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Obstacles on the road towards atomic resolution tomography

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Electrons are the ideal particles to unravel the atomic structure of non-periodic objects such as nanoparticles, crystal defects and amorphous structures. As compared to X-rays, the interaction between electrons and atoms is orders of magnitude larger with less radiation damage [1]. Moreover, with the development of aberration correctors, high resolution electron microscopy is now entering the domain where individual atoms can be resolved. This means that neighboring atoms can be discriminated in an image. Then, it is in principle possible to refine atom positions quantitatively. The key to successful quantitative refinement is the availability of a pertinent parametric model of the images. The model includes all the ingredients needed to perform a computer simulation of the image. It is parametric in the quantities of interest, such as the atom positions. Quantitative refinement is then done by fitting the model to the image with respect to the unknown quantities. Once atom positions can be determined with high precision they can be used as input data for ab-initio calculations in order to understand the properties and eventually design new structures. The ultimate challenge is to quantitatively determine atom positions in an amorphous structure with high precision. However, as shown in [2], the only way out to resolve amorphous structures is to explore also the third dimension by means of electron tomography with atomic resolution. It is the aim of this abstract to discuss the obstacles on the road towards atomic resolution tomography.

An important problem that will arise is the existence of the so-called Stobbs factor expressing that the contrast in experimental images is commonly three times lower than in simulated images [3]. Therefore, it is still premature to presume the availability of a parametric model that can be used in a full quantitative refinement of the atom positions. In our opinion, the existence of the Stobbs factor is caused by phonon scattering. Experiments that could be suggested in the search for this Stobbs factor are for example off-axis holography in combination with a channelling map of the reconstructed exit wave, cryo experiments and Cs corrected experiments. Recent results will be discussed in the presentation.

Simulation results show that amorphous structures can in principle be resolved using electron tomography. Figure 1 shows a 3D reconstruction of amorphous tungsten at the left-hand side and a slice of this reconstruction at the right-hand side. In this slice, nearly all tungsten atoms are well resolved. Then, atom positions can in principle be quantitatively determined using the reconstruction as a starting-point for refinement. The precision of the thus obtained atom positions is limited by the inevitable presence of noise in the images. Using parameter estimation theory, it can be shown that the precision can be improved by increasing the incident dose. However, every incident electron has a finite probability to damage the structure, either by displacing an atom from its position or by changing chemical bonds due to ionization. Therefore, a compromise between precision and radiation damage has to be made which turns out to be optimal when the accelerating voltage is reduced. However, at low accelerating voltages, the instrumental aberrations become important. For this reason, a Cs and Cc corrector will be essential. By means of statistical experimental design [4], it can be shown that for a microscope operating at 50 kV, the precision of the position of, for example, a Si atom, can be improved with a factor of about 6 using an aberration corrected microscope as compared to the uncorrected microscope.

The most difficult objects for electron tomography at high resolution are semi-crystalline structures where a substantial part of the structure is perfectly crystalline but where one is especially interested in three-dimensional deviations from the crystallinity. Examples are crystal defects, quasi-amorphous structures containing microcrystals, and nanoparticles. The problem with tomography of semi-crystalline structures is that, when crystalline parts are in zone axis orientation, the interaction is strong and nonlinear and dominates the signal. Classical tomographic reconstruction schemes cannot cope with such nonlinearities. One possibility is to leave out these orientations from the starting dataset but this leads to a reduction of the resolution and to missing data in the reconstruction process. Thus far, no successful scheme has been developed that can cope with these problems. Therefore we will propose a new method that is called precession laminography.

Recently it has been argued by several authors (Pennycook and Voyles) that when highly advanced correctors would be developed in the future, the opening angle of a STEM probe might be increased to such an extent that the vertical cross-over of the probe in the crystal would be of the order of the atomic dimensions. If this would be true one could visualize a three-dimensional structure on the crystal slice by slice by optical sectioning. However, the Heisenberg uncertainty relation shows that for a vertical resolution of 1 Å, the uncertainty s on the wave vector should be of the order of 1 Å^{-1}. The corresponding spatial frequency g is $(2ks)^{1/2}$ which for 300 kV yields g=7 Å^{-1}. However, in a classical HAADF experiment the hole in the detector has to be large enough to eliminate the central disc. This means that only signals are detected with a spatial frequency beyond 7 reciprocal Å which is far beyond the classical phonon scattering, usually the main signal for HAADF STEM. Concluding, although optical sectioning at atomic resolution is theoretically possible, it will be unpractical because of the lack of signal.

References
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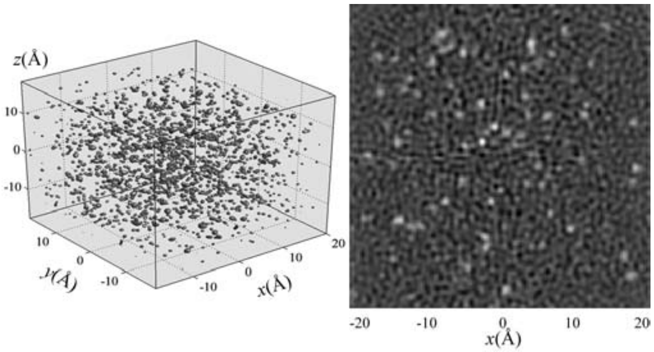

FIG. 1. A 3D reconstruction of amorphous tungsten from a simulated series of images is shown at the left of this picture, whereas a slice of this reconstruction is shown at the right.