Extracting single particle structure from heterogeneous disordered ensembles in two- and three dimensional space

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X-ray crystallography can be used to determine the structure of amorphous materials and many macromolecules, such as viruses and proteins. However, a lot of samples are incapable to crystallize or change their configuration during crystallization. Structure reconstruction from correlation functions is another approach to determine the particle structure. Averaging over a large number of angular correlation functions calculated from x-ray diffraction patterns cancels out the cross correlations between different particles and converge to the autocorrelation functions of single particles [1]. This technique was developed by modifying the fluctuation x-ray scattering technique developed by Kam et al.[2]. First experiments in two dimensional systems have already been carried out successfully on gold nanorods [3], triangular shaped particles [4], and nano-dumbbellsat both synchrotron [5] and FEL facilities [6].

In a first step we measured a two dimensional film of three differently-shaped silver colloids, namely rods, decahedral and plates. Therefore the sample was dried on a silicon nitrite membrane. The experiment was performed at the coherent scattering beamline P10. In our experiment the sample detector distance was about 5m. The beam was focused down to a few μm. A typical speckle pattern of the 2D film of the decahedral silver nanoparticles is shown in figure 1.

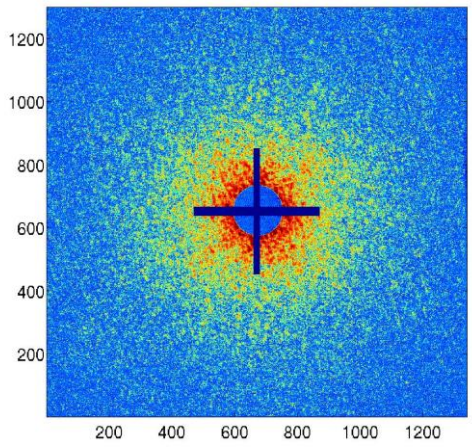

Figure 1: Speckle pattern of colloidal silver nanoparticles of the 2D film for the decahedral shaped nanoparticles.

Due to the small beamsize we were able to scan different spots of the sample and could extract an intensity map of the sample. As example we show the intensity map of the 2D film for the decahedral silver nanoparticles. Depending on the thickness of the film the intensity varies (Fig. 2a). For the further analysis we calculated the angular correlation function *C(Q,Δ):*

$$C(Q,\Delta) = \frac{\langle I(Q,\varphi) I(Q,\varphi+\Delta)\rangle_\varphi - \langle I(q,\varphi)\rangle_\varphi^2}{\langle I(q,\varphi)\rangle_\varphi^2}$$

To observe any periodic modulations we calculated $\hat{C}(Q,l)$, the Fourier transformation of *C(Q,Δ):*

$$\hat{C}(Q,l) = \int_0^{2\pi} C(Q,\Delta)\exp(il\Delta)\,d\Delta.$$

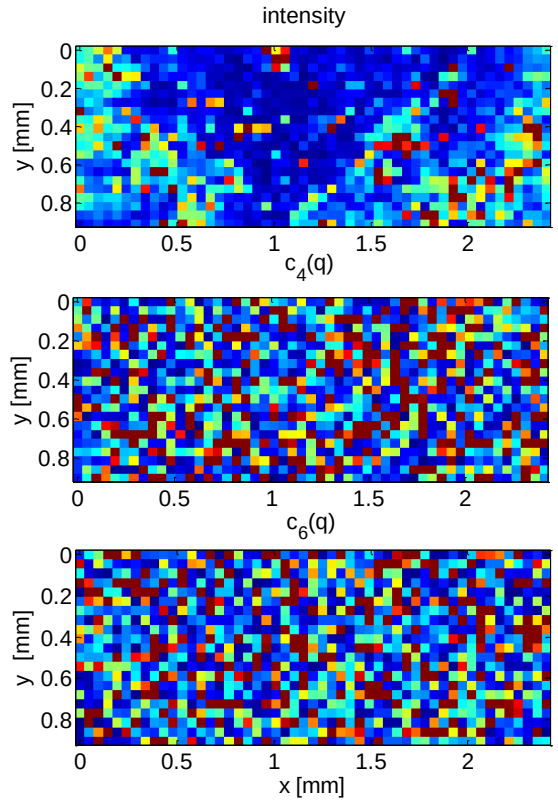

Figure 2: a) intensity map of the decahedral nanoparticles. Extracted Fourier coefficient of the angular corrleation function for l=4 (b) and l=6 (c).

The angular correlation function $\hat{C}(Q,l)$ differ for the different fourier coefficient, here shown for *l*=4 (Fig. 2b) and *l*=6 (Fig. 2c). A detailed analysis is still ongoing. However the data set are not complete. Due to technical problem, the beam was not continously available and the beamtime itself ended one day before the official end. As a next step we also measured the same samples in 3D. For the three dimensional samples the silver nanoparticles were dispersed in glycerol and put in capillaries of 0.7 mm.

Furthermore, recent simulations from two-dimensional systems demonstrated the feasibility of structural determination from mixtures from nanorods, nanoprism and nanorice [7]. The analysis is based on combining the corresponding single particle cases with an exact knowledge of the mixing ratios. Therefore we measured as well three different mixtures of decahedral, pentagonal cylinders and plates consisting out of silver particles. In a first approach we took images on a 2-dimensional film. Also here the data set is not complete due to technical problems.

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