FROM EXIT WAVE TO ATOMIC STRUCTURE

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Recently is has become a matter of routine to reconstruct the exit wave of an object at sub-angstrom resolution either by focal series reconstruction or by off-axis holography [1]. However, in order to interpret the amplitude and phase of the exit wave in terms of mass and position of the projected atom columns, one has to "invert" the dynamical scattering of the electrons in the object so as to obtain a starting structure which can the be used as a "seed" for further quantitative structure refinement.

Multislice methods or plane-wave based methods are not useful for this purpose since they do not explain on an intuitive basis why even in case of highly dynamical scattering the HREM exit wave can still be locally related to the projected structure. The classical picture of electrons traversing the crystal as plane-like waves in the directions of the Bragg beams, which stems from the X-ray diffraction picture and upon which most of the simulation programmes are based, is in fact misleading. The physical reason for this "local" dynamical diffraction is the channelling of the electrons along the atom columns parallel to the beam direction. Due to the positive electrostatic potential of the atoms, an atom column acts as a guide or channel for the electron [2] within which the electron can scatter dynamically without leaving the atom column.

If the distance between the atom columns is not too small, a one-to-one correspondence between the exit wave and the columnar structure of the crystal is maintained. Within the atom columns, the electrons oscillate as a function of depth without leaving the atom column. The periodicity is inversely related to the "density" of the atom column. It is important to note that channelling is not a property of a crystal, but occurs even in an isolated atom column and is not much affected by the neighbouring atom columns, provided the atom columns are not too closely spaced. Hence the one-to-one relationship is still present in case of defects such as translation interfaces or dislocations provided they are oriented with the atom columns parallel to the incident beam.

Furthermore, it is possible to parameterise the exit wave. This enables to retrieve the projected structure of the object from matching with the exit wave. In good approximation the S-state model can be written in closed analytical form (described by three parameters c_1, E_1, b respectively an excitation coefficient, the eigenenergy of the S-sate and the characteristic width of the uniform 1S-state):

$$\Psi(x,y,z) \simeq 1+\sum_j 2c_1^j \sin\left(\pi\frac{-E_1^j}{E_0}\frac{k_z}{2}z\right)\psi_1^j(x-x_j,y-y_j)\exp\left\{-i\pi\left(\frac{E_1^j}{E_0}\frac{k_z}{2}z-\frac{1}{2}\right)\right\}$$

with

$$\psi_1^j(x,y)=\sqrt{\frac{|E_1^j|}{\pi}}\frac{1}{b_j}\exp\left(-\frac{1}{2}\left(\sqrt{|E_1^j|}\frac{\sqrt{x^2+y^2}}{b_j}\right)^2\right)$$

with j the atom column index, E_0 the incident electron energy, k_z proportional to the inverse wavelength and z the crystal thickness [3]. This expression enables to determine the position and "weight" (E_1) of the atom columns by direct fitting with the exit wave.

It can be shown that the analytical S-state model describes the reality quite well for one atom column up to a thickness, which is equal to the first extinction distance [4].

from figure 1 that the systematic error on the atom column position as determined by direct fitting with the exit wave is smaller than 0.025 Å for Sn and 0.075 Å for Si up to relatively large thicknesses. The atom column positions in a dumbbell tend to move to each other to correct for the effect of overlap of their potential, which is not correctly described by the analytic S-state model. To prove this, the exit wave calculated with the S-state model, described by the equation above, is compared to the exit wave calculated by means of a multislice algorithm for various thicknesses. Figure 2 shows the amplitude and phase of respectively the analytical S-state model and a multislice calculation for Sn [110] at 90 Å specimen thickness. It is clear that the S-state model agrees fairly well with the multislice simulation.

Up to now an expression for the wave function $\Psi(x,y,z)$, given by the S-state model, was considered. However, in contrast with $\Psi(x,y,z)$, $\Psi(x,y,z)-1$ is much more direct interpretable. The amplitude of $\Psi(x,y,z)-1$ is strongly peaked at the atom column positions and the phase is constant over the atom column and proportional to the "weight" (E_1) of the atom column. According the S-state model, the amplitude of $\Psi(x,y,z)-1$ oscillates periodically in function of thickness and the phase increases linearly as function of thickness. The amplitude of $\Psi(x,y,z)-1$ allows to determine the atom column positions accurately and the phase of $\Psi(x,y,z)-1$ allows single atom sensitivity as will be shown in the next. A convenient way to visualise this is by plotting the pixels of the exit wave in a channelling map, which allows linearization of the data. This is an Argand diagram as first applied by Sinkler and Marks to thin crystals [5]. The complex pixel values are plotted as points using the x-axis as the real axis and the y-axis the imaginary axis. The points are now located on a disk which has a point of contact in the origin and which is symmetric with respect to the x-axis. The points can now be assigned to sectors, which correspond with a certain amount of atoms in the atom columns. In this way is single atom sensitivity feasible, as is shown in figure 3 where the pixels values of an experimentally reconstructed Au exit wave are plotted in a channelling map. The phase change at one gold atom is about 0.6 rad and is independent of thickness. Although here an example of a material with only one atom specie is given the method is applicable to any material even if atom species in atom columns are mixed and distinction of individual atoms is not possible.

References

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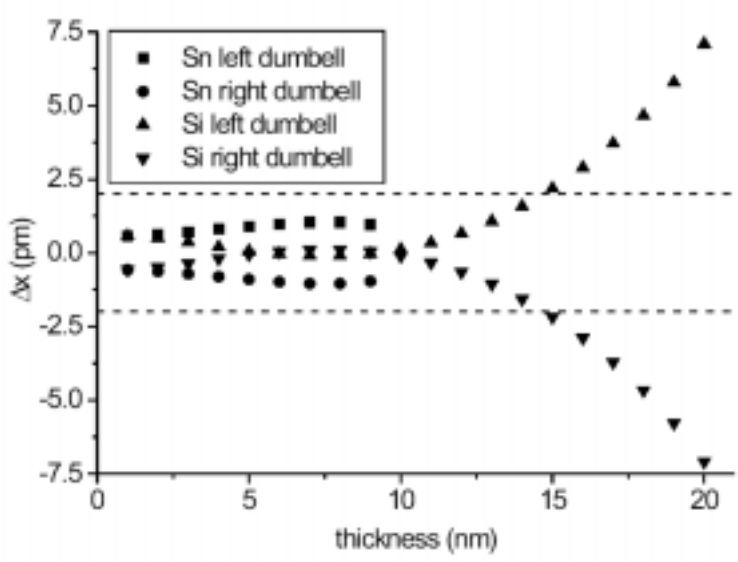

Figure 1: The systematic error on the fitted atom column positions of both the left atom column and right atom column of a dumbbell along [001], by matching the analytic S-state model to multislice calculations.

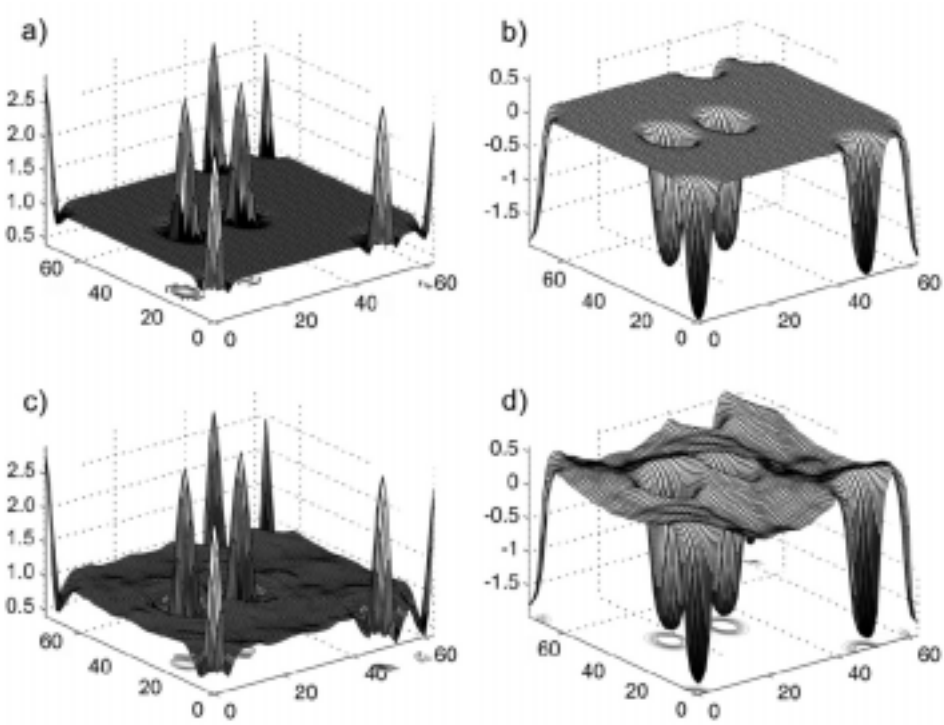

Figure 2: The exit wave of Sn [110] at a thickness of 90 Å. a.) the amplitude of the S-state model b.) the phase of the S-state model c.) the amplitude of a multislice calculation d.) the phase of a multislice calculation.

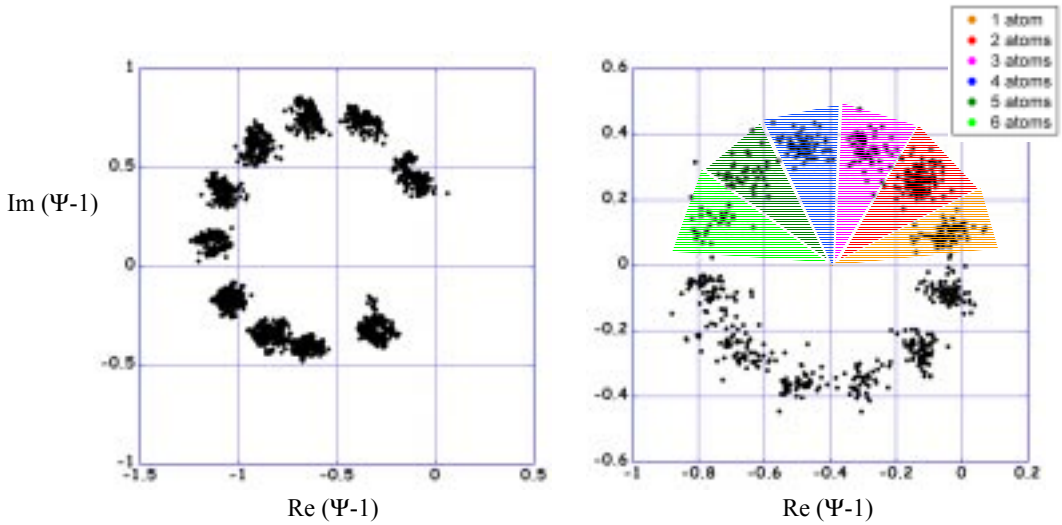

Figure 3: a) Channelling map of a simulated exit wave of a wedge shaped Au [110] sample (10 layers) by multislice calculations (300 keV, Debye Waller factor 0.5 $Å^{-2}$). b) Channelling map of an experimental reconstructed wedge shaped Au [110] sample. The complex pixel values can be assigned to sectors, which correspond to different numbers of atoms in the atom columns.