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IT-10-O-2253 Possibilities and limitations for atom counting using quantitative ADF STEM

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Advanced statistical methods can be used to count the number of atoms in each atom column of high-resolution ADF STEM images [1-3]. Here we discuss the possibilities and limitations of achieving single atom sensitivity.

Four images of the same Ir/Pt nanoparticle were recorded at different magnifications and electron doses. In order to allow comparison with simulations, the images were normalised with respect to the incoming electron beam intensity [4]. Next, using statistical parameter estimation theory, the total scattered intensities are quantified atom column–by–atom column. An example analysis for the image recorded at the highest magnification and electron dose is illustrated in Fig. 1; the total scattered intensities are visualised in the histogram. The number of significant components and their intensities were retrieved by evaluating the so-called integrated classification likelihood (ICL) criterion in combination with Gaussian mixture model estimation. These results allow us to quantify the number of atoms in each atom column. As shown in [3], the reliability of atom counts depends on the number of atom columns present in an image, the width of the components, and the performance of the ICL criterion. These parameters can be linked with the quality of the recorded images.

In Fig. 2, the intensities of the components resulting from the counting analyses are compared with the total scattered intensities resulting from simulated images using STEMsim. For image 3 an excellent match was found. However, analysing images of lower magnification and/or electron dose worsens the match with simulation. The same effect is observed when analysing an image composed of every second pixel of image 3. In this way, the lower magnification of images 1 and 2 is mimicked. This leads to less precise measurements of the total scattered intensities resulting in insufficient statistics for the determination of the number of components. However, when enhancing the statistics by combining the values of the scattered intensities of the four images collectively, the experimental intensities again match with simulated values. In addition, the statistical approach for atom counting provides us high precision leading to near single atom sensitivity for this combined set of images.

In conclusion, an advanced quantitative method to count the number of atoms is presented together with its possibilities and limitations. Single atom sensitivity may be achieved when the experimental images are of sufficient quality to yield sufficient statistics.

References

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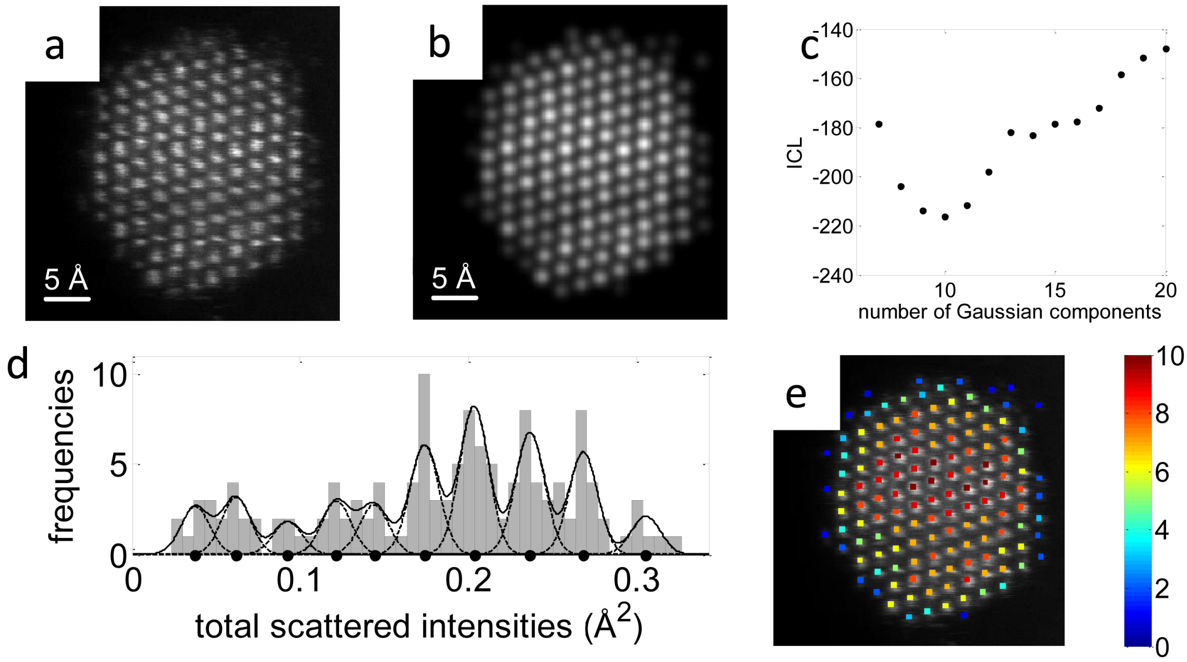

Fig. 1: Illustration of the atom counting procedure; a) experimental ADF STEM image, b) refined parameterised imaging model, c) evaluation of ICL as a function of the number of components, d) histogram of estimated scattered intensities together with the estimated Gaussian mixture model, e) quantification of the number of atoms in each atom column.

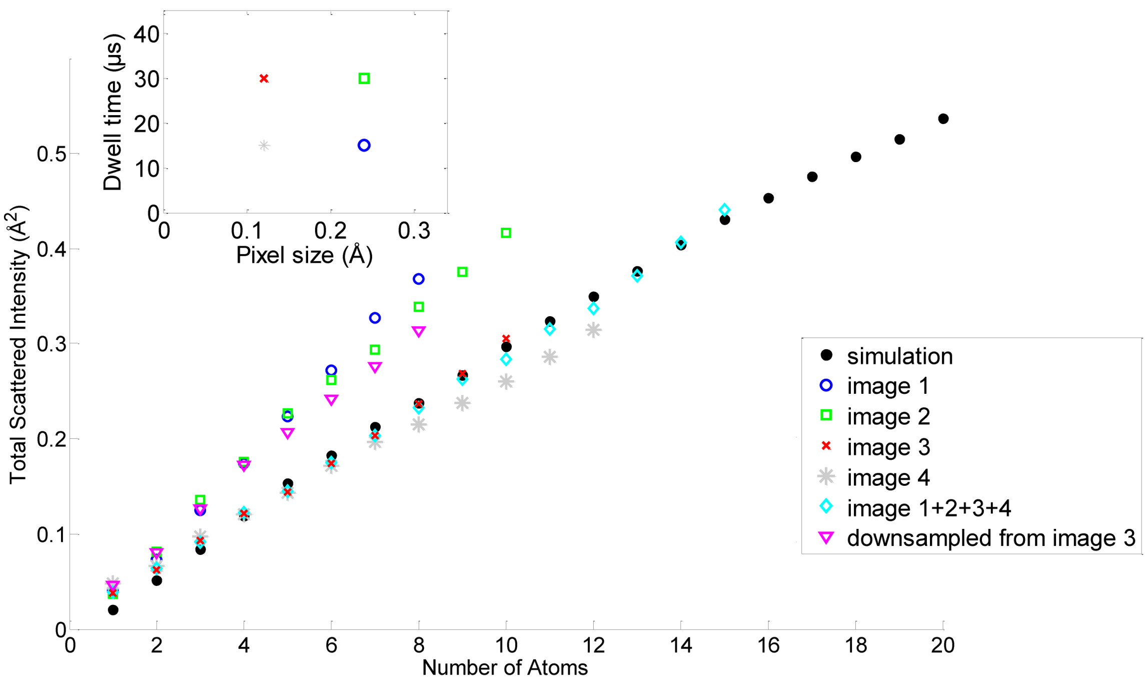

Fig. 2: Comparison of experimental and simulated total scattered intensities. The inset shows the specific pixel size and dwell time for the individual images of the Ir/Pt particle.