Delocalisation in images of Pt nanoparticles

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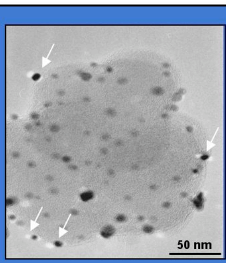
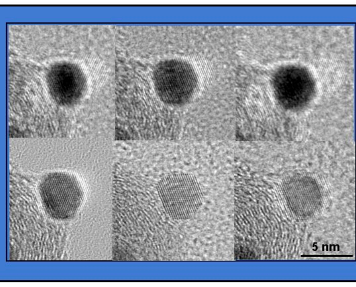
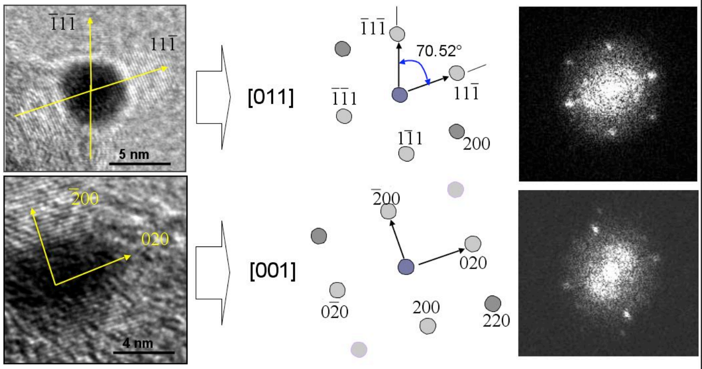
Abstract

Delocalisation is an imaging artefact that results in the presence of lattice fringes outside a crystal imaged in a transmission electron microscope (TEM) in the presence of spherical aberration and/ or defocus. Here, we show that delocalisation can be used to determine the absolute defocus values of chosen features in a specimen from a defocus series of images. We suggest that delocalisation in images of nanoparticles can be used to extract useful information about the specimen. We examine platinum nanoparticles that have an average size of 5nm and are supported on graphitic carbon. Experimental images are acquired at 200 kV using a JEOL 2200FS FEG (Field Emission Gun) TEM equipped with a spherical aberration corrector.





Qualitative information from delocalisation



When imaged out of focus, Fresnel fringes appear around the edge of a particle, and in addition the lattice fringes are displaced with respect to the true position of the particle, which becomes progressively darker with increasing defocus. The displaced lattice fringes move away from the particle linearly with defocus.

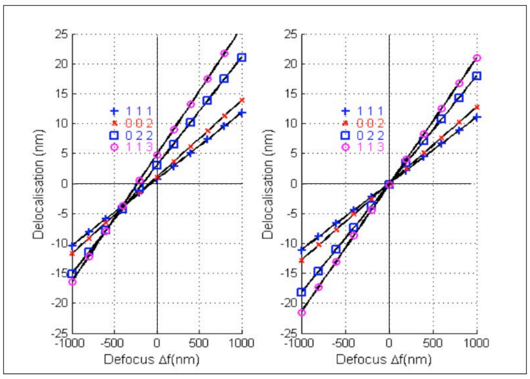
Qualitatively, the image shifts that accompany delocalisation can be used to provide information about the orientation of the particle. When the defocus is changed, bright contrast surrounding the particle moves in specific directions, which are related to its orientation. The relative intensities of the bright regions depend on the degree and direction of misalignment of the particle from the exact zone axis orientation.

Quantitative information from delocalisation

The extent of image delocalisation depends on the spatial frequency, g, and on the wave aberration function of the objective lens, $\chi(\mathrm{g})$. The equation that relates the delocalisation to the imaging conditions is

$$\Delta R = \lambda g \Delta f + C_s \lambda^3 g^3 \qquad [1]$$

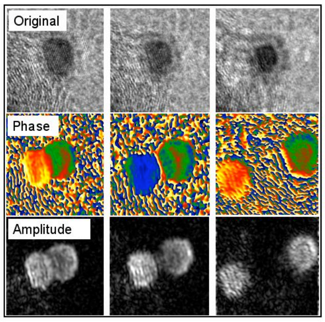
where ΔR is the image displacement, Δf is the defocus, λ is the electron wavelength and C_s is the spherical aberration coefficient. Delocalisation is linear with defocus for fixed λ and C_s. The graphs show Equation 1 plotted as a function of defocus for lower spatial frequency reflections in platinum.



The graphs show that when C_s is close to zero it is possible to find a defocus value for which the delocalisation can be minimised for *all* spatial frequencies at the same time. Clearly, for *every* spatial frequency there is a defocus value for which delocalisation is exactly zero.

Geometrical Phase analysis applied to delocalisation

Geometrical Phase Analysis allows the quantitative measurement of displacement and strain fields from lattice images. The method is based on centring a small aperture around a strong reflection in the Fourier transform of the image, and performing an inverse Fourier transform. The phase of the resulting complex image is used to provide information about local displacements of atomic planes from a reference lattice.

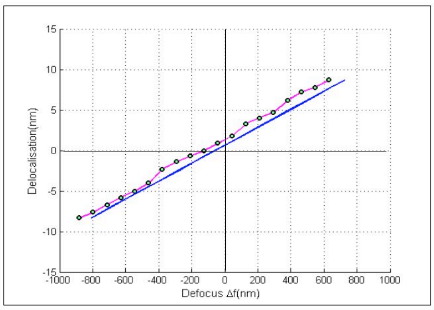


The images show a Pt particle at three different defocus values, and the phase and amplitude images that result from the application of *geometrical phase analysis* to the 111 reflection.

Method to calculate absolute defocus values from delocalisation

For nanoparticles, the defocus obtained from diffractogram analysis can be very different from the true defocus of the particle, which may be at a different height from the support film. A method to find the absolute defocus of a nanoparticle is now presented. On the assumption that *delocalisation is associated with a single spatial frequency*, the proposed method is as follows:

1. A through-focus series of the particle is acquired.
2. The extent of delocalisation in each image is measured, for example using *geometrical phase analysis*.
3. Defocus values are also measured from each image using diffractogram analysis.
4. The delocalisation values are plotted as a function of the defocus values obtained from diffractogram analysis.
5. The lateral displacement of the graph that is required for consistency with Equation 1 is determined.



The graph shows the application of this approach to the images of the particle shown above. The entire defocus series contained 20 images, with a defocus step size of 100 nm and with C_s adjusted to 0.5 mm. The slope of the experimental graph matches that of the theoretical graph for a spatial frequency that corresponds to the 111 reflection in Pt, and is displaced with respect to the predicted graph by an averaged value of 85 nm. The absolute defocus value of the particle in each image is therefore obtained by subtracting 85 nm from the values measured using diffractogram analysis.

References

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Conclusions

In the absence of spherical aberration, there is a defocus value at which delocalisation is absent for every spatial frequency in a lattice image. However, if spherical aberration is present, delocalisation can only be removed for one spatial frequency at a particular defocus value. Delocalisation can be used qualitatively to provide information about the defocus of a particle and its orientation. It can also be used to provide quantitative information about the absolute defocus of a nanoparticle, when combined with diffractogram analysis.