The tuning of a Zernike phase plate with defocus and variable spherical aberration and its use in HRTEM imaging

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Abstract

With the advent of the double-hexapole aberration corrector in transmission electron microscopy the spherical aberration of the imaging system has become a tunable imaging parameter like the objective lens defocus. Now Zernike phase plates, altering the phase of the diffracted electron wave, can be approximated more perfectly than with the lens defocus alone, and the amount of phase change can be adjusted within wide limits. The tuning of the phase change allows an optimum contrast transfer in high-resolution imaging even for thick crystalline objects, thus surpassing the limits of the well-known Scherzer $\lambda/4$ phase plate to the imaging of thin objects. The optimum values for the spherical aberration and the lens defocus are derived, and the limits and imperfections of the approximation explored. A mathematical link to the channelling approximation of high-energy electron diffraction shows how the image contrast of atomic columns can be improved systematically within wide thickness limits. Depending on the specimen thickness different combinations of spherical aberration and defocus are favourable: positive spherical aberration with an underfocus, zero spherical aberration with zero defocus, as well as negative spherical aberration with an overfocus.

Keywords: Transmission electron microscopy; High-resolution imaging; Aberration correction; Spherical aberration; Zernike phase plate; Optimum imaging

1. Introduction

In his paper from 1949 Otto Scherzer applied the phase-contrast method, devised by Frits Zernike in light optics [1,2], to the optimum imaging of thin specimens in electron microscopy [3]. He derived an optimum defocus, now called Scherzer’s defocus, for the given fixed and large spherical aberration of an electron microscope, in order to approximate a $\lambda/4$ phase plate by means of lens aberrations. This Scherzer phase plate has a serious shortcoming, which is particularly distinct in modern high-resolution electron microscopy reaching information limits of 0.1 nm and better: the appertaining point resolution is poor compared with the information limit.

Spherical aberration has now become a tunable imaging parameter through the development and integration of a double-hexapole aberration corrector into a transmission electron microscope, the Philips CM200 FEG ST installed at Jülich [4–8]. The compensating lens elements allow an adjustment of the spherical aberration from the value of the uncorrected instrument to small positive values, over zero, even towards negative values. The combination of small values of the spherical...
aberration with small lens defoci has given a new freedom for the construction of more perfect phase plates [9].

The imaging properties of three special phase plates have already been thoroughly investigated: the \( \frac{\lambda}{4} \) phase plate producing positive phase contrast, the \(-\frac{\lambda}{4}\) phase plate producing negative phase contrast, and the zero phase plate, representing perfect aberration correction [9,10]. In particular, the use of the \(-\frac{\lambda}{4}\) phase plate has proven to produce a high image contrast of thin crystalline objects, which made it possible to investigate even the occupancy of single oxygen columns in \( \text{SrTiO}_3 \) and \( \text{YBa}_2\text{Cu}_4\text{O}_7 \) [10,11].

In this article we follow the convention introduced by Zernike: advancing the scattered wave relative to the unscattered wave by a phase plate with positive values is called positive phase contrast. Then, in high-resolution electron microscopy, a Scherzer phase plate generated by positive spherical aberration and an underfocus provides dark-atom contrast of a thin specimen.

The present work explores now the construction of phase plates with an arbitrary phase change by the tuning of defocus and spherical aberration together, as well as the limits of the approximation of these Zernike phase plates by the two chief aberrations. The adaptation of an arbitrary phase change proves to be useful for the imaging of atom columns in crystalline objects of arbitrary thickness, where the propagation of the incident electron wave through the crystal is governed by electron channelling. In the second part of this work we repeat briefly a commonly used model of electron channelling, derive the phase of the diffracted wave with respect to the direct wave, and show how this phase can be combined with the phase of a tuned Zernike phase plate in order to attain a high image contrast from the columnar structure of crystalline objects.

2. Construction of a Zernike phase plate

Let the electron wave \( \psi(\vec{r}) \) entering the imaging system of the microscope be composed of the direct, unscattered part \( \psi_0 \) and the scattered part \( \psi(\vec{r}) - \psi_0 \). If a perfect Zernike phase plate [1,2] adds to the phase of the diffracted wave another constant phase \(-2\pi Z_0\), through a constant aberration \( Z_0 \) for the scattered wave, then the electron wave in the image plane is altered to

\[
\psi'(\vec{r}) = \psi_0 + (\psi(\vec{r}) - \psi_0)e^{-2\pi Z_0}.
\]

With the electron wavelength \( \lambda \), the scattering vector \( \vec{g} \), and the two chief aberrations defocus \( Z \) and spherical aberration \( S \), a real aberration function

\[
\chi(\vec{g}) = \frac{1}{2} Z \lambda g^2 + \frac{1}{2} C_S \lambda^3 g^4
\]

takes already the needed value of zero for the unscattered wave, \( \vec{g} = 0 \). Using the least-squares criterion

\[
\int |\chi(\vec{g}) - Z_0|^2 d\vec{g} \rightarrow \min,
\]

where the integration is performed over a circular aperture with radius \( g_{\text{max}} \), the best approximation for the constant aberration \( Z_0 \) is obtained for the defocus

\[
Z = \frac{8Z_0}{\lambda g_{\text{max}}^2}
\]

and the spherical aberration

\[
C_S = -\frac{40Z_0}{3\lambda^3 g_{\text{max}}^4}.
\]

The quality of the approximation is determined now using the \( \pi/4 \) criterion, which demands a deviation of the phase plate by less than \( \lambda/8 \). If

\[
|Z_0| < \frac{\lambda}{8}
\]

then the phase deviates by less than \( \pi/4 \) at the extremum of the aberration function. Fortunately this limit is wide enough to cover the important interval \(-\frac{\lambda}{8} < Z_0 < \frac{\lambda}{8} \), representing a full cycle!

A lower bound to the spatial frequency is given by

\[
g_{\text{low}} = g_{\text{max}} \sqrt{\frac{3}{5} - \sqrt{\frac{15 + 24|Z_0|}{400|Z_0|}}}.
\]

if \( |Z_0| > \frac{\lambda}{8} \), else \( g_{\text{low}} = 0 \). An upper bound to the spatial frequency is given by

\[
g_{\text{high}} = g_{\text{max}} \sqrt{\frac{3}{5} + \sqrt{\frac{15 + 24|Z_0|}{400|Z_0|}}}
\]
Within the interval given by \( g_{\text{low}} \) and \( g_{\text{high}} \) the \( \pi/4 \) criterion is fulfilled, and with the help of the diagram shown in Fig. 1 two properties of the phase plate can be seen: the upper bound is matching quite closely the information limit, but the lower bound cuts off a more or less large part of the small spatial frequencies, depending on the choice of \( w_0 \): The latter is a property of all phase plates which are mimicked by lens aberrations, because these follow power laws of the coordinate \( g \).

With a given choice of \( w_0 \) and the appertaining setting of defocus and spherical aberration, following Eqs. (4) and (5), the delocalisation in the image plane can be calculated. It describes the “blurring” of object points in the image plane, and it is given by the largest ray displacement \( \vec{R}(\vec{g}) = \nabla \phi(\vec{g}) \) of any ray inside the aperture [12,13]. Considering again a circular aperture with radius \( g_{\text{max}} \) the appertaining delocalisation is

\[
R = \frac{16|Z_0|}{3 g_{\text{max}}}. \tag{9}
\]

Now we have control over an improved Zernike phase plate which imposes, within the limits shown, an arbitrary and fairly constant phase on the scattered part of an electron wave.

### 3. Exit wave of a crystalline object

Let us now apply the improved Zernike phase plate to an exit wave leaving a crystalline object after electron scattering. The calculation will show how the arbitrary aberration \( w_0 \) has to be chosen in order to provide optimum high-resolution imaging. First, we repeat briefly a model of electron scattering in the framework of the high-energy channelling approximation.

The majority of high-resolution materials-science investigations are performed using low-indexed crystal orientations with respect to the incident electron wave, where single atomic columns can be resolved. In this situation the electron wave propagating through the crystal is modified by the local arrangement of atomic columns, and the individual scattered partial waves have corresponding local modulations. Then the mathematical treatment of electron scattering can be carried out atom column by atom column. A commonly used model of electron channelling by a single atom column [14–20], here in the form of Lentzen and Urban [18], leads to the wave function

\[
\psi(\vec{r}) = \cos \frac{\pi z}{\xi} + i \left( \frac{U(\vec{r}) \xi}{K} - 1 \right) \sin \frac{\pi z}{\xi} \tag{10}
\]

with the direct, unscattered part

\[
\psi_0 = \cos \frac{\pi z}{\xi} + i \left( \frac{U_0 \xi}{K} - 1 \right) \sin \frac{\pi z}{\xi} \tag{11}
\]

and the scattered part

\[
\psi(\vec{r}) - \psi_0 = i \left( \frac{U(\vec{r}) - U_0}{K} \xi \right) \sin \frac{\pi z}{\xi}. \tag{12}
\]

The model contains the projected electrostatic potential \( V(\vec{r}) \) via \( U(\vec{r}) = 2m e / h^2 V(\vec{r}) \), with the vacuum chosen as the reference, the mean inner potential \( U_0 \), the inverse electron wavelength \( K \), the specimen thickness \( z \), the extinction distance \( \xi \), the relativistically corrected electron mass \( m \), the elementary charge \( e \), and Planck’s constant \( h \). The model is valid for atoms with small or medium charge number: then the channelling can be well described by the superposition of two eigenstates of electron diffraction, which leads to the characteristic beating with a thickness period equal to the extinction distance \( \xi \). Comparing Eq. (10) with the form used in Lentzen and Urban [18] a term \( 2|z_1|^2 \), referring to the excitation of eigenstates, could be omitted here, because the vacuum was chosen as a reference for the potential. In Eq. (10),
a suitable common phase of the wave function $\psi(\vec{r})$ has been chosen in order to make the scattered part (12) purely imaginary, which simplifies further calculations.

Inserting Eq. (10) into (1), and with the abbreviation

$$c = \frac{U_0 \xi}{K} - 1,$$

(13)

the image intensity for coherent illumination becomes

$$I(\vec{r}) = |\psi(\vec{r})|^2 = \cos^2 \frac{\pi z}{\xi} + c^2 \sin^2 \frac{\pi z}{\xi} - 2 \frac{(U(\vec{r}) - U_0) \xi}{K} \sin \frac{\pi z}{\xi} \times \text{Im} \left( \left( \cos \frac{\pi z}{\xi} - ic \sin \frac{\pi z}{\xi} e^{-2ni\lambda_0} \right) \right) + \left( \frac{U(\vec{r}) - U_0) \xi}{K} \right)^2 \sin^2 \frac{\pi z}{\xi}. \quad (14)$$

The first part is the intensity of the direct wave; it provides a uniform background in the image plane. The second part is the linear interference between the direct and the scattered wave; it is linear with respect to the projected crystal potential and depends on the aberration $\lambda_0$ of the Zernike phase plate. The third part is the intensity of the scattered wave; it is quadratic with respect to the projected crystal potential.

A quantitatively more correct model would include the effects of a partially coherent illumination [21], that is the linear and non-linear damping envelopes, and the imperfections of the Zernike phase plate approximated by defocus and spherical aberration. Then, the whole calculation had to be performed in reciprocal space, and in particular the non-linear part of the image intensity would become less clearly interpretatable in real space than in the simplified treatment used above. With the expression for the image intensity in real space for coherent illumination (14) we gain here a clear light on the contrast mechanism investigated in this work: the mutual superposition of the linear and non-linear part of the image intensity. Later, in the simulation study, we will apply, of course, the complete imaging model including partially coherent illumination.

4. Adaptation of the Zernike phase plate

In the following section a favourable choice for the aberration is derived with respect to an optimum image of the column structure. From Eq. (14) we can see that both the linear and the quadratic part of the image intensity peak at the atom column position. The quadratic part is always positive; modulus and sign of the linear part, however, are determined by the term $\text{Im}(\ldots)$ in Eq. (14). Obviously, the strongest contrast modulation of the image intensity at the column position can be obtained, if the modulus of $\text{Im}(\ldots)$ takes a maximum and if its sign is opposite to the sign of $\sin(\pi z/\xi)$.

Both conditions are met by the solution

$$\tan 2\pi \lambda_0 = \frac{1}{c \tan \frac{\pi z}{\xi}} \quad (15)$$

and

$$\tan \frac{\pi z}{\xi} \sin 2\pi \lambda_0 > 0, \quad (16)$$

for the desired aberration interval $-\frac{1}{2} < \lambda_0 < \frac{1}{2}$. Then the image intensity (14) takes the form

$$I(\vec{r}) = \left( \sqrt{\cos^2 \frac{\pi z}{\xi} + c^2 \sin^2 \frac{\pi z}{\xi}} + \frac{(U(\vec{r}) - U_0) \xi}{K} \left| \sin \frac{\pi z}{\xi} \right| \right)^2, \quad (17)$$

and atom columns show up bright with respect to the mean intensity.

The prescription to obtain a strong contrast modulation runs now as follows: Determine the parameters $U_0$, $\xi$, $K$, $z$, and calculate the constant $c$ using Eq. (13). Then solve Eq. (15) for $\lambda_0$, and select the solution fulfilling Eq. (16). Finally, Eqs. (4) and (5) give the optimum setting for defocus and spherical aberration, once the information limit is given.

5. Dependence of the optimum phase plate on the specimen thickness

The previous section has given the mathematical framework for applying the optimum Zernike phase plate, and now we have all means to
demonstrate an important aspect for materials science work, that is its dependence on the specimen thickness.

Because the electron diffraction channelling has a thickness period equal to the extinction distance $\xi$, the image intensity has a thickness period of $\xi$ as well. Eqs. (15) and (16) show: the phase of the scattered wave with respect to the direct wave starts at $\pi / 2$, remains positive in the first half of the extinction period, and turns negative in the second half of the extinction period, approaching $-\pi / 2$ at the extinction distance. For $c < 0$ the phase increases in the first half of the extinction period from $\pi / 2$ to $\pi$, and in the second half from $-\pi$ to $-\pi / 2$. For $c > 0$ the phase decreases in the first half of the extinction period from $\pi / 2$ to 0, and in the second half from 0 to $-\pi / 2$.

Conditions (15) and (16) determine how the Zernike phase plate compensates the phase of the scattered wave favourably: for the first half of the extinction period $\chi_0$ shall be positive, for the second half it shall be negative. Hence the approximation of the Zernike phase plate by defocus and spherical aberration, Eqs. (4) and (5), yields an overfocus combined with a negative spherical aberration for the first half of the extinction distance, and an underfocus combined with a positive spherical aberration for the second half of the extinction distance. Approaching a specimen thickness just smaller than half the extinction distance the favourable compensation is achieved for $c < 0$ with a defocus

$$Z = \frac{1}{2\lambda g_{\text{max}}^2}$$

and a spherical aberration

$$C_S = -\frac{20}{3\lambda^3 g_{\text{max}}^4}.$$  

For a specimen thickness just larger than half the extinction distance the favourable compensation is achieved with

$$Z = -\frac{1}{2\lambda g_{\text{max}}^2}$$

and

$$C_S = \frac{20}{3\lambda^3 g_{\text{max}}^4}.$$  

If, at last, $c > 0$, then the favourable compensation at half the extinction distance is achieved with $Z = 0$, and $C_S = 0$. The favourable compensation by the Zernike phase plate is illustrated for both situations, $c > 0$ and $c < 0$, by the plots shown in Fig. 2.

6. Simulation study

Three approximations have been used in the above analytical treatment in order to derive a favourable adaptation of the contrast transfer of the microscope with respect to the exit wave function due to a crystalline object: the channelling model of high-energy electron diffraction [15]; the replacement of the true phase plate, generated by lens aberrations, by an ideal Zernike phase plate.
with a constant aberration for the scattered wave; and the assumption of a fully coherent illumination for the imaging process.

The following simulation study shall demonstrate that even if the full quantum-mechanical optical treatment of electron diffraction, using the multislice algorithm [22], and contrast transfer under partially coherent illumination is applied [21], the proper choice of defocus and spherical aberration still yields a favourable superposition of the linear and non-linear part of the image intensity. In particular, an investigation of the image contrast due to the crystal structure for different specimen thicknesses illustrates the benefit of tuned phase plates for materials science investigations.

As an example for a simple crystal structure we chose Ge [110], imaged with the aberration-corrected Philips CM200 FEG ST at an acceleration voltage of 200 kV, yielding a wavelength of $\lambda = 2.51 \text{ pm}$ and a reciprocal wavelength of $K = 398.7 \text{ nm}^{-1}$. Then the extinction length of Ge [110] is $\xi = 14.8 \text{ nm}$, and the constant $c = -0.57$. We assume an information limit of the microscope of $g_{\text{max}} = 8 \text{ nm}^{-1}$. Fig. 3 displays a through-thickness series of images of Ge [110] for the specimen thicknesses listed in Table 1, which also lists the favourable aberrations $\chi_0$ and the appertaining defocus and spherical aberration values, according to Eqs. (4), (5), (13), (15), and (16). All image calculations were carried out with the EMS simulation package [23].

The through-thickness series of images shows: The proper choice of defocus and spherical aberration yields for all thicknesses, covering a complete extinction distance, a favourable superposition of the linear and non-linear contributions to the image intensity. The atom columns show up bright with respect to the mean intensity, because the strongest contrast modulation is obtained if the direct wave and the scattered wave are imaged with the same phase. At a thickness equal to half the extinction length, Fig. 3C, extra spots appear at the tunnel positions and faint spots close to the dumbbell contrast at the column positions.

In the first half of the extinction distance, cases (A) and (B) in Table 1, the optimum aberration $\chi_0$ takes positive values, corresponding to an
optimum overfocus and an optimum negative spherical aberration; in the second half of the extinction distance, cases (D) and (E) in Table 1, the optimum aberration \( \xi_0 \) takes negative values, corresponding to an optimum underfocus and an optimum positive spherical aberration. Close to half the extinction distance, case (C) in Table 1, the optimum aberration \( \xi_0 \) approaches \(-\frac{1}{2}\), corresponding to an underfocus and a positive spherical aberration. The comparison of the image displayed in Fig. 3C with the image displayed in Fig. 4, simulated for \( z = 7.6 \text{ nm}, Z = 24.3 \text{ nm}, \) and \( C_S = -100.7 \text{ \mu m} \), shows: at a thickness close to half the extinction distance the optimum aberration can be reversed, resulting in the same image intensity. All these observations correspond to the description given in the previous Section for \( c < 0 \).

Finally, the superposition of the linear and non-linear contribution to the image intensity can be investigated more closely by comparing the non-linear image with the linear image, for example for the case (B) in Table 1 and the appertaining image (B) displayed in Fig. 3. Fig. 5 displays the appertaining linear image, together with the difference image of the non-linear and the linear image, representing the pure non-linear contribution of the image intensity: Both parts exhibit maxima at the atom column positions, and hence the linear part reinforces the non-linear part.

A close look at the linear and non-linear parts of the image intensity reveals the origin of the extra spots, which disturb the direct structure image at half the extinction distance. The extra spots at the tunnel positions are present in both parts, but they are more pronounced in the non-linear part. The faint spots close to the dumbbell contrasts appear only in the non-linear part. In the image displayed in Fig. 3C the direct view of the column structure is more disturbed than in case (B), because here the

### Table 1

| Specimen thicknesses \( z \) (nm), defoci \( Z \) (nm), and spherical aberrations \( C_S \) used in the image simulation study, together with the aberrations \( \xi_0 \) approximated by the phase plate |
|---|---|---|---|
| \( z \) (nm) | \( \xi_0 \) | \( Z \) (nm) | \( C_S \) (\( \mu m \)) |
| A | 0.8 | 0.266 | 13.2 | −54.8 |
| B | 4.0 | 0.342 | 17.0 | −70.5 |
| C | 7.6 | −0.488 | −24.3 | 100.7 |
| D | 11.2 | −0.330 | −16.4 | 68.0 |
| E | 14.0 | −0.266 | −13.2 | 54.8 |

Negative \( Z \) refers to underfocus; letters refer to the images displayed in Fig. 3.
direct wave takes a minimum amplitude and the scattered wave takes a maximum amplitude. In this situation the non-linear part of the image intensity is strongest with respect to the linear part, which lets the extra spots show up more distinctly.

At least the extra spots at the tunnel positions can be explained through the imperfect contrast transfer of the electron microscope at small spatial frequencies. At half the extinction distance of Ge [110] the favourable aberration for strong image contrast is $\frac{1}{2}$ or $-\frac{1}{2}$, but with the help of Eq. (8), and the plot displayed in Fig. 1, we find that the spatial frequency of the important {111} beams falls already below the lower limit of the phase plate. In other words, the {111} beams are transferred with a wrong phase, which prevents the proper superposition of the partial waves in the image plane with respect to a direct structure image. On the other hand, no other compromise for the phase plate can be made, because then the high-frequency components of the scattered wave would be wrongly transferred.

7. Discussion

The limitations of the models used in this analytical treatment have been mentioned already. It remains now to discuss a few details regarding electron diffraction, imaging, and hints for the practical work with more complicated crystal structures.

The channelling model of high-energy electron diffraction used in this work assumed the excitation of two eigenstates of the Schrödinger equation, which is good enough to describe the scattering by columns comprising atoms of small or medium charge number [15]. If more eigenstates are excited, for example for Au [111], then the superposition of these partial waves will lead to a complicated beating and the phase of the diffracted wave will be no more constant with respect to the direct, unscattered wave [15]. A compromise for a proper choice of the aberration of the Zernike phase plate, $x_0$, and the appertaining setting for defocus and spherical aberration can then only be found, if either the eigenenergies of the additional states do not differ too much or the specimen thickness is not too large. Then the dispersion of the additional partial waves as a function of the specimen thickness is not too large, and the partial waves may be arranged in two groups. The beating between the two groups of eigenstates may then be described again by a single extinction length.

A similar situation occurs, if more complicated crystal structures comprising columns with different charge per unit length are considered. There, also a dispersion of partial waves occurs, owing to the individual columns, and an optimum aberration of the Zernike phase plate can be found, if the eigenstates may be arranged in two groups.

The channelling model of high-energy electron diffraction contains the parameter $c$, Eq. (13), which determines the relative phase between the direct, unscattered wave and the scattered wave. Depending on the sign of $c$ the relative phase increases or decreases with the specimen thickness within the first extinction distance, as has been described above. Many practical examples show: at an accelerating voltage of 200 kV the parameter is negative for many crystal structures. Materials with a high electrostatic potential $U$ have in general short extinction distances $\xi$ for closely packed atomic columns, and materials with low potential long extinction distances. Therefore, the product $U_0\xi$ varies within certain limits, but not strongly enough to change the sign of $c$. Therefore we have investigated in the simulation study above, on Ge [110], only the case $c<0$.

Electron diffraction and imaging are in this work well described within a wide range of specimen thicknesses, covering the important range of the first extinction distances, and a wide range of aberrations, covering the full cycle from a $\lambda/2$ to a $-\lambda/2$ phase plate. It is now worth to look at the familiar limit of small specimen thicknesses. Then conditions (15) and (16) yield an optimum aberration of

$$x_0 = \frac{1}{4},$$

representing a $-\lambda/4$ phase plate, which produces negative phase contrast. Eqs. (4) and (5) yield an
optimum overfocus of
\[ Z = \frac{2}{\lambda g_{\text{max}}} \]
and an optimum negative spherical aberration of
\[ C_S = -\frac{10}{3 \lambda^3 g_{\text{max}}^4}. \]

These expressions are very similar to the expressions for the optimum setting, extending the point resolution to the information limit, given in our former work [9]. The slight difference of the coefficients arises from the different criteria used: in this work the least-squares criterion (3) is the starting point, whereas in the former treatment [9], the traditional formula for Scherzer’s point resolution was exploited [3]. There is, of course, a certain arbitrariness in choosing a criterion for an “optimum” aberration function yielding an “optimum” imaging of a thin object. In this work, we stressed the intention to find a uniform aberration \( w_0 \) up to the information limit; in our former work we stressed the intention to find a point resolution as large as possible.

The image intensity for a small specimen thickness becomes
\[ I(\vec{r}) = \left( 1 + \pi \frac{U(\vec{r}) - U_0}{K} z \right)^2 - (1 - c^2) \times \left( \frac{\pi^2}{\xi} \right)^2 + O(z^2), \]
where the first term describes the familiar imaging in the framework of the weak phase object approximation, using an amplitude of 1 for the direct wave, and the second term accounts for the small loss of intensity of the direct wave, because we expanded intensity (17) to second order in \( z \). The mean inner potential \( U_0 \) appears explicitly, because we used throughout this work the vacuum as a reference for the electrostatic potential, and not the mean potential. The negative phase contrast due to the \(-\lambda/4\) phase plate has changed the scattered wave to a real and positive value, and exactly as in the full solution (17) atom columns show up bright with respect to the mean intensity.

Next, we touch an important aspect for practical work with variable Zernike phase plates: that is the knowledge of the parameters linked to the object structure, namely the mean potential, the extinction length, and the specimen thickness. These parameters govern the optimum setting of defocus and spherical aberration via the optimum aberration \( x_0 \). The mean potential and the extinction length of a certain material can be determined prior to the experiment, whereas the specimen thickness at a desired object location is generally unknown; the thickness may even vary over the field of view, in particular if standard specimen preparation techniques are used, which produce wedge-shaped specimens.

The problem may be solved by taking an image series with variable aberration \( x_0 \), from \( x_0 = -\frac{1}{2} \) to \( \frac{1}{2} \), sampled at an equal distance of, say, \( \Delta x_0 = \frac{1}{8} \). It was already shown above: in the practically important case \( c < 0 \) the optimum phase change applied to the scattered wave covers the whole cycle, from \(-\pi\) to \( \pi \), if the specimen thickness varies over the full extinction distance. The “through-aberration” image series contains now all desired phase changes, and hence the optimum setting can be found after the experiment for each object location. Eq. (17) sets the criterion for the selection of each of the optimum images: it is the one with the strongest atom column contrast.

Today, control and adjustment of the lens defocus of a high-resolution instrument is viable within the precision of a \( \pi/4 \) phase change for spatial frequencies at the information limit. With the aberration-corrected Philips CM200 FEG ST, installed at Jülich, the adjustment of a positive or negative spherical aberration is viable within the \( \pi/4 \) limit as well [24], but quick adjustments, as quick as a defocus change, are not possible due to the hardware lay-out of the correcting lens elements and due to the state of the lens control program, which was not designed for this task. Future lay-outs and software improvements, however, will undoubtedly pave the way for the application of through-aberration image series.

8. Conclusions

The new freedom to tune the spherical aberration of a transmission electron microscope...
provides together with the tunable objective lens defocus a means to approximate a Zernike phase plate imposing an almost arbitrary aberration on the scattered electron wave. The available range of positive and negative aberrations covers a complete cycle, and the imperfections of the real phase plates turned out to be not too serious with respect to the high-resolution imaging of crystalline objects.

Depending on the specimen thickness of a crystalline object, a unique aberration can be found which maximises the image contrast of single atomic columns. The optimum value for the aberration of the scattered electron wave was derived under three assumptions: (1) electron diffraction was described by the electron channeling model including two excited states; (2) the phase plate was assumed to have a perfect, constant aberration for the scattered wave; (3) the image intensity was calculated for coherent illumination. The simulation study on the imaging of Ge [110] showed: even if real lens aberrations and a partially coherent illumination are considered, the optimum setting of the lens defocus and the spherical aberration still provides an image intensity with a high contrast of individual atom columns.

Neither positive nor negative spherical aberration is alone important for the optimum imaging in a real materials science investigation, but the whole interval from −100 to 100 μm, at an acceleration voltage of 200 kV and an information limit of 8 nm⁻¹. Already the thickness variation of a sample, in the range of the first extinction length, requires an individual adaptation of the aberrations from positive to negative values.

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