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An alternative approach to determine attainable resolution directly from HREM images

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ABSTRACT

The concept of resolution in high-resolution electron microscopy (HREM) is the power to resolve neighboring atoms. Since the resolution is related to the width of the point spread function of the microscope, it could in principle be determined from the image of a point object. However, in electron microscopy there are no ideal point objects. The smallest object is an individual atom. If the width of an atom is much smaller than the resolution of the microscope, this atom can still be considered as a point object. As the resolution of the microscope enters the sub-Å regime, information about the microscope is strongly entangled with the information about the atoms in HREM images. Therefore, we need to find an alternative method to determine the resolution in an object-independent way. In this work we propose to use the image wave of a crystalline object in zone axis orientation. Under this condition, the atoms of a column act as small lenses so that the electron beam channels through the atom column periodically. Because of this focusing, the image wave of the column can be much more peaked than the constituting atoms and can thus be a much more sensitive probe to measure the resolution. Our approach is to use the peakiness of the image wave of the atom column to determine the resolution. We will show that the resolution can be directly linked to the total curvature of the atom column wave. Moreover, we can then directly obtain the resolution of the microscope given that the contribution from the object is known, which is related to the bounding energy of the atom. The method is applied on an experimental $CaTiO_3$ image wave.

1. Introduction

Originally the concept of resolution was defined by Lord Rayleigh [1] as the power to discriminate point objects, such as stars, with a telescope. In a sense, the resolution is related to the width of the point spread function of the telescope or the microscope. In [2,3], a review of various resolution definitions is given according to the transfer function in HREM. The most commonly used definition is the inverse of the information limit of the microscope [2,4], which is determined by the damping envelope incorporating effects of partial coherence. In electron microscopy, the smallest objects are atoms and because the electron interacts with their electrostatic potential, the atoms cannot be considered as ideal point objects. This poses no problem when the resolution of the instrument is much larger than the width of the atom as was the case in the past. In that case, one can determine the resolution from the diffractogram of an amorphous thin film which can, to some extent, be considered as a white noise object [5,6]. Nowadays, with advanced techniques and aberration correctors [7], the resolution of the microscope has been greatly improved to the sub-Å regime [8–11]. In this case, objects can no longer be considered as weak phase objects and dynamical scattering may become important, meaning the non-linear interaction may not be ignored [12]. The information about the microscope such as the resolution is evidently strongly entangled with the information about the atoms in the HREM images. Thus, the resolution cannot be defined independently of the object. Therefore, we must try to find an alternative method to determine the resolution in the image in an object-independent way. This means that the resolution in an image should be measured directly using the object under study.

In this work we propose to use the image wave of a crystal in zone axis orientation. In such a “channeling condition” [13] the atoms in a column act as small lenses that focus the electron wave. In this way the image wave amplitude can be much more sharply peaked than the width of the electrostatic potential of a single atom and this channeling occurs for light atoms as well as for heavy atoms. Furthermore, it has been shown in [15] that to a good approximation the image wave at the atom column position has a Gaussian shape. Our approach here is to use the shape of the image wave of an atom column in a zone axis condition to determine the resolution.

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The outline of this paper is as follows. In Section 2, an overview of the channelling theory is given. Next, in Section 3, the approach to determine the attainable resolution is derived. In Section 4, the influence of microscope lens aberrations on the attainable resolution is studied. Then, in Section 5, a practical example is given. Finally, in Section 6, conclusions are drawn.

2. The channelling theory

Due to the strong electrostatic potential of the atoms, an atom column in the direction of the electron beam acts as a channel for the incoming electrons in which the electrons scatter dynamically. An intuitive way of understanding this is to consider each atom as a thin lens so that as the electron wave passes through the atoms, it is focused at periodic distances [14].

It has been proven in [15] that when the electron beam leaves an isolated atom column, the image wave can be expressed to a good approximation as

$$\psi(\mathbf{r},z)=\psi(\mathbf{r},0)+c_s\phi_s(\mathbf{r})\left[\exp\left(-i\pi\frac{E_s}{E_0}\frac{1}{\lambda}z\right)-1\right], \tag{1}$$

where $\psi(\mathbf{r},0)$ is the incident wave, $\mathbf{r}$ describes the two-dimensional vector in the plane of the image wave which is perpendicular to the beam direction, and z is the column thickness (relates to the number of atoms in a column given the distance between atoms in the column is known). The incident beam energy is given by E_0 and λ is the wavelength. The function $\phi_s(\mathbf{r})$ is the lowest energy bound state, the s-state, with E_s its energy. The s-state function can be approximated by a quadratically normalized and parameterized Gaussian function [15]

$$\phi_s(\mathbf{r})=\frac{1}{a\sqrt{2\pi}}\exp\left(-\frac{r^2}{4a^2}\right) \tag{2}$$

with a the column dependent width and $r=|\mathbf{r}|$. The column width is related to the energy of the s-state and is larger for a light atom column type and smaller for a heavy atom column type within the range of 0.1 Å to 0.5 Å. The excitation coefficient c_s is given by

$$c_s=\int\phi_s^{\star}(\mathbf{r})\psi(\mathbf{r},0)\,d\mathbf{r}, \tag{3}$$

where the symbol $\star$ denotes the complex conjugate. For an incident plane wave, i.e. $\psi(\mathbf{r},0)=1$, c_s equals $2\sqrt{2\pi}a$ when using Eq. (2). Eq. (1) is also referred to as the exit wave of an atom column to distinguish between the wave at the exit surface of the object and the wave at the image plane. Later, in Section 5, the reconstructed wave will be referred to as the experimental exit wave. A detail description will be given then.

3. Determination of the attainable resolution

As presented in [16], a complex plane, the so-called Argand plot, can be used to derive the structure parameters, such as the number of atoms in a column and the column height, using the pixel value at the atom column position of the image wave (or exit wave). Similarly, instead of plotting only the pixel at the column position, we can plot all the pixel values of an atom column wave in the Argand plot. According to the channelling theory, all pixels of a single atom column wave are located on a straight line in the Argand plot since they have the same phase (refer to Eq. (1) minus the incident beam wave $\phi(\mathbf{r},0)$).

An example is simulated for an isolated Au atom column with $z=16$ Å, $a=0.13$ Å and $E_s=-210.8$ eV [17]. The incident beam energy is 300 keV. Here we assume that the microscope is free of lens aberrations. Therefore, the resolution derived is solely contributed from the atom column. The amplitude (or magnitude) of the image wave is shown in Fig. 1(a). All pixels of the image wave lie on a solid straight line in the Argand plot as shown in Fig. 1(b). If the incident beam wave is removed, meaning that the plot passes through the origin $(0,0)$, this shows that the phase of every point in the exit wave is constant as stated above. When the image wave is defocused, meaning that the image plane is at a distance ε to the focal plane, the pixel values of the atom column will form a curve as shown by means of dotted lines in Fig. 1(b). The effect of defocus on the image wave can mathematically be described as a convolution product of Eq. (1) and a defocus propagator (see Appendix). Pixels closer to the column position give larger position changes in the Argand plot. This difference of position changes can also be explained from the fact that pixels closer to the column position contain more higher spatial frequency information. Thus, Fig. 1(b) also shows different defocus phase changes on the spatial frequencies contributing to the column image wave. It is observed in Fig. 1(b) that there are two stationary points (dotted circles), meaning that they do not change with defocus. One is at (1, 0) which is the constant background from the incident beam wave and the other one is at where the total curvature of the column wave equals zero. Note that here the surface of the column wave is defined in the three-dimensional space (bell-shaped function). The latter point will be used to define the attainable resolution. Note that this point is not strictly stationary but is a point with very little variance. This will be explained in detail in Section 4.1. In what follows, we will derive the resolution mathematically.

Substituting Eqs. (3) and (2) into Eq. (1), we can derive the curvature of the column wave using the Laplacian operator Δ

$$\begin{aligned}\Delta\psi&=\frac{\partial^2\psi}{\partial x^2}+\frac{\partial^2\psi}{\partial y^2}\\&=c_s\frac{1}{a\sqrt{2\pi}}\exp\left(-\frac{r^2}{4a^2}\right)\left[\exp\left(-i\pi\frac{E_s}{E_0}\frac{1}{\lambda}z\right)-1\right]\left(\frac{r^2}{4a^4}-\frac{1}{a^2}\right).\end{aligned} \tag{4}$$

Eq. (4) equals zero either when r approaches a large value or when $r=2a$. The former case corresponds to the stationary point representing the background while the latter case corresponds to the other stationary point. Thus, at

$$r=r_{\Delta=0}=2a, \tag{5}$$

the total curvature of the image wave of an atom column equals zero within a certain defocus range (see Section 4.1).

A common definition of resolution was given by Lord Rayleigh in 1879 [1]. Rayleigh stated that the resolution is the minimum resolvable distance in the sense that two point sources are just resolvable when the central maximum of one source coincides with the first zero of the other one. The Rayleigh resolution is thus given by the distance from the central maximum to the first zero of a point spread function. The Rayleigh resolution criterion can be generalized as the distance for which the ratio of the value at the central dip of composite point spread functions to the value at the central maximum of the point spread function is equal to 0.8. This corresponds to the original Rayleigh resolution for a rectangular aperture. Following this approach, the Rayleigh resolution ρ_p can be derived mathematically from Eq. (1)

$$0.8=2\exp\left(-\frac{\rho_p^2}{16a^2}\right), \tag{6}$$

from which it follows that:

$$\rho_p\approx 4a=2r_{\Delta=0}. \tag{7}$$

As a result, the resolution is directly related to the width of the atom column a in the image wave. However, the width a in an experimentally reconstructed wave is strongly influenced by lens aberrations such as defocus, spherical aberration, astigmatism and so on. Thus, it seems more appropriate to determine the resolution