasma. After removal of the remaining PMMA the Si was etched to the desired depth. To evaluate the performance of the fabricated optics we recorded images at the focal distance where a 300 μm thick LuAG:Ce scintillator converted the X-rays into visible light, magnified by 20 fold optics and finally recorded by the pco.edge 5.5 camera with a 6.5 μm pixel size (effective pixel size of 0.325 μm). Horizontal and vertical line profiles were used to estimate the full width half maxima (FWHM) in the x and y directions, which were $FWHM_x = 7.44\ \mu m$ and $FWHM_y = 3.76\ \mu m$ respectively. Ideally, the width of the focus should be equal to the width of the outer most zone, which in this case was 1.2 μm. However, due to the finite asymmetric source size of the TOMCAT beamline this discrepancy is observed. The asymmetry of the source will result in different noise performance in the horizontal and vertical directions. For the validation and dynamic imaging experiments a similar configuration was used but with optics designed for 25 keV and 1:1 optics for recording the diffraction patterns.

VALIDATION

Wave optics simulations To further validate our approach we developed a MATLAB based wave optics simulation tool. In particular, we wanted to demonstrate that the limited sample sampling points within each until cell are not hindering the retrieval of the microparticle diameter. The discrete simulation grid had a size of 1092 × 1092 which represented a single unit cell. We randomly generated positions of the spherical particles within each the unit cell. The number of particles, was choses such that volume fraction for each particles size is the same. For each particle size we run the simulation 20 times in order to collect sufficient statistics. The effective particle size was estimated by least mean square error fitting of the theoretical monodisperce model.

Micro computed tomography validation To evaluate if the observed effective diameter is a reasonable estimate, we performed high resolution tomographic scans of the sample when it had reached a steady state. The system was let to settle for 3 hours, no turbidity was visible in the main body of the sample. A 10x objective was utilised resulting in an effective pixel size of 0.65 μm which was sufficient to resolve particles down to a few micrometers. Due to the limited field of view of the high resolution computed tomography (CT) multiple scans were performed to cover the creamed area. The resolved particles were segmented [1] from the tomographic scans and finally the cumulative probability density function of the particle sizes was estimated as shown in Fig. 1. A representative CT slice used for the calculation of the cumulative probability density function is shown in the inset. From the cumulative probability density function we observe that approximately 70% of the particles are within the range of autocorrelation lengths probed. Therefore, until the time point $t = 2.8$ min a slight bias towards smaller effective diameters exists. After that time point, all particles are within the autocorrelation length range, and therefore no bias is present. This time is estimated as the time required for a particle with a diameter equal to the maximum autocorrelation length ξ_{max} to traverse the whole observation window based on its terminal Stokes velocity.

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[1] T. Hildebrand and P. Ruegsegger, Journal of Microscopy **185**, 67 (1997).

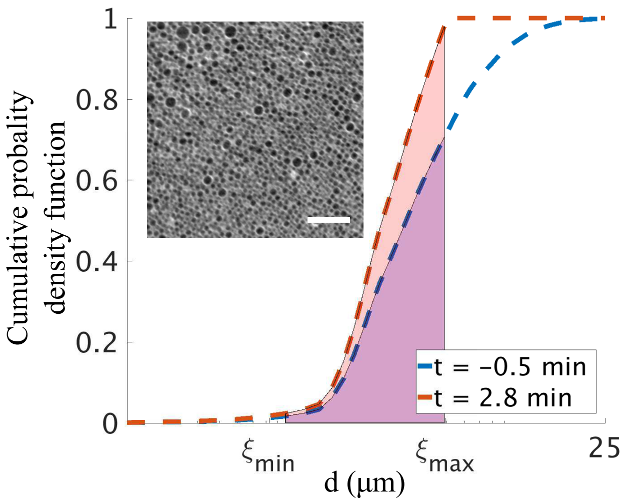

FIG. 1. **Cumulative distribution function of estimated particle diameters** The particle diameters were extracted from micro computed tomographic scans performed at the surface of the solution after 3 hours. The inset shows a representative micro computed tomographic slice, the scale bar corresponds to 100 μm. The estimated cumulative distribution function shows that the autocorrelation range of our system covers approximately 70% of the particles. Nonetheless, due to the delay between sample preparation and measurement, particles larger than 12 μm have already exited the observation window. At the time point $t = 2.8$ min all particles still in the observation window are 100% within the autocorrelation range, therefore no bias expected for the rest of the imaging.