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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, what may be used to improve computer reconstruction methods. These values are important for optimising the accuracy of information about the specimen obtained using techniques such as exit wavefunction restoration.

1. Introduction

The high spatial and temporal coherence of a field emission gun (FEG) transmission electron microscope (TEM) increase the information limit of the microscope and affect the degree of localisation of the high-resolution information in a recorded image. At large defocus settings, the contrast transfer function (CTF) of the microscope contains many oscillations, which not only lead to contrast reversals, but are also responsible for delocalisation of the information in the image. Detail in the image is then displaced from its "true" position relative to the specimen, by an amount that increases with defocus from Scherzer focus [1]. We suggest that delocalisation in images of nanoparticles can be used to extract useful information about the specimen [2]. 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.

2. Delocalisation of nanoparticles

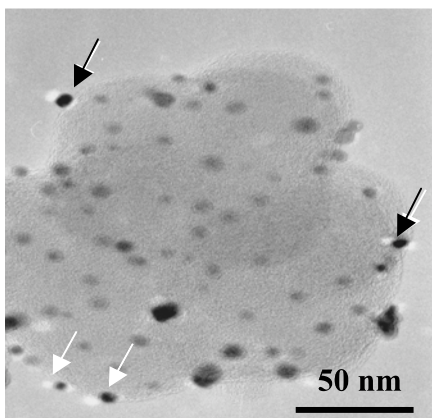

Figure 1. Bright-field image of Pt particles on a carbon support, showing the effects of delocalisation (arrowed).

The extent of image delocalisation, which occurs in every microscope, depends on the spherical aberration coefficient and the defocus value used. A perfect homogeneous crystal, with uniform thickness and no defects, has translational symmetry, and delocalisation effects are not readily apparent. Any imperfection or discontinuity such as at the edge of a specimen destroys this translational symmetry locally. Figure 1 shows platinum particles supported on graphitic carbon. Some of the particles are strongly diffracting and surrounded by bright contrast, which results from delocalisation.

Figure 2 shows a defocus series of high-resolution lattice images of a single Pt particle. When imaged out of focus, Fresnel fringes appear around the edge of the particle, and in addition the lattice fringes are displaced

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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.

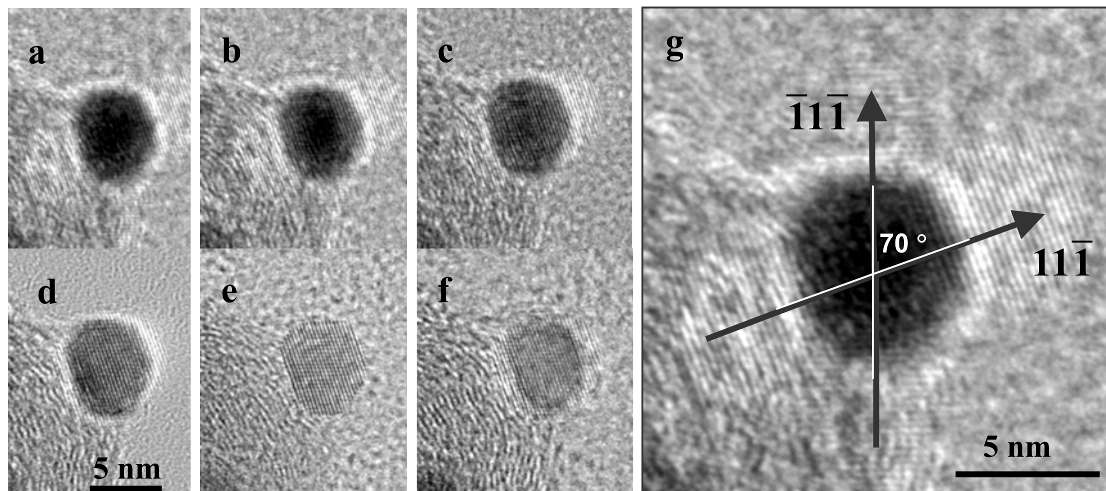

Figure 2. (a)-(f) Lattice images of a Pt particle shown as a function of defocus, for a spherical aberration coefficient of 0.5 mm. The defocus step size between successive images is 100 nm. When the particle is close to focus, delocalisation is minimised. The angle between the displaced images is 70º, as shown in (g).

3. Qualitative information from delocalisation

Qualitatively, the image shifts that accompany delocalisation can be used to provide information about the orientation of a particle. Figure 2 illustrates the fact that the regions of bright contrast that surround a particle move in specific directions, which are related to its orientation. The angle of 70º observed between the displaced images is close to the value of 70.52º expected between 111 reflections in a cubic structure oriented close to a [0 1 1] zone axis. The relative intensities of the bright regions depend on the degree and direction of misalignment of the particle from the exact zone axis orientation.

4. Quantitative information from delocalisation

A detailed discussion of delocalisation can be found in [1-3]. The extent of image delocalisation depends on the spatial frequency, g, and on the wave aberration function of the objective lens, $\chi(g)$. It is large when $\partial\chi(g)/\partial g$ is large. The equation that relates delocalisation to the imaging conditions is

$$\Delta R = \lambda g \Delta f + C_s \lambda^3 g^3 \qquad (1)$$

where ΔR is the displacement of the image, Δf is the defocus, λ is the electron wavelength and C_s is the spherical aberration coefficient. From Equation 1, it follows that delocalisation is linear with defocus for fixed λ and C_s. Figure 3 shows Equation 1 plotted as a function of defocus for lower spatial frequency reflections in platinum, an accelerating voltage of 200 kV and for $C_s = 0.5$ mm and -30 µm.

The graphs suggest 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.

One way of quantifying delocalisation is by measuring the displacement exhibited by the highest frequency limited by the objective aperture, because higher spatial frequencies suffer greater delocalisation and so allow more precise measurements of image displacements.

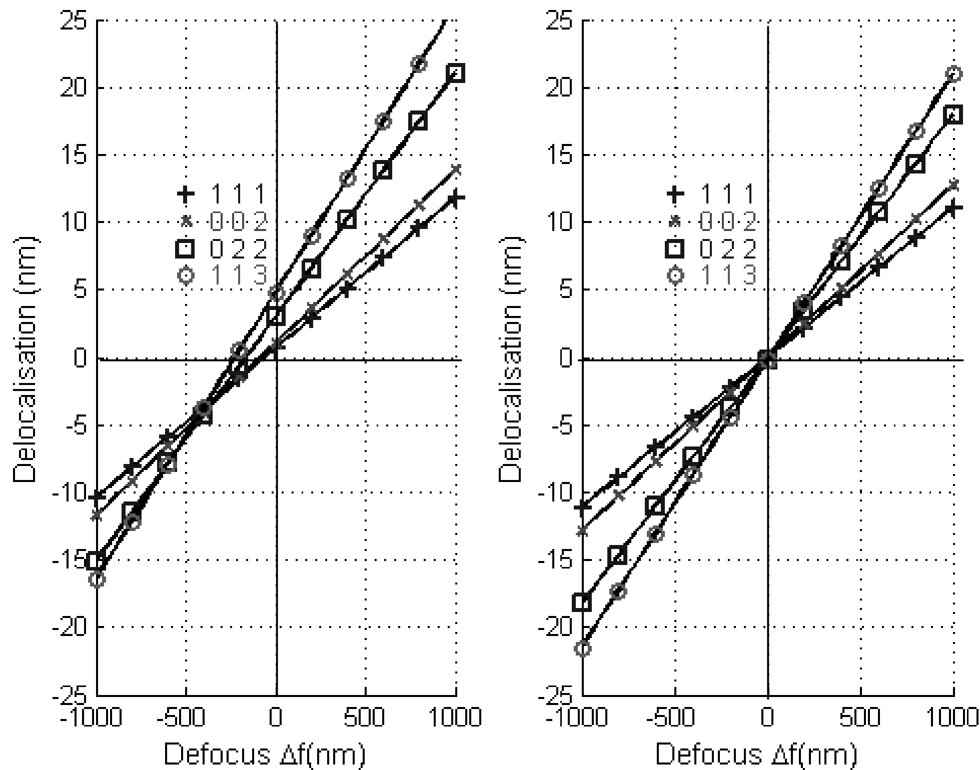

Figure 3. Theoretical relationship between defocus and delocalisation plotted as a function of spatial frequency for Pt at 200kV. (a) is for C_s= 0.5 mm. (b) is for C_s= -30

5. Absolute defocus measurement

The defocus value of an image is usually obtained from a comparison with simulated images or from a diffractogram acquired from amorphous material close to the area of interest. 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 delocalisationof the particle in each image is measured, for example using *Geometricl Phase Analysis* [4].
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.

Figure 4 shows the application of this approach to the images of the particle shown in figure 2. 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 predicted using Equation 1 for a spatial frequency that corresponds to the 111 reflection in Pt. It is displaced laterally 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.

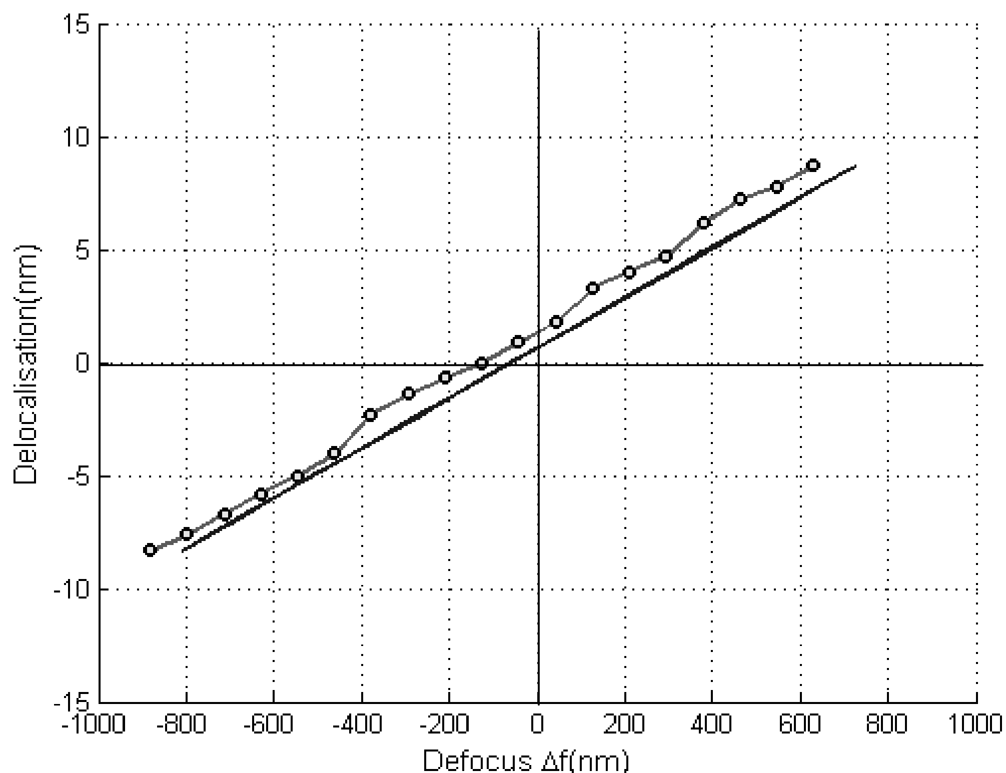

Figure 4. Predicted graph of delocalisation as a function of defocus, plotted for the 111 reflection in Pt for C_s=-30 μm. Alongside this line are plotted experimental values measured from the Pt particle shown in figure 2. These values are plotted as a function of defocus obtained from the carbon support film using diffractogram analysis.

6. Conclusion

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, what is useful for computer reconstruction methods like through-focus exit wave restoration (TF-EWR).

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