3D reconstruction of nanocrystalline particles from a single projection

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Abstract

This paper describes an approach to retrieve the three-dimensional atomic structure of a nanocrystalline particle from the reconstructed electron exit wave function in a single projection direction. The method employs wave propagation to determine the local exit surface of each atomic column together with its mass. The exit wave in between columns is used as internal calibration so as to remove the background noise and improve the precision to the level of single atom sensitivity.. The validity of the approach is tested with exit wave functions of a gold wedge reconstructed from simulated images containing different levels of noise.

1. Introduction

It is known that the structure-property relations of matter are encoded in the three-dimensional positions of the atoms [1]. Aberration-corrected transmission electron microscopy (TEM) is now well recognized for its ability to detect single atoms with Sub-Ångstrom resolution in 2D images [2-4] or reconstructed exit waves [5], But in general a single two-dimensional projection does not contain sufficient information to retrieve all atom locations within a three-dimensional object so that one needs multiple projections. However the classical tomographic reconstruction schemes based on combining images from many multiple viewing directions are not yet able to reconstruct objects at atomic resolution. The main reasons are the extreme stability requirements for the instrument, the limitations on the shape and the support of the object. Furthermore threshold values for electron beam-sample interactions are reported [6] which should not be overcome during image acquisition in order to guarantee the sample integrity. It was also shown that these undesirable sample alterations can be minimized by using low dose rates [7].

A particular problem is encountered for semi- or polycrystalline samples where in the direction of the atomic columns the electron scattering becomes dynamic and strongly nonlinear so that the classical tomographic schemes based on filtered backprojection are not applicable. Only in a few very favorable cases of crystalline samples atomic resolution could be obtained [8-13].

However for the study of nanoparticles such as for catalysts there is a need for a generic tomographic method that allows a fast characterization of the shape of pristine particles at atomic resolution. Here we present a new approach for a 3D reconstruction of all the atoms positions of an arbitrarily shaped crystalline particle that only needs one projection direction. In this way one can avoid the complicated tilting schemes and reduce the total electron dose. The starting point is the electron exit wave obtained in a zone axis orientation, that is reconstructed from focal series of images [14-16]

This exit wave is analysed in depth using the channelling theory [17,18] which provides a simple physical model for the interaction of the incident electron with the atoms of that sample.

In a zone axis orientation the impinging electrons are trapped in the positive potential of the column atoms and have not enough transverse kinetic energy to escape as long as the incident wave is planar and not converged. Thus in broad beam illumination schemes, which are used to produce HRTEM images, the dynamic scattering takes place within each column without interferences from the neighbouring columns as long as the lateral column separation is large enough and the object thickness does not exceed tens of nanometer. The effect is known as “electron channeling” and occurs in perfectly periodic objects as well as in defective crystals.

It is convenient to describe channeling effects using a wave picture. Figure 1(a) shows the geometry of imaging in zone axis condition when channeling occurs. In this case every atom acts as a lens that slightly focuses the electron wave. After passing across a number of atoms the electron wave is focussed in a point and further propagation it starts to defocus. The result is a periodical motion over a sample thickness t_0 , which is called an extinction distance. It is clear that the interaction is highly nonlinear. Furthermore, electron channeling makes it impossible to “focus” the electron beam inside of the object. On the other hand the electron exit wave function at the bottom of the sample is dynamically modulated by the location of atoms sites in beam (z) direction and their occupation with different elements. Therefore, it can be analyzed locally column-by-column and contains three-dimensional information. The channeling theory is simple [17] and provides a intuitive way to interpret the complex electron exit wave function by using amplitude and phase values in form of an Argand plot. The electron exit wave at the bottom of the column $\psi_e(r(x,y),z)$ can be equated as follows [17].

$$\psi_e(r,z)=\psi(r,0)+\Phi_{1s}(r)[\exp(-i\pi E_s k z/E)-1]..... (1)$$

Where $\psi(r,0)$ is the background (entrance) wave, $\Phi_{1s}(r)$ is the 1S state of the column and E_s its eigen energy. E is the energy of the incident electron and k its wave vector and z is the thickness of the sample.

Since the plane of the reconstructed exit wave does not coincide with the bottom of the column we have to propagate this column wave further over a distance Δf (which we will call defocus distance). After propagation over Δf the wave takes the explicit form [17].

$$\psi_e(r,z)=\psi(r,0)+[C(2\pi)^{1/2}a/(\pi a^2+i\Delta f\lambda)]\exp[-\pi^3/(\pi a^2+i\Delta f\lambda)] [\exp(-i\pi E_s k z/E)-1] \quad (2)$$

Where a is the $1/e$ radius of the 1S state (assumed gaussian) and λ the electron wavelength. This equation implicitly gives the information of the mass of the atomic column and the focal distance from the reconstructed exit wave plane to the bottom of the column. Figure 1(b) shows the expression for the exit wave of a particular column and its representation in the Argand plot a complex plane using the simple 1S state model derived in reference [17]. The 1S model predicts that when the mass of the column is varied, the solid black circular symbols describing the wave function at the exit surface of the sample (i.e. at zero focus) moves along the depicted grey circle. The associated angle θ is proportional to the mass thickness ($E_s \cdot z$) of the column. This “mass circle” is described by the factor $[\exp(-i\pi E_s k z/E)-1]$. By default samples exhibit shape variations across the field of view so that the defocus distance $\Delta f(r(x,y))$ varies with the projected position of the column (x,y) . This local variation is described by the defocus circle $[C(2\pi)^{1/2}a/(\pi a^2+i\Delta f\lambda)]\exp[-\pi^3/(\pi a^2+i\Delta f\lambda)]$ as indicated by the dashed circles in Figure 1. In a sense the Argand plot provides an orthogonal coordinate system that measures the column mass and defocus independently and locally. It provides simple insight into the physics of dynamic electron scattering. In Figure 1 only the pixel values from the center of the column are depicted.

As mentioned before, a given object geometry does not allow defining a global zero-focus value across the entire field of view of a micrograph unless all samples would be atomically flat sheets. Thus, the electron exit wave function is typically defocused by a value $\Delta f(x,y)$ away from the samples exact surface. Since the focal spread is small in state-of-the-art electron microscopes (~ 0.6 nm in TEAM 0.5), such improper defocus values alter the image contrast significantly, in particular if low acceleration voltages are used. For thin samples made from light atoms such as a few monolayers of graphene, the local, focal distance from the zero-focus plane of the electron exit wave function to the objects exit surface can be determined with a precision as high as $0.1\text{\AA}$ using the Big-Bang scheme [5]. This method was based on the idea that an atom is a weak phase object with a sharply peaked shape. But since the interaction of the electron and a column is very strong we cannot use the weak phase approximation. Nevertheless we can still exploit the fact that the $\Phi_{1s}(r)$ wave at the bottom of the column is still sharply peaked.

We therefore proceed as follows:

From (2) it follows that in the center of the column ($r = 0$) and for relatively small defocus values Δf , the intensity varies with defocus as

$$I = 1 + C/[(\pi a^2)^2 + (\Delta f \lambda)^2] \quad (3)$$

This is a Lorentzian function with a maximum at zero defocus and a FWHM (above the background) of

$$\text{FWHM} = 2\pi a^2/\lambda \quad (4)$$

Using typical values of

$a = 35$ pm and $\lambda = 2$ pm (300keV) we get $\text{FWHM} = 3.8$ nm.

As we can see later in fig. 3 (a), the FWHM of the intensity vs focus curve is consistent with this value. It is worth mentioning that the accuracy in determining the vertical position of the bottom of a column depends quadratically on the resolution of microscope. Thus by proper fitting and sufficient signal to noise it is possible to determine the vertical position of a column with a precision comparable to [5].

As shown in [17] the mass of the atomic column can in principle be determined from the position along the mass circle starting from the origin. However the main problem is that the position of the origin corresponds with the background wave. And it is known that one cannot retrieve the low spatial frequencies from a focal series. For this reason the reconstructed background wave is unreliable over long distances. And hence the determination of the origin of Argand space is subject to inaccuracy and noise. Therefore we devised a local calibration procedure which is much more reliable and more routinely applicable to experimental exit waves. This procedure measures the reference wave locally at positions in between the atom columns where the incident wave only feels the mean inner potential of the sample. In this way we can reach single atom sensitivity which allows to count the number of atoms in every column. By combining both the defocus distance and the number of atoms in of every column we are able to reconstruct the shape of the nanocrystalline particle to the level of the individual atoms.

The method is then applied successfully to exit waves of a gold wedge nanocrystal reconstructed from simulated images containing three different noise levels.

2. Implementation

The numerical procedures to determine the focus value and column mass from the electron exit wave function exit wave plane was developed and tested against simulated images from a gold wedge:

- (i) A wedge-shaped gold nanocrystal is modeled and its electron exit wave function $\Psi_e(r, z)$ is reconstructed from 20 simulated images using the McTempas software package [19]. The optical parameters of TEAM 0.5 are used (accelerating voltage = 300 keV, $C_s = -0.013 \mu\text{m}$, convergent angle = 0.1 mrad focal spread = $10 \text{ \AA}$). The Debye-Waller factor for Au = $0.5 \text{ \AA}^2$. The reconstructed exit wave contains structural information up to the information limit which is reported to be near $0.5 \text{ \AA}$. The signal to noise in the simulation is set to be about 0%, 10% and 50%, respectively (experimentally the signal/noise for a similar Au sample was determined to be around 10%). The wedge crystal is oriented such that the flat [001] top surface of the wedge crystal faces the incident electron beam. The stacking sequence along Au[001] is “abab...”. The distance between “a” and “b” layers is $2.04 \text{ \AA}$. Fig. 2(a) and 2(b) are the modulus respectively the phase of the reconstructed exit wave function $\Psi_e(r, z)$ from simulated images containing 10% signal/noise. The thicker part of the nanocrystal (point “A” on line A-B) contains five atoms and the thinnest part “B” on the line A-B contains only one atom. The distance between two gold atoms along the [001] direction is $4.08 \text{ \AA}$. Therefore the focus difference between “A” and “B” along line A-B is about $18 \text{ \AA}$. The complex value at the center of each column of the wave functions reconstructed from simulated images is extracted and plotted in the Argand plot in fig. 2(c) for three different values of the signal/noise (blue, green and red dots for 0%, 10% and 50% respectively). The corresponding number of atoms for each accumulation point is marked. The {001} planes are composed of an “abab...” stacking sequence. The gold [001] structure has “a” and “b” layers and surface corrugation but they are no lateral displacements. Although the two columns have the same number of atoms, their positions in the argand plane are slightly different. This is due to the potential eccentricity effect [20] by which the atom in the “b” layer has only an effective “half potential” of the atom in “a” layer.
- (ii) In the next step, we calibrate the reconstructed wave function $\Psi_e(r, z)$ using the background wave $\Psi_o(r)$. We determine this background wave by fitting the complex value at “valley” points between the atomic columns which are marked by red crosses in fig. 2(a) and blue, green and red dots near (1,0) in the Argand space of fig. 2(c). The fitted phase map of the background wave is shown in the figure 2(d). A line plot of background phase Φ_o along the line A-B is also given in the figure 2 (e) for three different s/n cases. We can see that the background phase increases monotonically from B to A. This is due to the fact that the background wave that passes in between the atom columns is not totally free but feels the “mean inner potential” that causes a phase shift that is proportional to the travelled distance.
- (iii) The background subtracted exit wave function $\Psi'_e(r, z)$ is given as:

$$\Psi'_e(r, z) = [\Psi_e(r, z) - \Psi_o(r)] / \Psi_o(r) \dots\dots\dots (5)$$

$$= [\Psi_e(r, z) / \Psi_o(r)] - 1 \dots\dots\dots (6)$$

- (iv) After internal calibration the exit wave is propagated back numerically over defocus distances with increments of $0.1 \text{ \AA}$ while monitoring the intensity. Fig. 3(a) shows the intensity profile of the

defocused wave versus propagation (focus) distance f for two columns in edge of A (blue) and B (red) as shown in fig. 2(b). The curve is obtained from the wave with 10% noise. As it is shown the FWHM for both curves is about 3.5 nm which is in consistent with estimation of equation (4). The exact focus value associated with each column is set by the maximum intensity for each column. The intensity maximum of both intensity curves occur roughly at -2.5 and 16 Å, respectively. Fig. 3(b) shows cross-sections of the propagation intensity from the wave with 10% signal/ noise for all columns along the red line in fig. 2(b). Connecting the positions of maximum intensity reveals the wedge shape of the exit surface. The extracted focus differences correspond to the geometric differences in height of 18 Å of the two edges of the sample. The solid and dashed lines are associated with atoms located in “a” and “b” layers of the structure, respectively. A plot of focal values of each column along AB line is given in fig. 3(c) for three waves associated with s/n value of 0% (blue), 10% (green) and 50% (red), respectively. The differences along the y-axis show the error bars for the focal values which ranges from 0.1-0.4 Å for s/n=0% and 10%, and 0.3-1.0 Å for s/n=50%. With these error bars, the expected surface corrugation between a and b layers which is 2.04 Å. (1/2 lattice parameter) is reproduced, except for the case of s/n=50% (red dots) where we get 14 and 16 Å. A focal map of selected 285 atom columns from waves associated with s/n=10% is shown in fig. 3(c). The expected focus gradient along the line A-B is recovered accurately in Fig. 3(d), which shows the expected surface corrugation between a and b layers which is 2.04 Å.

(v) In order to determine the position on the mass circle the wave of every column a has first to be set to its exact zero-focus value. The Figure 4(a)-4(c) depicts the original (blue, green and red dots) and the corrected waves (black dots, the mass circle) for the examples of this gold wedge. Further, the mass value of the column mass can be calibrated using the background wave as yardstick. The phase of the background wave varies with depth and with the mean inner potential.

$$\phi_o = \sigma V_o t \dots\dots\dots (7)$$

Obviously such an assignment holds only if the resolution of a microscope is sufficient to allow revealing the undistorted space between atoms. According to the equation (7). The phase of the mean inner potential (valley wave) ϕ_o varies linearly with thickness. The mean inner potential of Au is 32.2V [21]. For electrons accelerated by 300 keV, σ is 0.00065 rad/ (eVÅ). The mass of each column can then be calibrated by the mean inner potential since a background correction corresponds to a shift of the wave function by (1,0) in Argand space. If we now plot the phases of the mass circle θ against the the phases of the valley wave ϕ_o in Argand space (fig. 2(e)) we expect a straight line through the origin as shown in Fig. 4(d). The blue, green and red dots in fig. 4(d) are associated with different s/n cases correspondingly. The slope s of this graph is a proportionality factor that quantifies the number of atoms in each column. The slope s in fig. 4(d) is fitted to be about around between -12.7 and -13.3 for three cases. For gold [100], for instance, the distance between successive layers in beam direction is 2.04 Å. The number of atom n in each column is given by:

$$n = (\theta/s) / (2.04\sigma V_o) \dots\dots\dots (8)$$

Fig. 4(e) shows a plot of phases of the mass circle θ against the number of atoms n given in equation (8). The number of atoms of every column is rounded to the nearest integer and the assignment is plotted in figure 4(e). The standard deviation in the number atoms is less than 1 atom for all three cases. Fig. 4(e) confirms that the modelled nanocrystal contains 1 to 5 atoms as in agreement with the structure used as input for the simulations.. Both, the focus dependences (fig.

3(b)) and the column mass variation (fig. 4(c)) provide sufficient information to construct a three dimensional image of the gold wedge as shown in Figure 5 from a single projection if the crystal structure of the material is known.

Conclusions

In this paper, we developed a generic procedure that allow retrieving the three dimensional atomic structure of a nanocrystalline particle from a single projection. The application assumes that the crystal structure is known and the material is homogeneous. At most, two other projections of the same material are needed if these assumptions cannot be made. Our approach allows minimizing the object exposure to an electron beam and is compatible experiments that use ultra low dose rates for the reconstruction of electron exit wave functions [10].

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Fig. captions:

Figure 1 (a) In the channeling condition the electron wave is trapped in an atomic column (blue dots) inside the crystal and the exit wave is presented conceptually as a red line **(b)** the complex value of the exit wave at the bottom surface of the column is presented as a solid dot in the Argand plot. When the mass of the column increases the dots move along a circle called the mass circle. When the wave is further defocused over a distance Δf away from the bottom surface, the point moves along another circle called the defocus circle (dashed circle)

Figure 2 (a) and (b) modulus and phase of the reconstructed exit wave of an Au wedge crystal reconstructed from simulated images containing 10% noise. **(c)** is the Argand plot of the original column waves (within black dash-circles) and background waves near (1,0) for three different noise levels 0% (blue), 10%(green) and 50%(red). The column waves with the same number of atom are grouped. **(d)** phase of the background wave obtained by interpolation from values of the valley points. The colour bar is indicated **(e)** phase line plot along line A-B.

Figure 3 (a) profile of intensity versus defocus of two columns at position A and B in the red line A-B. This curve is obtained from the wave with 10% noise. **(b)** intensity of de-focused wave from all atomic columns in the red line A-B of fig. 2(b). The range of focus is from $-120\text{\AA}$ to $120\text{\AA}$ **(c)** plot of defocus values for atomic columns along the line A-B. Blue, green and red dots are from the waves reconstructed from images with respective noise levels 0%, 10% and 50% **(d)** perspective view (top) and projected (bottom) views of the focal map determined from the intensity maximum of the defocused wave. The surface corrugation of “a” and “b” columns is clearly revealed.

Fig. 4 (a)–4(c) The blue, green and red dots are the background subtracted waves associated with three different noise levels 0%, 10% and 50%, respectively. The black dots are the focus corrected waves located on the mass circle **(d)** graph of the angle along the mass circle (black dots in 4(a)–4(c)) vs the phase of the background wave (phase value in 2(e)) **(e)** plot of the angle along the mass circle vs number of atoms (determined by equation (8)) **(f)** mass map.

Fig. 5 The reconstructed tomogram of the full Au wedge crystal based on the combination of focal map in figure 3(c) and the mass map in figure 4(f)

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a

b

Figure 1 (a) In the channeling condition the electron wave is trapped in an atomic column (blue dots) inside the crystal and the exit wave is presented conceptually as a red line **(b)** the complex value of the exit wave at the bottom surface of the column is presented as a solid dot in the Argand plot. When the mass of the column increases the dots move along a circle called the mass circle. When the wave is further defocused over a distance Δf away from the bottom surface, the point moves along another circle called the defocus circle (dashed circle)

a

b

imaginary

1.0

0.0

-1.0

real

0.0

1.0

c

N=0%

N=10%

N=50%

background
wave

ϕ_o
(rad)

-0.02

-0.04

-0.06

-0.08

-0.1

-0.12

-0.14

-0.16

-0.18

-0.2

$N=0\%$

$N=10\%$

$N=50\%$

A

position

B

d

e

Figure 2 (a) and (b) modulus and phase of the reconstructed exit wave of an Au wedge crystal reconstructed from simulated images containing 10% noise. **(c)** is the Argand plot of the original column waves (within black dash-circles) and background waves near (1,0) for three different noise levels 0% (blue), 10%(green) and 50%(red). The column waves with the same number of atom are grouped. **(d)** phase of the background wave obtained by interpolation from values of the valley points. The colour bar is indicated **(e)** phase line plot along line A-B.

$\Delta f = 120 \text{ \AA}$

$\Delta f = 0 \text{ \AA}$

$\Delta f = -120 \text{ \AA}$

b

18 \AA

d

Figure 3 (a) profile of intensity versus defocus of two columns at position A and B in the red line A-B. This curve is obtained from the wave with 10% noise. (b) intensity of de-focused wave from all atomic columns in the red line A-B of fig. 2(b). The range of focus is from $-120\text{\AA}$ to $120\text{\AA}$ (c) plot of defocus values for atomic columns along the line A-B. Blue, green and red dots are from the waves reconstructed from images with respective noise levels 0%, 10% and 50%(d) perspective view (top) and projected (bottom) views of the focal map determined from the intensity maximum of the defocused wave. The surface corrugation of “a” and “b” columns is clearly revealed.

f

Fig. 4 (a)-4(c) The blue, green and red dots are the background subtracted waves associated with three different noise levels 0%, 10% and 50%, respectively. The black dots are the focus corrected waves located on the mass circle **(d)** graph of the angle along the mass circle (black dots in 4(a)-4(c)) vs the phase of the background wave (phase value in 2(e)) **(e)** plot of the angle along the mass circle vs number of atoms (determined by equation (8)) **(f)** mass map.

Fig. 5 The reconstructed tomogram of the full Au wedge crystal based on the combination of focal map in figure 3(c) and the mass map in figure 4(f)