Automated Scattering Cross-section Analysis of HAADF STEM Data for Atom-counting & Nano-particle Metrology

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Background

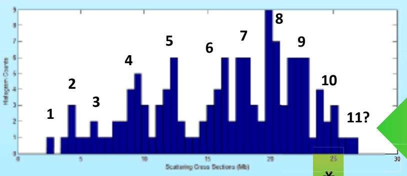
Nano-particle metrology is extremely important in the study of catalysts and the mass contrast of HAADF STEM provides a powerful tool for measuring the shape *and* thickness of particles from a single image[1].

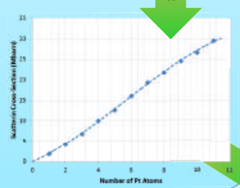
Here we describe our new 5-step high-throughput code for the automated peak-finding, background subtraction and normalisation of HAADF STEM images, from this data we perform image-segmentation and atom-counting within each atomic-column and determine the nanoparticle's 3D morphology.

④ Atom Counting

With the data now expressed on an absolute scale, integrating within polygons about each atomic-column position and multiplication by the pixel area gives the total scattering cross-section of each column in mega-barns (1 mb = 0.01 Å^2).

This data can then be compared with simulation[3] or analysed statistically[4] to perform atom counting and to subsequently deduce the size and structure of the nanoparticle.



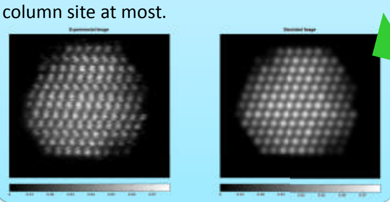


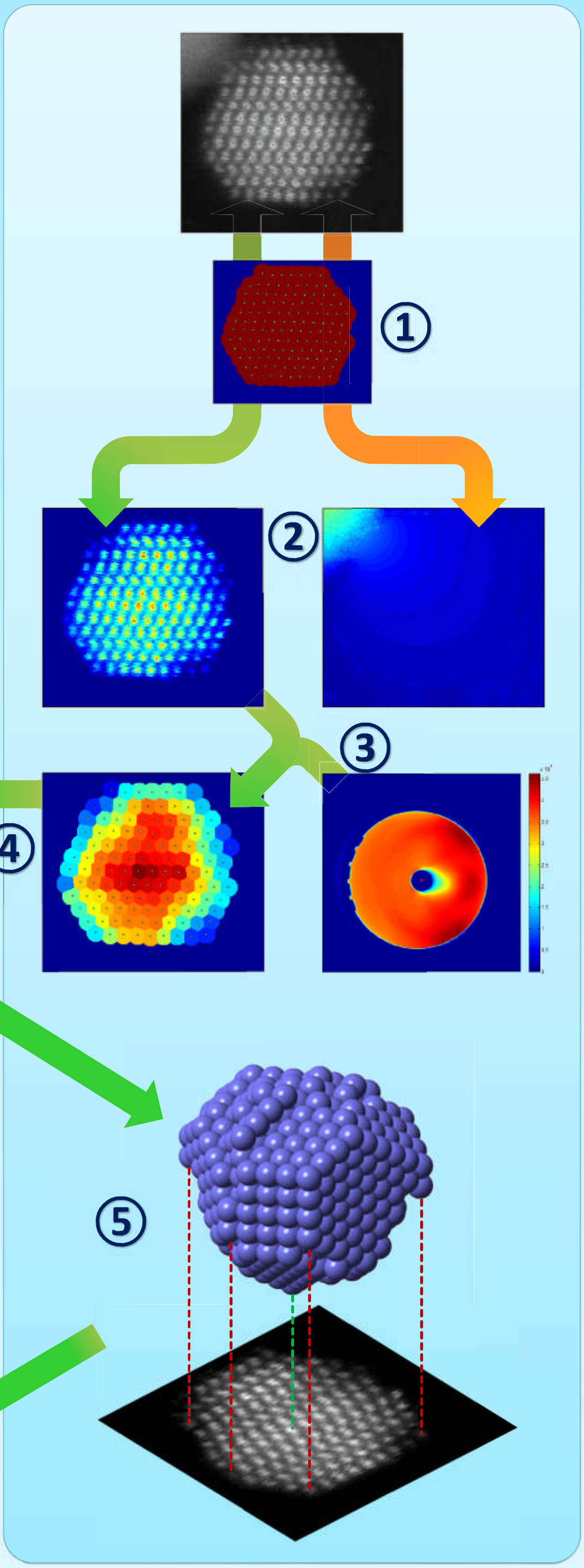
* Simulation library calculated using the same accelerating voltage and detector angles as the experiment.

⑤ Particle Reconstruction & Verification

With the number of atoms in every atomic-column identified the 3D morphology can be reconstructed (assuming the particle has no vacancies and a broadly smooth surface).

To verify the results of the reconstruction, the structure can be used as the input for an image simulation. Again the beam voltage and detector collection angles must match the experimental conditions. From the comparison below (matched greyscales) we believe the reconstruction is correct to within ± 1 atom per column site at most.





① Peak-finding & Masking

The analysis begins with a fully automated and robust peak-finding algorithm. This brand-new routine has been designed to make the best use of the a-priori form of the STEM point-spread function to outperform more simple intensity-maxima or template matching based methods.

Once the peak positions have been identified a mask is defined that encompasses all atomic-column positions. The region inside the mask includes sample with carbon support and outside the mask the image intensity represents the support only.

② Background In-painting & Subtraction

Discounting the portions of the image defined by the mask to contain intensity contributions from the nanoparticle sample, we are left with an image of the background that contains one or more holes. These holes are filled with a method similar to those in the fine-art restoration world.

An iterative Gaussian-blurring in-painting is used to reconstruct the intensity in the missing parts of the image. The result can be refined through 250 iterations in a few tens of seconds and produces a smooth estimate of the intensity 'behind' the nanoparticle sample. This can then be subtracted from the experimental image to leave the sample and the support clearly separated.

③ Detector Calibration & Image Normalisation

The detector map is created by scanning the STEM probe directly across the hardware[2]. From this we measure the number of counts that correspond to the vacuum-level (sometimes called the D.C. offset) and the number of extra counts recorded on the detector – its sensitivity or efficiency. Subtracting the offset and dividing by the sensitivity yields images expressed in units of fractional beam-current.

Conclusions

- The mass-thickness contrast of HAADF STEM makes it an incredibly powerful technique for nano-particle metrology,
- A high-throughput automated analysis code was designed that incorporates robust peak-finding, background reconstruction and subtraction, detector normalisation and atomic-column scattering cross-section integration,
- Comparison with simulation was used to directly count atoms and reconstruct the 3D particle morphology which was subsequently verified by image simulation.

Acknowledgments

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References

1) Singhal, Yang & Gibson (1997) *Ultramicroscopy* **67**, 191–206.
2) Lebeau & Stemmer (2008) *Ultramicroscopy* **108**, 1653–8.
3) E, MacArthur et. al. (2013) *Ultramicroscopy*.
4) Van Aert, Batenburg et. al.(2011) *Nature* **470**, 374–7.

Further Information

Copies of this poster, the associated manuscript and the analysis software described are available online at:

www.lewysjones.com