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Title: Imaging with aberration-corrected low energy electron microscopy  
Issue Date: 2013-04-25
We introduce an extended Contrast Transfer Function (CTF) approach for the calculation of image formation in low energy electron microscopy (LEEM) and photo electron emission microscopy (PEEM). This approach considers aberrations up to fifth order, appropriate for image formation in state-of-the-art aberration-corrected LEEM and PEEM. We derive Scherzer defocus values for both weak and strong phase objects, as well as for pure amplitude objects, in non-aberration-corrected and aberration-corrected LEEM. Using the extended CTF formalism, we calculate contrast and resolution of one-dimensional and two-dimensional pure phase, pure amplitude, and mixed phase and amplitude objects. PEEM imaging is treated by adapting this approach to the case of incoherent imaging. Based on these calculations, we show that the ultimate resolution in aberration-corrected LEEM is about 0.5 nm, and in aberration-corrected PEEM about 3.5 nm. The aperture sizes required to achieve these ultimate resolutions are precisely determined with the CTF method. The formalism discussed here is also relevant to imaging with high resolution transmission electron microscopy.

3.1 **INTRODUCTION**

In recent years, low energy electron microscopy (LEEM) and photo electron emission microscopy (PEEM) have developed into powerful methods for the study of surfaces and interfaces in real time, with a spatial resolution of several nanometers [1, 2]. In LEEM imaging, the sample is illuminated with a coherent electron beam, and one of the reflected low energy electron diffraction (LEED) beams, aligned with the optical axis of the instrument, is selected for image formation. Using the (0, 0) LEED beam we obtain a bright field image, while use of any other LEED beam results in a dark field image. As the electron beam is reflected by the sample, it may undergo a change in amplitude, in phase, or both. As in any electron microscope, the electron wave front that is emitted by the sample is modified by the objective lens due to defocus, chromatic, spherical, and higher order aberrations, as well as cut-off in reciprocal space by the contrast aperture, often referred to as the diffraction limit. In order to fully understand image formation in a quantitative manner, a theory of LEEM image formation must take these modifications into account. In PEEM imaging, electrons emitted from the sample are incoherent, both in space and in time. Yet the same aberrations that affect coherent image formation also affect incoherent image formation, although not in quite the same manner.

Recently, Pang et al. [3] introduced a wave-optical approach to calculate image formation in LEEM. Using a Fourier optics formalism they calculated contrast and resolution of pure amplitude and pure phase objects in one dimension. Mixed phase and amplitude objects can also be treated with Fourier optics, but were not considered by Pang et al. In these calculations, only the third order spherical ($C_3$) and the lowest order chromatic ($C_C$) aberrations of the objective lens, diffraction cut-off at the aperture, energy spread and size of the electron source, defocus, as well as instabilities in lens current and voltage were considered. On the other hand, in the Transmission Electron Microscopy (TEM) community an alternative and essentially equivalent wave-optical formalism for image calculation using the Contrast Transfer Function (CTF) has been successfully applied for more than 30 years [4, 5]. The CTF considers the same imaging errors and modifications as the Fourier optics formalism used by Pang et al. [3]. It is an attractive approach because it facilitates image calculations that are particularly easy in the special case of weak phase objects, as appropriate for high resolution atomic imaging of thin objects by TEM. Nonetheless, the CTF formalism in its general formulation can also be applied to calculate image formation for arbitrary phase, amplitude, and mixed amplitude and phase objects that are routinely encountered in imaging with LEEM and PEEM.

In the present work, we apply the CTF formalism to image calculations for phase, amplitude, and mixed phase and amplitude objects in LEEM. Application
of the CTF formalism to LEEM has recently been explored [6]. Here, we expand
the formalism to include the effects of the chromatic and spherical aberrations
of the objective lens up to fifth order. This approach allows for calculating im-
age formation in state-of-the-art aberration corrected microscopes with improved
resolution. In these microscopes, the third order spherical and the lowest order
chromatic aberrations of the objective lens are compensated by an electron mirror
[7–9]. In this case, the contributions of the next higher order spherical and chro-
natic aberrations need to be taken into account. Finally, we consider the case of
incoherent imaging in PEEM. A modified CTF which models a spatially and tempo-
rally incoherent source is introduced and applied to calculate image formation in
PEEM for different emitted electron energy distributions with and without energy
filtering. Based on these calculations we show that the ultimate resolution in aber-
ration corrected LEEM is about 0.5 nm, and in aberration corrected PEEM is about
3.5 nm. While we focus our attention in this paper on LEEM/PEEM imaging, the
extension of the CTF formalism presented here is equally applicable to TEM imag-
ing in and beyond the weak phase approximation, with aberration correction.

### 3.2. The CTF formalism

A real instrument always introduces image modifications. For example, a point
in the object appears as an extended region in the image. This broadening can
be described by the so-called Point-Spread-Function (PSF) [5]. It describes how
information is transferred from the object to the image. The convolution of the PSF,
\( h(r) \), with the object wave function, \( f(r) \), yields the image wave function, \( \psi(r) \). The
observed image intensity is obtained by taking the squared modulus of the image
wave function, i.e. \( I = \psi \psi^* \).

The modifications introduced by the optical system of the microscope are best
described in the spatial frequency space. The spatial frequency is here approxi-
mated by \( q = \alpha / \lambda \), where \( \lambda \) is the electron wavelength and \( \alpha \) the emission angle.
Using the convolution theorem, the Fourier transform of the image wave function
is given by \( \Psi(q) = F(q) H(q) \), where \( F(q) \) and \( H(q) \) are the Fourier transforms of
\( f(r) \) and \( h(r) \). \( H(q) \) is called the Contrast Transfer Function (CTF) since it de-
scribes how information or contrast is transmitted from the object to the image.
The basic idea is to include all the relevant image modifications introduced by the
optical imaging system in the CTF. The CTF can be written as a product of all rele-
cant contributions imposed by the optical system [4, 5]

\[
H(q) = M(q) W(q, \Delta z) E_C(q, \Delta E) E_S(q) E_{\mu/
u}(q) 
\]  

(3.1)

Here, \( M(q) \) is the aperture function and accounts for the effects introduced by
the contrast aperture located in a diffraction plane. \( W(q, \Delta z) \) is the wave aber-
ration function which accounts for effects due to spherical wave aberrations and defocus, $\Delta z$. $E_C(q, \Delta E)$ is the chromatic envelope function that accounts for effects due to chromatic aberrations. $E_S(q)$ is the envelope function due to a finite source size and $E_{U/l}(q)$ is the envelope function caused by instabilities in the lens current and the lens voltage.

State-of-the-art electron microscopes use aberration correcting elements for improved resolution. In LEEM an electrostatic electron mirror is placed in the optical path after the objective lens [7, 8]. The mirror introduces spherical and chromatic aberrations with opposite sign compared to the objective lens. Therefore, the spherical and chromatic aberrations of the objective lens can be compensated to yield an imaging system with zero net third order spherical and lowest order chromatic aberrations, i.e. $C_3 = 0$ and $C_C = 0$, resulting in a better microscope resolution. Image calculations in aberration corrected microscopes require an extended CTF which takes higher order spherical and chromatic aberrations into account.

In LEEM the sample acts as the cathode in an electrostatic immersion lens. The front of the objective lens constitutes the anode. The incident electrons are decelerated by the electric field between the sample and the anode. Then, the back reflected electrons from the sample are reaccelerated by the same electric field. The electron energy with which electrons impact the sample and with which they also leave the sample can be adjusted by the offset between the potential of the sample and the electron gun and is referred to as the starting electron energy or the electron energy at the sample. The electron energy before deceleration and after reacceleration following reflection from the sample is given by the potential of the gun and is referred to as the nominal or the column electron energy. The column electron energy is typically $E = 15 – 20$ keV. The immersion lens produces a virtual image plane behind the real sample position [3, 6, 10]. This virtual image plane is then imaged by the remaining optics of the microscope. Here, the CTF formalism is applied to calculate LEEM images in an image plane with magnification $M = 1$.

Other effects that might cause additional image modifications include astigmatism, coma, and drift and vibrations of the sample. Since these effects usually can be minimized, they are not considered further here.

In the following sections, we describe each of the above mentioned terms of the CTF in more detail for standard and aberration corrected electron microscopes.

### 3.2.1 Aperture Function

The contrast aperture, located in a diffraction plane, limits the spatial frequencies used for image formation to a certain maximum value, $q_{\text{max}} = \frac{\alpha_{\text{max}}}{\lambda}$, where $\alpha_{\text{max}}$ is the maximum emission angle permitted by the aperture and $\lambda$ is the electron wavelength. A circular aperture can be represented by a simple boxcar function
which is equal to 1 for all spatial frequencies lower than $q_{\text{max}}$ and zero for all frequencies higher than that:

\[
M(q) = \begin{cases} 
1, & |q| < q_{\text{max}} \\
0, & |q| \geq q_{\text{max}} 
\end{cases}
\]  \hspace{1cm} (3.2)

Selection of aperture size that optimizes spatial resolution is discussed further in Section 3.3.3 on resolution.

### 3.2.2 WAVE ABERRATIONS

Imaging errors due to deviations of the wave path from an ideal reference wave are described by the wave aberration function, $W(q, \Delta z)$. There are two general sources causing wave aberrations for off-axis waves ($q > 0$), namely, spherical aberrations and defocus. Third order spherical aberrations cause electrons that travel further away from the optical axis to be focused stronger. This introduces an optical path length difference and corresponding phase shift that increases with increasing angle from the optical axis. Considering only defocus and third order spherical aberrations, the phase shift due to wave aberrations is expressed as [3–5]

\[
\chi(q, \Delta z) = \frac{1}{4} \left( C_3 \lambda^3 q^4 - 2 \Delta z \lambda q^2 \right) 
\]  \hspace{1cm} (3.3)

where $C_3$ is the third order spherical aberration coefficient of the objective lens and $\Delta z = \Delta f - \Delta a$ is the defocus with $\Delta f$ and $\Delta a$ being the deviations of focus and sample position, respectively, from ideal values (see Ref. [3]). The wave aberration function is calculated from the phase shift as follows [4, 5]:

\[
W(q, \Delta z) = \exp \left( i 2 \pi \chi(q, \Delta z) \right) 
\]  \hspace{1cm} (3.4)

Figure 3.1a shows the real and imaginary parts of the wave aberration function for standard LEEM at in-focus condition for aberration coefficients at starting electron energy of $E_0 = 10$ eV (see Section 3.2.7 on aberration coefficients). The frequency of the observed oscillations increases with increasing spatial frequency.

For image formation in aberration-corrected electron microscopes, the wave aberration function needs to include higher order spherical aberrations. The next higher order spherical aberration coefficient is the fifth order term $C_5$. These fifth order spherical aberrations cause an additional phase shift to $\chi(q, \Delta z)$ which is given by $\frac{1}{6} C_5 \lambda^5 q^6$ [11–13]. Therefore, the phase shift due to wave aberrations has a more general form

\[
\chi(q, \Delta z) = \frac{1}{4} \left( C_3 \lambda^3 q^4 + \frac{2}{3} C_5 \lambda^5 q^6 - 2 \Delta z \lambda q^2 \right) 
\]  \hspace{1cm} (3.5)
In the case of aberration-correction, i.e. $C_3 = 0$ and $C_C = 0$, this becomes

$$\chi(q, \Delta z) = \frac{1}{2} \left( \frac{1}{3} C_5 \lambda^5 q^6 - \Delta z \lambda q^2 \right)$$

It follows from raytracing calculations (see Ref. [14] and Section 3.2.7 on aberration coefficients) that the third order spherical aberration coefficient and the fifth order spherical aberration coefficient have positive sign, i.e. $C_3$ and $C_5$ cause phase shifts in $W(q, \delta z)$ with the same sign.

### 3.2.3 TEMPORAL COHERENCE

The imaging fidelity of a real instrument is further degraded by limited temporal coherence caused by a finite energy spread in the electron beam and instabilities in the current and high voltage of the objective lens. The energy spread of the electron beam introduces imaging errors due to chromatic aberrations of the objective lens. In other words, the focal length becomes energy dependent leading to deviations of electron trajectories from ideal. Considering an electron with an energy that deviates by $\epsilon$ from the nominal energy $E$, the lowest order chromatic aberration of the objective lens gives rise to an additional phase shift

$$\xi = \frac{1}{2} \epsilon C_C \lambda q^2$$

where $C_C$ is the chromatic aberration coefficient of lowest order ref. In the CTF
this leads to another phase factor given by [11–13]

\[ K(q, \epsilon) = \exp (i2\pi \xi) \] (3.6)

The energy distributions can generally be approximated by a Gaussian function centered at the nominal energy \( E \). The full-width at half-maximum (FWHM) of the Gaussian is equivalent to the energy spread of the electron source \( \Delta E = 2\sigma_E \sqrt{2\ln2} \). Electron emitters used in LEEM exhibit an energy spread typically in the range \( \Delta E = 0.25 - 0.75 \text{eV} \). Integration over the weighted contributions of the different energies within the Gaussian energy distribution of the source yields the chromatic envelope function first derived by Hanšen and Trepte [15]

\[ E_C(q) = \frac{1}{\sqrt{2\pi\sigma^2_E}} \int_{-\infty}^{\infty} K(q, \epsilon) \exp \left( -\frac{\epsilon^2}{2\sigma^2_E} \right) d\epsilon = \exp \left( -\frac{(\pi C_C \lambda q^2)^2}{16 \ln2} \left( \frac{\Delta E}{E} \right)^2 \right) \] (3.7)

An energy spread of the source causes strong damping in the transfer of the spatial frequencies from the diffraction plane to the image that is exponential in \( q^4 \). Fig. 3.1b shows a plot of \( E_C(q, \Delta E) \) and the corresponding CTF with a typical energy spread for a cold field emitter of \( \Delta E = 0.25 \text{eV} \). In Fig. 3.1c we compare the real parts of the CTFs with an energy spread of \( \Delta E = 0.25 \text{eV} \) and \( \Delta E = 0.75 \text{eV} \) for standard or non-aberration-corrected (nac) as well as for aberration-corrected (ac) LEEM (see below).

Instabilities in the lens current and voltage also lead to fluctuations in the lens focal length \( f \). The time-averaged effect of these fluctuations on the image formation is analogous to the modifications caused by the energy spread [15]. In the presence of current and voltage instabilities, the phase shift \( \xi \) becomes [4, 15]

\[ \xi = \frac{1}{2} \left( \frac{\epsilon}{E} + \frac{u}{U} + 2\frac{i}{I} \right) C_C \lambda q^2 \] (3.8)

with \( U \) and \( I \) being the nominal lens voltage and current, respectively, and \( u \) and \( i \) are the deviations from \( U \) and \( I \). The time-averaged current and voltage distributions are assumed to be Gaussian with FWHMs \( \Delta I = 4\sigma_I \sqrt{2\ln2} \) and \( \Delta U = 2\sigma_U \sqrt{2\ln2} \) (see Ref. [4]). Using Eq. 3.8, the phase factor for arbitrary but fixed energy, current, and voltage yields

\[ K(q, \epsilon, u, i) = \exp \left( i\pi \left( \frac{\epsilon}{E} + \frac{u}{U} + 2\frac{i}{I} \right) C_C \lambda q^2 \right) \] (3.9)
The integration over all three Gaussian weighting distributions yields

$$E_{C/U/I}(q) = \exp \left( -\frac{(\pi C_C \lambda q^2)^2}{16 \ln 2} \left[ \left( \frac{\Delta E}{E} \right)^2 + \left( \frac{\Delta U}{U} \right)^2 + \left( \frac{2 \Delta I}{I} \right)^2 \right] \right) = E_CE_UE_I$$  \hspace{1cm} (3.10)

Therefore, instabilities in the lens current and voltage give rise to additional exponential envelope functions $E_U$ and $E_I$ in the CTF. In modern LEEM instruments, the ratio $\Delta U/U = \Delta I/I$ is typically in the order of $10^{-6}$. This is about one order of magnitude smaller compared to $\Delta E/E \approx 1.7 \times 10^{-5}$.

Since the exponential envelopes in Eq. 3.10 scale with the squared of the ratios of $\Delta E/E$, $\Delta U/U$, and $\Delta I/I$, the impact of the envelope functions $E_U$ and $E_I$ on the CTF in LEEM is about a factor 100 – 1000 smaller compared to $E_C$. Hence, we will not further consider the minor effects of objective lens current and voltage instabilities on image formation for standard LEEM for the rest of the discussion.

Here, we consider the next two leading chromatic aberrations which comprise the aberration coefficients $C_{3C}$ and $C_{CC}$ [11–13]. The modifications caused by these aberrations in the case of a monochromatic source with electron energy deviation $\epsilon$ from the nominal energy $E$ are given by [11–13]

$$K_{3C/CC}(q, \epsilon) = \exp(i2\pi \xi_{3C/CC})$$

with

$$\xi_{3C} = \frac{1}{4} \frac{\epsilon}{E} C_{3C} \lambda^3 q^4$$

and

$$\xi_{CC} = \frac{1}{2} \left( \frac{\epsilon}{E} \right)^2 C_{CC} \lambda q^2$$

In analogy with the lowest order chromatic aberration term $C_C$, we sum up the weighted contributions for the different energies within a Gaussian energy distribution of the source to yield the effect of a polychromatic beam.

In a rigorous treatment, the chromatic aberration terms must be taken into account jointly, i.e. $K_{\text{total}}(q, \epsilon) = \exp(i2\pi \xi_{\text{total}})$ with $\xi_{\text{total}} = \xi_C + \xi_{CC} + \xi_{3C}$, because the integral of the product of the phase factor and the Gaussian energy distribution of the source
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\[ K_{\text{total}}(q, \epsilon) \exp \left( -\frac{\epsilon^2}{2\sigma^2} \right) \]

is not equal to the product of the separate integrals

\[ K_C(q, \epsilon) \exp \left( -\frac{\epsilon^2}{2\sigma^2} \right), \quad K_{3C}(q, \epsilon) \exp \left( -\frac{\epsilon^2}{2\sigma^2} \right) \]

and \[ K_{CC}(q, \epsilon) \exp \left( -\frac{\epsilon^2}{2\sigma^2} \right) \]

Carrying out the integral yields

\[
E_{\text{total}}(q) = \frac{1}{\sqrt{2\pi}\sigma^2} \int_{-\infty}^{\infty} K_{\text{total}}(q, \epsilon) \exp \left( -\frac{\epsilon^2}{2\sigma^2} \right) d\epsilon \\
= E_{CC}(q) \exp \left( -E_{CC}^2(q) \frac{\pi^2}{16\ln 2} \left( \frac{\Delta E}{E} \right)^2 \left( C_C \lambda q^2 + \frac{1}{2} C_{3C} \lambda^3 q^4 \right)^2 \right) \quad (3.11)
\]

where

\[
E_{CC}(q) = \left( 1 - i \frac{\pi \lambda}{4\ln 2} \left( \frac{\Delta E}{E} \right)^2 C_{CC} q^2 \right)^{-1/2} \quad (3.12)
\]

In order to get an understanding of the contributions of each of the two additional chromatic aberration terms to the CTF, we will consider them now separately. In the case of \( C_{3C} \) this yields

\[
E_{3C}(q) = \frac{1}{\sqrt{2\pi}\sigma^2} \int_{-\infty}^{\infty} K_{3C}(q, \epsilon) \exp \left( -\frac{\epsilon^2}{2\sigma^2} \right) d\epsilon \\
= \exp \left( -\frac{(\pi C_{3C} \lambda^3 q^4)^2}{64\ln 2} \left( \frac{\Delta E}{E} \right)^2 \right) \quad (3.13)
\]

Therefore, the \( C_{3C} \) aberrations lead to an exponential damping in \( q^8 \) for higher \( q \)-values that is similar to but stronger than the \( q^4 \) damping due to the \( C_C \) aberrations, Eq. 3.7. The modifications introduced by the \( C_{CC} \) aberration term is given by
\[ E_{CC}(q) = \frac{1}{\sqrt{2\pi}\sigma^2} \int_{-\infty}^{\infty} K_{CC}(q, \epsilon) \exp\left(-\frac{\epsilon^2}{2\sigma^2}\right) d\epsilon \]

\[ = \left(1 - i\gamma \frac{\pi}{4\ln 2} \left(\frac{\Delta E}{E}\right)^2 C_{CC} q^2\right)^{-1/2} = (1 - i\gamma q^2)^{-1/2} \quad (3.14) \]

This can be rewritten as

\[ (1 - i\gamma q^2)^{-1/2} = \left(1 + (i\gamma q^2)^2\right)^{-1/4} \exp\left(\frac{i}{2} \tan^{-1}(\gamma q^2)\right) \quad (3.15) \]

The aberration coefficient \( C_{CC} \) introduces a weak damping envelope and also an additional phase shift. The phase shift for small spatial frequencies \( q \) is proportional to \( q^2 \). For large \( q \)-values the phase shift goes asymptotically to \( \pi/4 \) and becomes independent of \( q \). This behavior is also observed for the rigorous treatment (see Eq. 3.11). To our knowledge, this is the first known case that chromatic aberration causes a phase shift. This phase shift has the same sign as the phase shift due to fifth order spherical aberrations since the chromatic aberration coefficient \( C_{CC} \) and the spherical aberration coefficient have the same sign (see Section 3.2.7 on aberration coefficients). However, in the case if the IBM LEEM [8] the phase shift due to chromatic aberration is small in the \( q \)-range passed by the aperture and is negligible compared to the phase shift due to wave aberrations in that \( q \)-range.

The three separate chromatic envelope functions \( E_C(q) \), \( E_{3C}(q) \), and \( E_{CC}(q) \) can also be obtained from Eq. 3.11 by setting each of the other two aberration coefficients to zero.

Similar to the case of the lowest order chromatic aberrations discussed above, the presence of current and voltage instabilities can be considered in the phase shift \( \xi_{\text{total}} \) by replacing the term

\[ \frac{\epsilon}{E} \quad \text{with} \quad \left(\frac{\epsilon}{E} + \frac{u}{U} + 2i I\right) \]

where \( u, i, U \) and \( I \) are defined as stated above. We already know that these effects can be neglected in the case of the lowest order chromatic aberrations. The same holds for the case of \( C_{3C} \), since the corresponding envelope function \( E_{3C}(q) \) (Eq. 3.13) has the same form as \( E_C(q) \). Solving the integral of Eq. 3.14 with current and voltage instabilities included and use of Eq. 3.15 yields

\[ \left(1 + (i\gamma q^2)^2\right)^{-1/4} \left(1 + (\gamma U q^2)^2\right)^{-1/4} \left(1 + (4\gamma I q^2)^2\right)^{-1/4} \]
where the superscripts $U$ and $I$ indicate that the ratio $\Delta E/E$ in Eq. 3.15 is replaced by $\Delta U/U$ and $2\Delta I/I$, respectively. The phase factors due to current and voltage instabilities have already been neglected. A series expansion gives $(1 + (\gamma q^2)^2)^{-1/4} \approx 1 - (\gamma^2 q^4/4)$. The second term is about a factor $10^4 - 10^6$ smaller in the case of current and voltage instabilities compared to the case of energy spread. Therefore, the instabilities in lens current and voltage are also negligible in the case of next higher order chromatic aberrations and are not considered further.

The effects of all chromatic aberrations discussed here can be considered in the CTF by replacing $E_C(q)$ in Eq. 3.1 with $E_{\text{total}}(q)$, Eq. 3.11.

### 3.2.4 Spatial coherence

Here we consider the effect of an extended source. In practice electrons are emitted from a finite area on the source cathode rather than a point source. Therefore, electrons that are emitted from a position some distance away from the point on the optical axis will be incident with a tilt angle, $\alpha_{\text{ill}}$, at the sample. This angle causes a shift in spatial frequency given by

$$q_{\text{ill}} = \frac{\alpha_{\text{ill}}}{\lambda}$$  \hspace{1cm} (3.16)

The extended source can be described by a source density with a Gaussian distribution with FWHM given by $q_{\text{ill}} = 2\sigma_{\text{ill}} \sqrt{2 \ln 2}$. This leads to an exponential envelope damping function given by [16]

$$E_S(q) = \exp \left( -2\pi^2 \sigma_{\text{ill}}^2 |\nabla \chi(q)|^2 \right)$$  \hspace{1cm} (3.17)

Using the expression for the phase shift due to wave aberrations given in Eq. 3.3, the envelope damping function due to an extended source reads

$$E_S(q) = \exp \left( -\pi^2 q_{\text{ill}}^2 \frac{4}{4\ln 2} \left( C_3 \lambda^3 q^3 - \Delta z \lambda q \right)^2 \right)$$  \hspace{1cm} (3.18)

Figure 3.1b shows a plot of the source envelope damping function with a typical angle spread of a cold field emitter, $\alpha_{\text{ill}} = 0.25$ mrad, for standard LEEM. It is clear that it has no impact on the CTF because of the overriding effect of the chromatic envelope. Therefore, we will neglect the effect of source extension in standard LEEM in the following.

When the contribution of higher order wave aberrations to the phase shift, Eq. 3.5, is considered, the exponential damping function due to an extended source becomes

$$E_S(q) = \exp \left( -\pi^2 q_{\text{ill}}^2 \frac{4}{4\ln 2} \left( C_3 \lambda^3 q^3 + C_5 \lambda^5 q^5 - \Delta z \lambda q \right)^2 \right)$$  \hspace{1cm} (3.19)
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Again, like in the case of standard LEEM, the effect of an extended source is negligible compared to the chromatic envelope and is therefore neglected in the following.

3.2.5 PROPERTIES OF THE CTF

The CTF describes the transmission of information from the object wave function to the image wave function as a function of the spatial frequency. The transmission depends on the amplitude and phase of the CTF which oscillate with increasing frequency for increasing \( q \)-values. The point where the real or imaginary part, depending on the nature of the object, of the CTF first crosses the abscissa defines the point resolution of the microscope. Up to the first zero crossing all the transmitted phase information has the same sign and contributes therefore in the same manner to the image.

In addition, the CTF amplitude is damped by the chromatic envelope function with increasing spatial frequency. Therefore, only information up to a certain spatial frequency is transmitted. The point where the amplitude is too small to contribute significantly to the image is called the information limit. In well-designed instruments this information limit is located beyond the point resolution. Figure 3.2a and b shows plots of the real part of CTFs for energy dependent aberration coefficients at 1 eV, 10 eV, and 30 eV for standard and aberration-corrected LEEM, respectively. The point of the first zero crossing shifts to higher \( q \)-values with increasing starting energy improving the point resolution. Aberration-correction improves the point resolution at a fixed starting electron energy significantly.

Figure 3.2: (a) Real parts of CTFs with \( E_0 = 1 \) eV (green dashed line), \( E_0 = 10 \) eV (red dotted line), and \( E_0 = 30 \) eV (blue solid line) for standard LEEM at in-focus condition and \( \Delta E = 0.25 \) eV. (b) Real parts of CTFs with \( E_0 = 1 \) eV (green dashed line), \( E_0 = 10 \) eV (red dashed line), and \( E_0 = 30 \) eV (blue solid line) for aberration-corrected LEEM at in-focus condition and \( \Delta E = 0.25 \) eV. (c) Real part (solid lines) and imaginary part (dashed lines) of the CTF at in-focus condition (red lines), at \( \Delta Z_\phi^2 \) defocus (blue lines), and at \( \Delta Z_\phi^3 \) defocus (green lines) for standard LEEM with \( \Delta E = 0.25 \) eV and \( E_0 = 10 \) eV.
Weak phase approximation

Usually in high resolution transmission electron microscopy thin objects are used for imaging. In that case, the CTF approach focuses on the special case of weak phase objects. Considering a pure phase object (i.e. an object that does not contain variations in scattering amplitude) the corresponding object wave function is given by $f(r) = \exp(i\phi(r))$, where $\phi$ is the phase. In the case of a weak phase object, i.e. $\phi << 1$, the object wave function can be written as an expansion of $\exp(i\phi(r))$

$$f(r) \approx 1 + i\phi(r)$$  \hspace{1cm} (3.20) 

where all terms containing $\phi^2$ and higher orders of $\phi$ are neglected [17]. The image wave function is given by the convolution of the object wave function with the PSF, $h(r)$, which is given by the inverse Fourier transform of the CTF. In the non-aberration-corrected case, the CTF consists of a product of real and even functions except for the wave aberration function which is complex and even. Therefore, the imaginary part of the PSF is only given by the inverse Fourier transform of the imaginary part of the CTF. Then, the image intensity of a weak phase object, to first order, is given by [4]

$$I = \Psi \Psi^* \approx 1 + 2\phi \otimes F^{-1}\left\{Im\left[W(q,\Delta z)\right]M(q)E_C(q,\Delta E)\right\}$$  \hspace{1cm} (3.21) 

where $\otimes$ is the symbol for the convolution and $F^{-1}$ denotes the inverse Fourier transform. This means that only the imaginary part of the wave aberration function, i.e. $Im[W(q,\Delta z)] = \sin(2\pi\xi(q,\Delta z))$, is of significance for weak phase objects. In LEEM and PEEM, phase shifts can be anywhere between 0 and $2\pi$, and structure factor differences across the object can give rise to strong amplitude contrast, as it is also the case for thicker specimens in transmission electron microscopy. In LEEM/PEEM therefore, the weak phase approximation does not apply in general, and we must utilize the full wave aberration function, i.e.

$$W(q,\Delta z) = \cos(2\pi\xi(q,\Delta z)) + i\sin(2\pi\xi(q,\Delta z))$$

3.2.6 Scherzer defocus

The amplitude of the CTF is seen to oscillate between positive and negative values with increasing $q$. The regions between each two zero crossings are called passbands. The first passband extends from $q = 0$ up to the first zero crossing, i.e. the point resolution. Ideally the first passband is as broad as possible to assure the best point resolution. The first zero crossing can be shifted to larger $q$ and the point resolution of an electron microscope can be optimized by defocussing slightly. A defocus is chosen that balances the effects of spherical aberrations and defocus term in Eq. 3.3. At the same time, the defocus should give rise to an amplitude
of the CTF that is ideally close to a transmission of 1. Such an optimized defocus value in the case of a weak phase object is called Scherzer defocus, first introduced by Scherzer [18].

In the case of a weak phase object, the effective part of the wave aberration function is given by \( \text{Im}[W(q, \Delta z)] = \sin(2\pi \xi(q, \Delta z)) \). Although the amplitude is ideally close to 1, a less strict requirement for the amplitude is chosen to allow for a broader passband. Considering an amplitude of \( 1/\sqrt{2} \approx 0.71 \) to be sufficient, the maximum argument of the sine to fulfill that requirement before the first zero crossing is given by \( \pm 3\pi/4 \). However, only the negative argument gives a real solution for the optimized defocus value. Therefore, this requirement is given by

\[
2\pi \xi = -\frac{3\pi}{4},
\]

By differentiating Eq. 3.3, the criterion for a nearly flat \( W(q, \Delta z) \) in the range of the first passband, i.e. \( d\xi/dq = 0 \), leads to

\[
q^2 = \frac{\Delta z}{C_3 \lambda^2}.
\]

By combining the latter two equations we obtain the value of the Scherzer defocus [5, 18] for weak phase (\( \phi \)) objects, with dominant third order spherical aberration:

\[
\Delta Z_3^3 = \sqrt{\frac{3}{2}} C_3 \lambda \approx 1.22(C_3 \lambda)^{1/2}.
\]

Below, we will equivalently define and derive Scherzer defocus values for weak phase objects with dominant fifth order spherical aberration (\( C_3 = 0 \)), as well as for pure amplitude (\( A \)) and strong phase (\( \Phi \)) objects, both with dominant third and fifth order spherical aberrations.

The CTF for standard LEEM at zero defocus and at \( \Delta Z_3^3 \) Scherzer defocus is plotted in Fig. 3.2c. It is clear that this defocus shifts the zero crossing of the imaginary part to higher \( q \)-value improving the point resolution for weak phase objects. The zero crossing of the real part is shifted to lower \( q \)-values at the same time. However, for most objects that are encountered in LEEM and PEEM, the real part of the CTF is dominant. Therefore, a different defocus value that improves point resolution for amplitude and strong phase objects has to be used.

**Amplitude and strong phase objects**

In LEEM a sample usually exhibits a multitude of phase and amplitude objects. Therefore, in general the approximations for weak phase objects cannot be applied in LEEM, and the CTF has to be used in its general form for image calculations. Then, the image formation of an object is determined by both the real and imaginary parts of the CTF. We consider here the case that image formation is dominated...
by the real part. In this case, the first zero crossing of the real part is used as the point resolution.

Now we derive a defocus value which optimizes the point resolution of an object in standard LEEM for which the image formation is dominated by the real part of the CTF, i.e., for $A$ and $\Phi$ objects. Following Scherzer’s example for weak phase objects, we consider a transfer amplitude of $1/\sqrt{2}$ to be sufficient. Hence, the maximum argument of the cosine, i.e., the real part of the wave aberration function, to fulfill that requirement before the first zero crossing is given by $\pm \pi/4$. Again, only the negative argument leads to one real solution. Therefore, we obtain $C_3 \lambda^3 q^4 – 2\Delta z \lambda q^2 = -1/2$. Substitution of $q$ using Eq. 3.22 gives a defocus of

$$\Delta Z_{A\Phi}^3 = \sqrt{\frac{1}{2} C_3 \lambda \approx 0.71 (C_3 \lambda)^{1/2}}$$  (3.24)

The Scherzer defocus value is therefore about 40% smaller for this case than for weak phase Scherzer defocus $\Delta Z_{\Phi}^3$. The real situation for LEEM may be somewhere in between, depending upon the object. Figure 3.2c shows the real and imaginary part of the CTF at $\Delta Z_{3\Phi}^3$ Scherzer defocus (green solid and dashed lines). The position of the first zero crossing of the real part is at significantly larger $q$ compared to zero defocus. However, the first zero crossing of the imaginary part is the same as for in-focus although the amplitude has opposite sign.

So far we have discussed Scherzer defocus values for standard LEEM and TEM. In the case of aberration-corrected instruments, these defocus values have to be adapted. In the following we derive a defocus value which optimizes the point resolution of a weak phase object in aberration-corrected LEEM and TEM, i.e., a weak phase fifth order Scherzer defocus for aberration-corrected instruments, and a defocus value which optimizes the point resolution of strong phase/amplitude objects in aberration-corrected LEEM, i.e., a fifth order amplitude/strong phase Scherzer defocus. The derivation is equivalent to the derivation of $\Delta Z_{\Phi}^3$ in Eq. 3.23 and $\Delta Z_{A\Phi}^3$ in Eq. 3.24 for the case of non-aberration-corrected instruments, except that the third order spherical aberration term is zero and only fifth order spherical aberration is considered. The requirements $2\pi \xi = -(3\pi/4)$ and $2\pi \xi = -(\pi/4)$ for the $\phi$ and $A\Phi$ Scherzer defocus, respectively, give

$$C_5 \lambda^5 q^6 – 3\Delta z \lambda q^2 = -\frac{9}{4}$$  (3.25)

and

$$C_5 \lambda^5 q^6 – 3\Delta z \lambda q^2 = -\frac{3}{4}$$  (3.26)
The requirement of a nearly flat function, \( d\xi/dq = 0 \), gives \( q^4 = \Delta z/C_5 \lambda^4 \). Using this equation to replace \( q \) in Eq. 3.25 and Eq. 3.26 gives one real solution each. The fifth order weak phase Scherzer defocus is given by

\[
\Delta Z_\phi^5 = \left( \frac{81}{64} C_5 \lambda^2 \right)^{1/3} \approx 1.08(C_5 \lambda^2)^{1/3}
\]

and the fifth order \( A\Phi \) Scherzer defocus is given by

\[
\Delta Z_{A\Phi}^5 = \left( \frac{9}{64} C_5 \lambda^2 \right)^{1/3} \approx 0.52(C_5 \lambda^2)^{1/3}
\]

The point resolution in aberration-corrected LEEM can possibly be further improved by using an approach that involves the use of an optimized defocus value and a negative value for the third order spherical aberration term to balance at least partly the fifth order spherical aberrations [19]. We will discuss such an optimization in chapter 4 and 5.

### 3.2.7 Aberration Coefficients

We used aberration coefficients calculated with two different raytracing programs, namely the MIRDA program from Munro’s Electron Beam Software Ltd. (London, UK) and the COSY INFINITY code (M. Berz, K. Makino, COSY INFINITY 9.0 Beam Physics Manual, MSU Report MSUHEP-060804, Department of Physics and Astronomy, Michigan State University, 2006). A detailed description of the computation and comparison with an analytical theory can be found in Ref. [14]. Aberration coefficients obtained for both the uncorrected and the corrected microscopes are given in Table 3.1, for starting electron energies of 1 eV, 10 eV, and 30 eV. The coefficients are the sum of objective lens aberrations and electron mirror aberrations and are given for an image plane with magnification \( M = 1 \) and nominal energy \( \bar{E} = 150\,\text{eV} \). The aberration coefficients depend on the starting energy of the electrons, decreasing with increasing starting energy. A method to experimentally determine the values of the aberration coefficients in LEEM is discussed in Ref. [20].

### 3.3 Image Contrast Calculations in LEEM

We calculate the image contrast created by pure phase and pure amplitude objects as well as objects consisting of a superposition of phase and amplitude contributions. First we consider spatial variations, i.e. objects, only in one dimension. Then, we discuss two-dimensional objects and the respective image features. The phase and amplitude objects are represented by step functions as shown in Fig. 3.3. A pure amplitude object has a constant phase and produces only variations in the
3.3. IMAGE CONTRAST CALCULATIONS IN LEEM

Table 3.1: Calculated aberration coefficients (in meters) for the IBM LEEM system for different starting electron energies. The coefficients are the sum of objective lens aberrations and electron mirror aberrations and are referenced to an image plane with $M = 1$ and $E = 15010$ eV.

<table>
<thead>
<tr>
<th>Energy (eV)</th>
<th>$C_3$</th>
<th>$C_5$</th>
<th>$C_C$</th>
<th>$C_{3C}$</th>
<th>$C_{CC}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Non-aberration-corrected</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>0.492</td>
<td>768</td>
<td>-0.13</td>
<td>-1484</td>
<td>719</td>
</tr>
<tr>
<td>10</td>
<td>0.345</td>
<td>39.4</td>
<td>-0.075</td>
<td>-59.37</td>
<td>23.09</td>
</tr>
<tr>
<td>30</td>
<td>0.305</td>
<td>14.5</td>
<td>-0.052</td>
<td>-16.12</td>
<td>4.58</td>
</tr>
<tr>
<td>Aberration-corrected</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>0</td>
<td>749</td>
<td>0</td>
<td>-1433</td>
<td>731</td>
</tr>
<tr>
<td>10</td>
<td>0</td>
<td>92.8</td>
<td>0</td>
<td>-67.4</td>
<td>27.9</td>
</tr>
<tr>
<td>30</td>
<td>0</td>
<td>66.4</td>
<td>0</td>
<td>-23.2</td>
<td>8.2</td>
</tr>
</tbody>
</table>

amplitude, $\sigma(r)$. A pure phase object, on the other hand, creates only a phase shift and has no effect on the amplitude. A surface step connecting two atomic terraces with the same reflection coefficients is an example of a phase object [21]. This phase shift is given by

$$\Delta \psi = kd = \left( \frac{2\pi}{\lambda_0} \right) 2a_0$$

where $d = 2a_0$ is the path length difference between waves that are reflected from opposite sides of a step with step height $a_0$, and $\lambda_0$ is the wavelength of the low energy electrons that are elastically backscattered from the object. The border between regions that have different reflection coefficients without an intervening step is an example of a pure amplitude object.

All image intensity calculations, except where noted otherwise, have been performed with the aperture size for which the best lateral resolution is obtained at zero defocus. How we obtain these values is discussed in Section 3.3.3 on resolution calculations. For all calculations presented here, the starting electron energy is $E_0 = 10$ eV and the energy spread is $\Delta E = 0.25$ eV. We obtain the optimum aperture angle at in-focus condition of $\alpha_{\text{nac}} = 2.34$ mrad and $\alpha_{\text{ac}} = 7.37$ mrad for the standard and the aberration-corrected microscope, respectively, for amplitude and strong phase objects. In the case of weak phase objects the optimum aperture angle at in-focus condition is $\alpha_{\text{nac}} = 2.78$ mrad and $\alpha_{\text{ac}} = 8.32$ mrad for the standard and the aberration-corrected LEEM, respectively. The corresponding cut-off frequencies are given by $q_{\text{max}}^{\text{nac}} \approx 0.234$ nm$^{-1}$ and $q_{\text{max}}^{\text{ac}} \approx 0.737$ nm$^{-1}$ for ampli-
Figure 3.3: Amplitude, $\sigma$, (red dashed lines) and phase, $\phi$, (black solid lines) components of a one-dimensional pure $1 : 1/\sqrt{2}$ amplitude object (a) and a pure $\pi$ phase object (b).

tude/strong phase objects and by $q_{\text{max}}^{\text{nac}} \approx 0.278 \text{ nm}^{-1}$ and $q_{\text{max}}^{\text{ac}} \approx 0.832 \text{ nm}^{-1}$ for weak phase objects for the standard and the aberration-corrected LEEM, respectively.

3.3.1 ONE-DIMENSIONAL OBJECTS

Figure 3.4 shows the one-dimensional image intensity profiles of a pure $1 : 1/\sqrt{2}$ amplitude object (a-c), a pure $\pi$ phase object (d-f), and a pure $\pi/2$ phase object (g-i) considering only the effects of the contrast aperture (a, d, and g), the contrast aperture and the wave aberration function (b, e, and h), and the entire CTF (c, f, and i). The image contrast is shown for in-focus for standard (thick black lines) and aberration-corrected LEEM (thin red lines). The aperture function eliminates the contributions of spatial frequencies above a sharp cut-off. Sharp features like the amplitude object step function can only be reproduced by an infinite Fourier series. The use of the contrast aperture leads to the loss of higher $q$-values and therefore introduces broadening and intensity fringes (see Fig. 3.4, left panels). The width and spacing of these features scales with the inverse of the cut-off frequency $q_{\text{max}}$. The image modifications introduced by the aperture degrade the resolution and are responsible for the well-known diffraction limit in resolution [4].

In the case of a phase object, contrast can only be observed due to the cut-off of higher spatial frequencies. When the calculation considers the cut-off of the aperture only (Fig. 3.4d, and g), a core destructive fringe is located near the position of the phase jump in the corresponding object (here located at $x = 0$) and is surrounded by diffraction fringes. In the limit of $\alpha_{\text{ap}} \to \infty$, the lateral dimensions of the phase object approach zero and phase contrast is lost. The same fundamental effect is created by all of the other factors that act as low pass filter like the chromatic envelope function and the envelope function due to an extended source.
3.3. IMAGE CONTRAST CALCULATIONS IN LEEM

Figure 3.4: Image contrast of a $1: 1/\sqrt{2}$ amplitude object (a)-(c), a $\pi$ phase object (d)-(f), and a $\pi/2$ phase object (g)-(i) obtained by using only the aperture function, $M(q)$, ((a), (d), and (g)), using the aperture function and the wave aberration function, $W(q)$, ((b), (e), and (h)), and using the aperture function and the wave aberration function plus the total chromatic envelope function, $E(q)$, with $\Delta E = 0.25$ eV ((c), (f), and (i)). The thick black lines correspond to standard LEEM and the thin red lines to aberration-corrected LEEM. All calculations were performed at in-focus condition with optimum aperture angle and $E_0 = 10$ eV.
We have already discussed that the wave aberration function causes an increasing phase shift with increasing spatial frequency. This behavior also leads to intensity fringes and broadening. The cut-off of an aperture of optimum size is located at the first zero crossing of the relevant part of the CTF, i.e. the point resolution. Consequently, the modifications caused by the oscillations of the aberration function at larger $q$-values are suppressed and any remaining effects are minor compared to the effect of the contrast aperture. This is true for amplitude objects and $\pi$-phase objects. However, for phase objects like $\pi/2$ or $3\pi/2$ the wave aberration function introduces asymmetric (with respect to the phase step position) modifications in the image contrast (see Fig. 3.4g, and h) as already observed by Chang et al. [21] and by Pang et al. [3] for non-aberration-corrected LEEM.

The chromatic aberration envelope function (see Eq. 3.11) dampens higher $q$-values and reduces their transmittance. For a non-aberration-corrected LEEM the dampening is determined by the lowest order chromatic aberration term. In the case of the aberration-corrected LEEM the dampening behavior is determined by the chromatic aberration term $C_{3C}$ and to a lesser extend by the $C_{CC}$ aberrations for an energy spread of $\Delta E = 0.25$ eV. A larger energy spread can cause a change in the relative importance of the two higher order chromatic aberrations. The dampening leads to a reduction of the amplitude of the diffraction fringes and broadens them further. The effect of the chromatic damping envelope is independent of the focus condition. For in-focus condition with an optimum aperture the chromatic envelope effect introduces significant changes in standard LEEM but has only a minor impact in the case of the aberration-corrected microscope for a reasonably small energy spread ($\Delta E < 1$ eV). The reason for this is that the chromatic envelope reduces the transmittance at the cut-off $q_{\text{max}}$ by 33 % in the case of the standard LEEM and only 2 % in the case of the aberration-corrected LEEM. A similar scenario is observed when we apply the Scherzer defocus together with the correspondingly larger optimum aperture sizes. Since the point resolution and therefore the cut-off frequency are now shifted to higher $q$-values but the chromatic envelope stays unaffected by the defocus, the maximum amount of chromatic dampening at the cut-off is now increased to 78 % and 9 % for the standard and the aberration-corrected LEEM, respectively.

Figure 3.4a-c shows the contrast profile of a pure $1 : \frac{1}{\sqrt{2}}$ amplitude object at in-focus condition for standard and aberration-corrected LEEM. The object produces an intensity change of one half because it is calculated as the square of the wave function. The high intensity side as well as the low intensity side exhibit intensity fringes which are most pronounced around the amplitude step and decay with distance from the step. This behavior is observed for all pure amplitude objects with jumps from a value greater than zero to another value greater than zero.
given that the low amplitude side is not too close to zero, compared to the fringe amplitude. In the case of an object with an amplitude step from a value greater than zero to zero the intensity fringes are almost completely suppressed on the low intensity side [3]. In any case, aberration correction produces intensity profiles of amplitude objects that exhibit a steeper slope between the high and the low intensity sides and fringes that are narrower and oscillate with higher frequencies compared to the standard uncorrected case.

In the case of pure phase objects the CTF reproduces several behaviors that were observed previously with Fourier optics [3] and also with an analytical wave-optical model [21]. In agreement with the previously published work we observe that the corresponding image contrast highly depends on the magnitude of the phase jump. For the so-called in-phase condition, $\Delta \psi = 2n\pi$ with $n =$integer, the phase shift amounts to zero since the phase factor has a periodicity of $2\pi$. Therefore, contrast is absent for the in-phase condition. In the case of the out-of-phase condition, $\Delta \psi = (2n + 1)\pi$, the intensity fringes are located symmetrically around a completely destructive interference fringe at the phase step position [3]. Figure 3.4d-f shows the image contrast for an out-of-phase object with $n = 0$, namely $\Delta \psi = \pi$, which clearly shows a mirror symmetry around $x = 0$.

Objects with intermediate phase condition produce image contrast with clear asymmetric features. This asymmetry is most pronounced at $\Delta \psi = ((2n + 1)\pi)/2$ [3, 21]. Our calculations show for $\Delta \psi = \pi/2$ and equivalent conditions ($n =$even), that the intensity fringes including the first intensity maximum after the global minimum are more pronounced on the down side than on the upper side of the step at in-focus condition. This asymmetric behavior has been reported for imaging in an uncorrected instrument at defocus [3, 21] but not for in-focus imaging as observed here. The wave aberration function gives rise to asymmetric fringes. We observe asymmetry at in-focus for optimum aperture size. In case of a larger energy spread (e.g. $\Delta E = 1.2$ eV as used in Ref. [3]), the chromatic envelope is narrower, and that suppresses the effect of the wave aberration function and the contrast becomes almost completely symmetric. We also observe asymmetric fringes for a $\Delta \psi = \pi/2$ phase object in the aberration-corrected case at in-focus for the first time. These fringes exhibit the same asymmetry as the uncorrected case because the spherical aberration coefficient $C_3$ and $C_5$ have the same sign, therefore introduce phase shifts in the wave aberration function with the same sign. For $\Delta \psi = 3\pi/2$ and equivalent conditions ($n =$odd), the asymmetry is reversed with more intense fringes on the upper side of the step for standard LEEM [3, 21] and for aberration-corrected LEEM.

As a generic example of a mixed amplitude/phase object, Fig. 3.5a shows the image contrast of a mixed $1: \frac{1}{\sqrt{2}}$ amplitude and $\pi$ phase object with the amplitude
and phase step coinciding at $x = 0$ for standard and aberration-corrected LEEM. Comparison with the pure $1 : \frac{1}{\sqrt{2}}$ amplitude object (Fig. 3.4c) shows that the superposition with a $\pi$ phase object causes larger amplitudes of the intensity fringes next to the step.

In Fig. 3.5b-d the image contrast is shown for a pure $1 : \frac{1}{\sqrt{2}}$ amplitude, a $\pi$ phase, and a $\pi/20$ phase object, respectively, for aberration-corrected LEEM at in-focus (red solid lines), at $\Delta Z_{\phi}^{S}$ Scherzer defocus, Eq. 3.27, (blue dashed lines) and at $\Delta Z_{A\phi}^{S}$ Scherzer defocus for amplitude/strong phase objects, Eq. 3.28, (green dotted lines). For the amplitude and $\pi$ phase object the first zero crossing of the real part of the CTF is taken as the aperture size for each focus condition. In the case of the $\pi/20$ phase object the first zero crossing of the imaginary part of the CTF is taken as the aperture size for each focus condition.

If we apply $\Delta Z_{A\phi}^{S}$ defocus the slope of the step in the image contrast of the $1 : \frac{1}{\sqrt{2}}$ amplitude object (Fig. 3.5b, green dotted line) becomes steeper and therefore improves the resolution (see Section 3.3.3 on resolution) to $R = 0.5$ nm compared to $R = 0.6$ nm at in-focus. The $\Delta Z_{\phi}^{S}$ defocus causes a less steep slope in the image that results in a resolution of $R = 1.2$ nm. Therefore, the resolution is about a factor 2.4 worse for the latter defocus condition. This result clearly demonstrates the difference between weak phase and non-weak phase imaging and that different Scherzer defocus values are necessary.

In the case of the $\pi$ phase object (Fig. 3.5c) the observed central dip becomes narrower for $\Delta Z_{A\phi}^{S}$ defocus and gets wider for $\Delta Z_{\phi}^{S}$ defocus. The corresponding resolution is $R = 0.6$ nm, $R = 0.5$ nm, and $R = 0.7$ nm for imaging at in-focus. Therefore, we conclude that also in the case of strong phase objects the resolution in aberration-corrected instruments is improved by $\Delta Z_{A\phi}^{S}$ defocus.

The image contrast of a weak phase object is shown in Fig. 3.5d for different defocus conditions. The depth and height of the central intensity fringes are increased and asymmetry of the fringes is reversed for $\Delta Z_{\phi}^{S}$ defocus and, to a lesser extent, also for $\Delta Z_{A\phi}^{S}$ defocus compared to the in-focus case. The inversion of the asymmetry of the fringes around the position of the phase step is, although to a lesser extent, also observed for $\pi/2$ and $3\pi/2$ phase objects. From Fig. 3.2c we know that the imaginary part of the CTF has up to the first zero crossing opposite sign for $\Delta Z_{\phi}$ and $\Delta Z_{A\phi}$ compared to at in-focus condition. The real part of the CTF, however, has up to the first zero crossing the same sign for all three focus conditions. This explains why the asymmetry of the fringes in the contrast of weak phase objects, where the imaginary part of the CTF is relevant, is reversed for $\Delta Z_{\phi}$ and $\Delta Z_{A\phi}$. It also shows that for $\pi/2$ a $3\pi/2$ phase objects the imaginary part of the CTF cannot be completely neglected and both parts of the CTF have an impact.
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**Figure 3.5:** (a) Contrast of a mixed $1:1/\sqrt{2}$ amplitude and $\pi$ phase object for standard LEEM (thick black line) and aberration-corrected LEEM (thin red line) at in-focus with optimum aperture angle. Image contrast of a $1:1/\sqrt{2}$ amplitude object (b), a $\pi$ phase object (c), and a $\pi/20$ phase, i.e. weakphase, object (d) for aberration-corrected LEEM at in-focus (red solid lines), at $\Delta Z_{\text{inf}}$ defocus (blue dashed lines), and at $\Delta Z_{\text{def}}^5$ defocus (green dotted lines). The aperture angle is $\alpha = 15$ mrad. All calculations were performed with $E_0 = 10$ eV and $\Delta E = 0.25$ eV.
the image formation. The resolution of the $\pi/20$ phase object is $R = 0.6 \text{ nm}$, $R = 0.8 \text{ nm}$, and $R = 0.7 \text{ nm}$ for imaging at in-focus, $\Delta Z^{5}_{A\Phi}$ defocus, and at $\Delta Z_{\phi}$ defocus, respectively. Therefore, both defocus values degrade the resolution compared to the in-focus condition. However, at the same time they improve the image contrast. Although the in-focus condition yields the best resolution it might be still advantageous to use $\Delta Z^{5}_{\phi}$ defocus to improve image contrast of weak phase objects (see Section 3.3.3 on resolution for a more detailed discussion).

The CTF formalism shows that all phase objects (excluding the in-phase case which is equivalent to a constant phase) produce significant image contrast for in-focus condition. This is in contradiction to the reported experimental observation that phase contrast disappears for in-focus condition. Like with Fourier optics, contrast is produced with the CTF formalism for in-focus condition because alterations like phase shifts and suppression of high $q$-values introduced by the chromatic envelope and the contrast aperture are always present and independent of defocus. As pointed out before [3], the loss of phase contrast in experiment at in-focus condition has to stem from a different source. We suggest that one possible reason is the modification introduced by the detection system which is characterized by its modulation transfer function (MTF). The MTF of the detector can lead to further broadening and loss of image details. Defocus can increase the width and can change the depth of the main destructive fringe of phase objects (see Fig. 3.5 and [3, 21]) in order that it can be recorded with a lower detector resolution. We will discuss the qualitative effect of a detector in Section 3.3.3 using a simple model.

### 3.3.2 2D IMAGE CALCULATIONS

The CTF formalism can readily be extended to image calculations of two-dimensional objects, i.e. real objects in LEEM. The 2D CTF is generated by rotation of the 1D CFTs around the optical axis at $q = 0$. Again, like in the one-dimensional case it is possible to calculate image contrast of pure amplitude, pure phase, and mixed arbitrary phase and amplitude objects.

We calculate here the image contrast of square-shaped amplitude and phase objects surrounded by a uniform background. We have chosen an amplitude step of $1: \frac{1}{\sqrt{2}}$ and the special case of an $1: 0$ amplitude step. The side length of the square, is varied form 10 nm, well above the resolution limit, down to 0.1 nm, below the resolution limit, with a step size of 0.1 nm. Figure 3.6a, d, g, j shows a $1: 0: 1$ amplitude object, the inverse $0: 1: 0$ amplitude object, $\pi$ phase and arbitrary amplitude objects, respectively, that are used to perform 2D image contrast calculations. The calculated images are shown, respectively, in Fig. 3.6b, e, h, k for standard LEEM and Fig. 3.6c, f, i, and l for aberration-corrected LEEM.

Intensity line profiles through the center of the object functions and the cal-
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(a) object

(b) nac

(c) ac

(d) object

(e) nac

(f) ac

(g) object

(h) nac

(i) ac

(j) object

(k) nac

(l) ac
calculated 2D images in horizontal direction (black dashed lines in Fig. 3.6) are also shown in Fig. 3.6. In addition, Fig. 3.6 shows line profiles taken along the diagonal of the object (green dashed lines in Fig. 3.6). These profiles are projected on to the x position axis so that they can be easily compared with the horizontal line profiles. It is clear that the line profiles along these two directions yield different results. Therefore, additional information is obtained from the 2D images compared to the one-dimensional images. While the expect the horizontal line profile to resemble the one-dimensional calculation result when the object dimension is large compared to fringe spacing, the new information in the diagonal line profile of the 2D object arises because of corners that are not present in the one-dimensional calculation. With decreasing object size, deviation from the one-dimensional result will be exacerbated.

From the horizontal profile plots we extracted the contrast. The contrast of a pure $\alpha : \beta$ amplitude object is defined as

$$\text{Intensity of the global maximum (minimum) - Intensity far away} \over \alpha^2 - \beta^2$$

In the case of a pure phase object the contrast is defined as

$$\text{Intensity of the global maximum (minimum) - Intensity far away} \over \text{Intensity far away}$$

for Intensity far away $\neq 0$. The contrast of different amplitude and phase objects is plotted as a function of their size in Fig. 3.7 for standard (a, c, and e) and aberration-corrected (b, d and f) LEEM.

The image contrast of a square two-dimensional $0 : 1 : 0$ amplitude object shown in Fig. 3.6d is significantly higher than the inverse case of a $1 : 0 : 1$ object shown in
3.3. IMAGE CONTRAST CALCULATIONS IN LEEM

Figure 3.7: Contrast as a function of object size (see text for details). (a) Square object with $\sigma = 1$ surrounded by $\sigma = 0$ (red dashed line) and with $\sigma = 0$ surrounded by $\sigma = 1$ (black solid line) for standard LEEM. Also shown are the contrast of a square object with $\sigma = 1$ with a background of $\sigma = 1/\sqrt{2}$ (green dotted-dashed line) and contrast of an object with $\sigma = 1/\sqrt{2}$ surrounded by $\sigma = 1$ (blue dotted line). (b) Results for the same objects for aberration-corrected LEEM. The contrast of a $\pi : -\pi$ phase object is shown in (c) for standard LEEM and in (d) for aberration-corrected LEEM. The contrast of a $\pi/2 : -\pi/2$ phase (black solid lines) and a $3\pi/2 : -3\pi/2$ phase object (red dotted lines) is shown in (e) for standard LEEM and in (f) for aberration-corrected LEEM. The starting electron energy is $E_0 = 10$ eV, $\Delta E = 0.25$ eV, at in-focus condition. The dashed vertical lines indicate the corresponding calculated resolution limits (see Section 3.3 on resolution).
Fig. 3.6a for an object size in the range of about 3 – 8 nm for standard LEEM and in the range of about 1 – 2.5 nm for aberration-corrected LEEM. A similar behavior is observed for the cases of a $1: \frac{1}{\sqrt{2}} : 1$ and the inverse $1: \frac{1}{\sqrt{2}} : 1$ amplitude objects, although the difference is significantly less compared to the case of the $0 : 1 : 0$ and $1 : 0 : 1$ amplitude objects. This observation can be of value in practice to obtain better image contrast. For example, $1 : 0$ objects are frequently observed in dark field imaging mode. Dark-field contrast can be reversed choosing different diffraction spots for imaging. Therefore, dark field contrast of small objects can be optimized by choosing the appropriate diffraction spot for imaging.

The observed image contrast of a $\pi$ phase object (Fig. 3.7c and d) is symmetric across the phase step (see discussion in Section 3.3.1 on one-dimensional image contrast). Therefore a phase step of $\Delta \psi = \pi$ causes the same image contrast as a phase step of $\Delta \psi = -\pi$. That means that the contrast of a $\pi : -\pi$ phase object is the same as for a $-\pi : \pi$ phase object. The contrast of the $\pi : -\pi$ phase object is significantly enhanced in the range of about 4 – 7 nm with the maximum enhancement of about 60% located at about 5.3 nm for standard LEEM. In the aberration-corrected case, contrast of a $\pi : -\pi$ phase object is enhanced in the range of 1 – 2 nm, with the maximum enhancement of about 90% at 1.6 nm. The enhancement in contrast is due to the overlap of constructive interference fringes from both sides of the object that occurs at these dimensions. This agrees with the experimental observation by Wu et al. [22].

As already discussed for the one-dimensional case above the image contrast for a $\pi/2$ phase and a $3\pi/2$ phase object are related, i.e. they are mirror symmetric around a vertical plane at the step position. That means that a $\pi/2 : -\pi/2$ phase step gives rise to the same image contrast as a $-3\pi/2 : 3\pi/2$ step, and a $3\pi/2 : -3\pi/2$ phase step yields the same image as a $-\pi/2 : \pi/2$ phase step. Therefore, it is sufficient to look at phase steps of $\pi/2 : -\pi/2$ and $3\pi/2 : -3\pi/2$ for standard and aberration-corrected LEEM. The contrast of both phase objects oscillates as a function of object size. Their image contrast (Fig. 3.7e and f) goes asymptotically to about 50% for large objects. For phase objects with a size below about 15 nm and below about 5 nm the contrast varies vastly with the object size for standard and aberration-corrected LEEM, respectively.

### 3.3.3 Resolution of LEEM

#### Geometric optics model

The resolution is determined by diffraction, chromatic aberrations, and spherical aberrations. In the simplest approach, the resolution of the microscope, $R$, is often estimated using a simple geometric optics model approach. Including higher
order aberrations, the expression for the resolution takes the following form \[8, 13\]

\[
R = \left[ \left( \frac{0.61\lambda}{\alpha} \right)^2 + \left( C_3 \alpha^3 \right)^2 + \left( C_5 \alpha^5 \right)^2 + \left( C_C \left( \frac{\Delta E}{E} \right) \alpha \right)^2 \right]^{1/2} \tag{3.29}
\]

All five aberration terms listed here, plus the Rayleigh resolution term \(0.61\lambda\) were taken into account in our calculations of the resolution using the geometric optics method. Figure 3.8 shows the resolution as a function of the acceptance angle plus the single contributions of the five aberration terms and the diffraction term for an energy spread of \(\Delta E = 0.25\) eV and a nominal electron energy of \(E = 15010\) eV for standard LEEM (a) and for aberration-corrected LEEM (b). The starting electron energy is \(E_0 = 10\) eV.

In standard LEEM, the resolution is limited by diffraction and third order spherical aberrations. The chromatic aberrations are less important because of the small energy spread appropriate for a field emission source that is used in the calculation. All higher order aberration terms contribute only little to the resolution limit.
In an aberration-corrected microscope the resolution is limited by diffraction and fifth order spherical aberrations. The higher order chromatic aberrations do not contribute significantly to the resolution limit for the energy spread chosen here.

**Resolution using CTF**

We will use now the image profiles of different objects calculated with the CTF method to determine lateral resolution. A uniformly-illuminated circular aperture gives rise to an Airy diffraction pattern, i.e. a central bright Airy disk surrounded by a series of concentric bright rings [23]. The Airy disk extends to the first minimum located at a radius \( R = 0.61 \lambda / \alpha \). If we only consider diffraction or if diffraction is dominant, it is common practice to estimate the resolution from the electron wavelength and the size of the aperture by the Rayleigh criterion. It states that two point objects are just resolved if the global diffraction maximum produced in an image by one of the objects coincides with the first diffraction minimum produced by the other object. Then, the resolution is given by \( R = 0.61 \lambda / \alpha \).

The CTF considers diffraction by the aperture and also chromatic and spherical aberrations of the objective lens. This gives rise to a slightly modified Airy pattern and we will refer to this as a point spread function (PSF). The Airy disk can be approximated with a Gaussian if we ignore the smaller surrounding bright rings. If we optimally fit a Gaussian to the central Airy disk with the constraint that the peak amplitude of the Airy pattern is equal to the maximum of the Gaussian, the width of this Gaussian becomes \( 2 \sigma = 0.42 \lambda / \alpha \).

The integral of a Gaussian is the error-function which has the property that the lateral separation between 84 % and 16 % of the error-function is equal to the 2\( \sigma \)-width of the Gaussian. Now, if we extract the lateral distance between the 84 % and 16 % intensity values of an image intensity profile of an amplitude object we find a resolution \( R = 0.37 \lambda / \alpha \) in the diffraction limited regime. This value is close to the width of the Gaussian fitted to the central peak in the PSF. We conclude that the width of the central peak in the PSF determines diffraction limit and resolution. Therefore, the resolution of amplitude objects is here defined as the spatial separation between 84 % and 16 % of the contrast between the intensity far away on the left side and the intensity far away on the right side of the object, i.e. for \( \alpha : \beta \) amplitude objects this means the resolution is given by the spatial separation between 84 % and 16 % of \(|\alpha^2 - \beta^2|\).

We are interested in finding the aperture angle that optimizes resolution. On that account we repeat the resolution calculation outlined above for a series of different aperture angles. This gives resolution plots analogous to those obtained by the geometric optics method (see Fig. 3.8). Figure 3.9a shows resolution plots of a 1 : \( \frac{1}{\sqrt{2}} \) amplitude object, for an electron starting energy of 10 eV and an energy spread of 0.25 eV for standard (thin black solid lines) and aberration-corrected
LEEM (thick red solid lines) at in-focus condition, and at $\Delta Z_{A\Phi}$ defocus for standard (green dotted-dashed lines) and aberration-corrected (blue dotted lines) LEEM. Aberration-correction improves optimum resolution and increases optimum aperture angle by about a factor 3. The optimum aperture angle at $\Delta Z_{A\Phi}$ defocus is about 30 % and 15 % larger for standard and aberration-corrected LEEM, respectively, compared to the in-focus condition. At the same time resolution is improved by $\Delta Z_{A\Phi}$ defocus by about a 10 % for the non-corrected and the corrected case. Weak phase $\Delta Z_\phi$ Scherzer defocus, on the other hand, degrades resolution of a $1 : \frac{1}{\sqrt{2}}$ amplitude object by about 35 % and 45 % compared to resolution at in-focus for standard and aberration-corrected LEEM, respectively.

The resolution of a phase object is defined here as the FWHM of the central dip in the corresponding image contrast, where the maximum is defined as the difference of the intensity far away and the intensity at the central dip. Again, by calculating the image contrast of a phase object for a series of acceptance angles and extracting the resolution from each of these images one obtains the resolution as a function of the acceptance angle. Such resolution plots are shown in Fig. 3.9b-d for a $\pi$ phase, a $\pi/2$ phase, and a $\pi/20$ phase object, i.e. a weak phase object, respectively, for an electron starting energy of 10 eV and an energy spread of 0.25 eV for standard (thin lack solid lines) and aberration-corrected LEEM (thick red solid lines) at zero defocus. In the case of the $\pi$ phase object (see Fig. 3.9b) the resolution for $\Delta Z_{A\Phi}$ defocus is also shown for standard (green dotted-dashed line) and aberration-corrected (blue dotted line) LEEM. The increase in the optimum aperture angle and the improvement in the resolution at $\Delta Z_{A\Phi}$ defocus compared to the in-focus case is the same as for the $1 : \frac{1}{\sqrt{2}}$ amplitude object. Both, $\Delta Z_{A\Phi}$ and $\Delta Z_\phi$ Scherzer defocus, degrade the resolution in the case of the $\pi/2$ phase object and the $\pi/20$ phase object compared to the in-focus condition (see discussion below). The resolution of a $3\pi/2$ phase object is equal to the resolution of a $\pi/2$ phase object since they are connected by a symmetry operation.

The main reason for choosing $\Delta Z_\phi$ Scherzer defocus in the case of a weak phase object is to maximize phase contrast in the image [24]. The resolution, which is determined by the width of the PSF, is not necessarily optimized at the same time as discussed, e.g. by Lichte [25]. Figure 3.10 shows the resolution and contrast of a $1 : \frac{1}{\sqrt{3}}$ amplitude object (a), a $\pi$ phase object (b), and a $\pi/20$ phase object (c) as a function of defocus for standard and aberration-corrected LEEM with a starting electron energy of $E_0 = 10$ eV and $\Delta E = 0.25$ eV. The aperture angle is kept at a constant value at every defocus. For the amplitude and $\pi$ phase objects the aperture angle is given by the value of the first zero crossing of the real part of the CTF at $\Delta Z_{A\Phi}$ defocus values for standard and aberration-corrected LEEM. In the case of the weak phase object, $\pi/20$, the aperture angle is given by the first zero cross-
Figure 3.9: Resolution calculated with the CTF formalism for a $1:1/\sqrt{2}$ amplitude object (a), a $\pi$ phase object (b), a $\pi/2$ phase object (c), and a $\pi/20$ phase object (d) for standard LEEM (thin black solid lines) and aberration-corrected LEEM (thick red solid lines). All calculations were performed with $E_0 = 10$ eV, $\Delta E = 0.25$ eV and at in-focus condition. The thin green dotted-dashed and thin blue dotted lines in (a) and (b) correspond to the resolution at $\Delta Z_{\Delta\Phi}^3$ defocus and $\Delta Z_{\Delta\Phi}^5$ defocus, respectively. Additionally, the corresponding resolution obtained with the geometric optics model for non-corrected and aberration-corrected instruments is plotted in each figure (dashed lines).
3.3. IMAGE CONTRAST CALCULATIONS IN LEEM

Figure 3.10: Resolution (solid lines) and contrast (dotted lines) as a function of defocus for a $1 : 1 / \sqrt{\pi}$ amplitude object (a), a $\pi$ phase object (b), and a $\pi / 20$ phase object (c) for standard LEEM (thin black lines) and aberration-corrected LEEM (thick red lines) at a fixed aperture size (see text for details). The vertical dashed lines indicate the $\Delta Z_{A \Phi}$ defocus values. The vertical dashed-dotted lines indicate the $\Delta Z_{\phi}$ defocus values. All calculations were performed with $E_0 = 10 \text{ eV}$ and $\Delta E = 0.25 \text{ eV}$.

We find that the contrast of the amplitude object (Fig. 3.10a) and the $\pi$ phase object (Fig. 3.10b) changes by less than 10% over the full defocus range considered here. The resolution, on the other hand, changes about a factor of 3 with defocus. The optimum resolution is obtained with defocus values close to $\Delta Z_{A \Phi}^3$ defocus for standard LEEM (Fig. 3.10, black vertical dotted lines) and $\Delta Z_{A \Phi}^5$ defocus for aberration-corrected LEEM (Fig. 3.10, red vertical dotted lines). The contrast of the weak phase object shown in Fig. 3.10c is significantly weaker compared to the strong phase object in Fig. 3.10b and varies by about a factor of 4 over the full defocus range. The maximum contrast is obtained close to $\Delta Z_\phi^3$ and $\Delta Z_\phi^5$ defocus for standard and aberration-corrected LEEM, respectively. The defocus value that gives the weakest contrast also produces the best resolution. Scherzer defocus yields an increase of contrast by about a factor of 4 compared to the contrast at the infocus condition. The resolution at Scherzer defocus is about 25% worse compared to that at zero defocus. This discussion shows that in the case of weak phase objects a change in defocus causes a significant change in resolution and in contrast. In addition, it shows that the latter two variables cannot be optimized at the same time. The appropriate defocus value, therefore, depends on whether resolution or contrast is to be optimized.

The calculated resolutions (Fig. 3.9) follow a $1/\alpha$ behavior similar to the resolution obtained with the geometric optics method up to the first minimum, independent of the nature of the object. After reaching a minimum, the resolution plot does not rise like the resolution plot obtained with the geometric optics method. Instead it is flat with some oscillations that damp out with increasing aperture an-
The cause of these oscillations is the sign change of the CTF due to the wave aberrations as a function of spatial frequencies (see Fig. 3.1). The flattening is due to the damping of the chromatic envelope for larger $q$. The exponential envelope functions due to chromatic aberrations create an information limit which is related to the spatial frequency at which the amplitude in the CTF is damped below a critical value. No more contrast is transmitted to the image as the acceptance angle increases beyond the information limit, i.e. the resolution does not decay nor improve any further as the transmission increases. In practice, however, the contrast will decay with increasing acceptance angle due to increased background intensity in the image. The oscillations are more pronounced for the aberration-corrected case, because the damping due to the chromatic envelope function is weaker. This weaker damping gives rise to a greater separation in spatial frequency between the first zero crossing of the CTF and the information limit.

Converting the spatial frequencies into acceptance angle values using $q = \alpha/\lambda$ allows one to compare the behavior of the resolution plot with the behavior of the CTF. Figure 3.11 compares the resolution of amplitude and phase objects with the real part and the imaginary part of the corresponding CTF. These plots show that the resolution generally improves up to the first zero crossing of the relevant part of the CTF; although not strictly so for the $\pi$ and $\pi/2$ phase objects. A sign change of the amplitude in the CTF causes a decay of the resolution until the amplitude switches sign again. At this point the amplitude has the same sign as the starting amplitude and the resolution improves again. This behavior continues until the oscillations in the amplitude of the CTF are damped out by the chromatic envelope function and the resolution stays at a constant level. Figure 3.11 shows clearly that the positions of the zero crossings of the real part of the CTF coincide with the positions of the maxima and the minima of the resolution of the amplitude object. That supports the interpretation that the imaginary part of the CTF plays a minor role for amplitude objects. On the other hand, the positions of the maxima and the minima in the resolution of a $\pi/20$ phase object (Fig. 3.11, green solid line, bottom), a weak phase object, coincide with the positions of the zero crossings of the imaginary part of the CTF. In the case of the resolution of a $\pi$ phase object and a $\pi/2$ phase object the situation is not as clear as for the weak phase object and the amplitude object. For strong phase objects, like a $\pi$ phase object, both contributions of the CTF, real and imaginary, can play a role and need to be considered. These examples demonstrate how imaging of weak phase and arbitrary objects can differ, and that the distinction must be made.

The geometric optics model gives a reasonable estimate for the resolution limit and the optimum acceptance angle. However, it uses the Rayleigh resolution limit which is found to be a conservative estimate compared to the 40% smaller resolution limit determined using the 84/16 criterion with the CTF method. Further-
3.3. Image Contrast Calculations in LEEM

Figure 3.11: Comparison of the resolution of a $1 : 1/\sqrt{2}$ amplitude object (thin black dashed line, top), a $\pi$ phase object (thick orange solid line, top), a $\pi/2$ phase object (purple dotted line, top), and a $\pi/20$ phase object (green solid line, bottom) with the real part (red solid line, center) and the imaginary part (blue dotted line, center) of the corresponding CTF. The calculations were performed for aberration-corrected LEEM at in-focus condition with $E_0 = 10$ eV and $\Delta E = 0.25$ eV.
more, it underestimates the best achievable resolution by about 60 % for standard and 50 % for aberration-corrected LEEM compared to the more accurate results obtained with the CTF formalism. Connected to this, we find that the optimum aperture angle is underestimated by the geometric optics method by about 30 % compared to the CTF results. The acceptance angle is related to the spatial frequency by $a = \lambda q$. The exact optimum aperture angle for given conditions can therefore easily be extracted from the zero crossings of the corresponding CTF plot and do not require a full CTF resolution calculation. The inverse of these optimum acceptance angles gives a good upper limit of the achievable resolution. The CTF approach also allows one to consider the effects of defocus as well as the effects due to the nature of the object (e.g. weak phase object, strong phase object) on the aperture angle and resolution. These considerations are not addressed by the geometric optics approach.

The best theoretically achievable resolution as calculated by the CTF formalism for aberration-corrected LEEM at in-focus condition is about 0.6 nm for amplitude objects and strong phase objects. The best obtainable resolution in aberration-corrected LEEM is lowered to 0.5 nm for weak phase objects at in-focus condition and for amplitude and strong phase objects at their respective Scherzer defocuses.

**Off-axis aberrations**

The magnetic prism arrays in the IBM LEEM used to separate the incoming from the reflected electron beam introduce second-order aberrations such as image tilt and off-axis astigmatism [8]. In order to see the effects of these factors on the image formation we performed additional calculations which explicitly include both magnetic prism arrays. The results of ray tracing $10^5$ electrons through the entire imaging system from points on the sample that are located at different lateral distances from the optical axis are shown in Fig. 3.12 for a starting electron energy of $E_0 = 10$ eV and electron energy spreads of $\Delta E = 0$ eV and $\Delta E = 0.25$ eV. The calculations are performed in an image plane with magnification $M = 1$. The axes of the cones shown in Fig. 3.12 are along the radial displacement direction. Figure 3.12b, d, and f shows profile plots taken across the whole width of the data presented in Fig. 3.12a, c, and e. We take the FWHM of these profile plots as a measure for the effect of off-axis aberrations. The FWHM of these off-axis aberrations is about 11.8 nm, 1.24 nm, and 0.6 nm for distances from the optical axis of 5 mm, 0.5 mm, and 0.25 mm. The detector resolution is about 20 nm, 2 nm, and 1 nm for fields-of-view of 10 mm, 1 mm, and 0.5 mm, respectively. The off-axis aberrations are therefore smaller by about a factor 2 than the spatial resolution of the currently employed multichannel plate (MCP)/phosphorscreen/charge-coupled device detection system. Therefore, off-axis aberrations do not limit the resolution.
3.3. **IMAGE CONTRAST CALCULATIONS IN LEEM**

**Figure 3.12**: Image spread caused by off-axis aberrations at different distances \( d \) from the optical axis for energy spreads of \( \Delta E = 0 \) eV (red) and \( \Delta E = 0.25 \) eV (green) and a starting electron energy of \( E_0 = 10 \) eV. Line profiles taken horizontally across figures (a), (c), and (e) with line widths as broad as the entire images are shown in (b), (d), and (f), respectively.
Effect of a detector on image contrast

So far, the image contrast was calculated by considering only the modifications introduced by the microscope excluding the detection system. It is clear that the final image contrast and resolution depend on the properties of the detector. Here, we use a simple model to show the qualitative effect of a detector on the image contrast and resolution. We convolute the calculated image contrast with Gaussian smearing function with FWHM of \( d = 2 \text{ nm} \), \( d = 5 \text{ nm} \), and \( d = 10 \text{ nm} \). Figure 3.13 shows image contrast plots of a \( 1: \frac{1}{\sqrt{2}} \) amplitude object (a-b) and a \( \pi \) phase object (c-d) for standard LEEM (a and c) and aberration-corrected LEEM (b and d) for Gaussians with different FWHM each. The resolution of the \( 1: \frac{1}{\sqrt{2}} \) amplitude and a \( \pi \) phase object for the different detector smearing functions is shown in Table 3.2. This data shows that the resolution for standard and aberration-corrected LEEM approach each other and become equal with increasing Gaussian smearing. The resolution of a \( \pi \) phase object can even be worse for aberration-corrected LEEM than for standard LEEM if the smearing function is large. Therefore, to fully exploit the improvement in resolution due to aberration-correction a good detection system is necessary.

The Gaussian smearing effect of the detector also degrades the contrast in the final image. For all amplitude objects this is only a minor effect but it is significant in the case of phase objects. We use here the definition of contrast as defined in Section 3.3.2. The contrast extracted from Fig. 3.13c and d is shown in Table 3.3. The decay of contrast is significantly higher for the aberration-corrected case. Already for a smearing function with a FWHM of \( d = 2 \text{ nm} \) the decay is about a factor 4 higher for aberration-corrected LEEM compared to standard LEEM. From this we conclude that the loss of phase contrast for in-focus conditions as experienced by LEEM users in practice could be - at least partially - caused by a detection system with insufficient spatial resolution. These calculations show that a good detection system becomes crucial to profit from the advantages of aberration-corrected LEEM systems. A promising alternative to the conventional used MCP based detectors is the hybrid pixel solid state detector called Medipix. LEEM and PEEM images obtained with a Medipix detector show improved contrast and a resolution better by a factor of about 2 compared to MCP detectors [26].

3.4 PEEM

In the case of PEEM, photo emitted electrons are used to form an image. In contrast to the electrons emitted by an electron gun, the photo emitted electrons are incoherent in time and space. The image intensity for perfectly incoherent illumination, \( I_{\text{inco}} \), is given by the convolution of the intensity distribution in the object plane, \( |f(r)|^2 \), with the square modulus of the point spread function, \( |h(r)|^2 \)
Figure 3.13: Image contrast of a $1:1/\sqrt{2}$ amplitude object (a-b) and a $\pi$ phase object (c-d) for standard LEEM (a,c) and aberration-corrected LEEM (b,d) at in-focus condition for different detector smearing functions (black solid: $d=0$ nm; red dotted: $d=2$ nm; blue dashed: $d=5$ nm; green dotted-dashed: $d=10$ nm). All calculations were performed with optimum aperture angle, $E_0 = 10$ eV, and $\Delta E = 0.25$ eV.
Table 3.2: Resolution of a $1 : 1/\sqrt{2}$ amplitude and a $\pi$ phase object for different detector smearing functions (FWHM) for standard and aberration-corrected LEEM.

\[
\begin{array}{|c|c|c|c|c|}
\hline
\text{FWHM} & \text{w/o} & 2 \text{ nm} & 5 \text{ nm} & 10 \text{ nm} \\
\hline
\text{Resolution} & \\
\text{Amplitude} & \\
nac & 1.9 & 2.3 & 4.3 & 8.5 \\
ac & 0.5 & 1.7 & 4.2 & 8.4 \\
\hline
\pi \text{ phase} & \\
nac & 1.9 & 2.3 & 4.3 & 9.3 \\
ac & 0.6 & 1.8 & 4.8 & 9.8 \\
\hline
\end{array}
\]

Table 3.3: Contrast in % of a $\pi$ phase object imaged for different detector smearing functions (FWHM) for standard and aberration-corrected LEEM.

\[
\begin{array}{|c|c|c|c|c|}
\hline
\text{FWHM} & \text{w/o} & 2 \text{ nm} & 5 \text{ nm} & 10 \text{ nm} \\
\hline
\text{Contrast} & \\
nac & 100 & 68 & 26 & 11 \\
ac & 100 & 16 & 6 & 3 \\
\hline
\end{array}
\]
The Fourier transform of the point spread function is the CTF, $H(q)$. The CTF for a monochromatic point source for the coherent case (compare Eq. 3.1), is given by

$$H_{\text{mono}}(q, \epsilon) = M(q) W(q) K_{\text{total}}(q, \epsilon)$$  \hfill (3.30)

where $K_{\text{total}}(q, \epsilon)$ is given by Eq. 3.6 for the standard case and includes $K_{3C/CC}(q, \epsilon)$ (see Section 3.2.3) for the aberration-corrected case, and $\epsilon$ is the deviation of energy from some nominal energy $E$. For incoherent imaging we need to consider the squared modulus of the PSF. The Fourier transform of the squared modulus of a function is equal to the autocorrelation of the function according to the Wiener-Khinchin theorem [28]. We define the autocorrelation of the CTF $H_{\text{mono}}(q, \epsilon)$ as

$$\left[ \rho_H(q, \epsilon) \right]_{\text{mono}} = H_{\text{mono}}(q, \epsilon) \star H_{\text{mono}}(q, \epsilon)$$  \hfill (3.31)

In the case of a source with finite energy spread we have to integrate the product of $\left[ \rho_H(q, \epsilon) \right]_{\text{mono}}$ with the source energy distribution, $s(\epsilon)$, over the energy

$$\left[ \rho_H(q) \right]_{\text{inco}} = \int_{-\infty}^{\infty} \left[ \rho_H(q, \epsilon) \right]_{\text{mono}} s(\epsilon) d\epsilon$$  \hfill (3.32)

assuring temporal incoherence. The image intensity for perfectly incoherent illumination is then given by

$$I_{\text{inco}} = |f(r)|^2 \otimes |h_{\text{inco}}(r)|^2$$  \hfill (3.33)

where $|h_{\text{inco}}(r)|^2$ is the inverse Fourier transform of $\left[ \rho_H(q) \right]_{\text{inco}}$. $\left[ \rho_H(q) \right]_{\text{inco}}$ cannot be presented in an analytical form anymore but needs to be evaluated numerically. This makes the calculations for the incoherent case more time consuming than for coherent imaging.

The energy distribution of photo emitted electrons depends on the energy of the light used to excite the electrons. In the case of ultraviolet (UV) PEEM the starting energy of the electrons ranges from $E_0 = 0$ eV to a few eV with an energy spread typically between $\Delta E = 1.5$ eV and $\Delta E = 3$ eV depending on the UV source and the work function of the sample. In the case of X-ray PEEM the range of the starting energy becomes larger and also the energy spread gets broader. The broad energy spread in the case of X-ray PEEM is the determining factor for the resolution limit. The situation can be improved significantly, as we will see, by employing energy filtering.
In the case of UV-PEEM the energy distribution can be well approximated by a Gaussian with a FWHM determined by the source energy spread. The energy spread depends on the used light source and the work function of the sample. A light source typically used for UV-PEEM is a Hg discharge lamp with $\hbar \nu \approx 5$ eV. Assuming a typical work function of 4 eV the energy spread is about 1 eV. Here, we use energy spreads of $\Delta E = 1.5$ eV and $\Delta E = 3$ eV to perform the UV-PEEM image contrast calculations. We use the aberration coefficients for an electron starting energy of $E_0 = 1$ eV to perform the following calculations. That assumes that the maxima of the Gaussian energy distributions are located at an energy of $E_0 = 1$ eV.

Figure 3.14 shows a typical image contrast of a $1 : \sqrt{2}$ amplitude object obtained for a spatially and temporally incoherent source. No oscillations or fringes are observed in these contrast profiles contrary to the image contrast plots for a coherent source. Also, no contrast is observed for phase objects.

Here, we calculate the resolution as a function of the acceptance angle in a manner similar to the resolution calculations for coherent imaging (see Section 3.3.3). Figure 3.15 shows resolution plots for Gaussian energy source distributions with spreads of $\Delta E = 1.5$ eV (dotted-dashed and solid lines) and $\Delta E = 3$ eV (dotted and dashed lines) for standard PEEM (blue dotted and dotted-dashed lines) and aberration-corrected PEEM (red dashed and solid lines). The behavior of these resolution plots is similar to the resolution plots obtained with the geometric optics method (see Fig. 3.8), i.e. the resolution increases again for aperture angles larger than the optimum aperture angle at optimum resolution. This is due to the
temporal incoherence of the imaged electrons which prevents the damping of the contrast transfer for higher $q$-values. In the case of an energy spread of $\Delta E = 3 \text{ eV}$ we obtain an optimum resolution of $R = 11.4 \text{ nm}$ and $R = 7.3 \text{ nm}$ for standard and aberration-corrected PEEM, respectively. The best resolution for an energy spread of $\Delta E = 1.5 \text{ eV}$ improves from $R = 8.1 \text{ nm}$ for the standard microscope to $R = 3.7 \text{ nm}$ for the aberration-corrected instrument. Again, the best resolution is observed at an optimum aperture angle. This optimum acceptance angle increases for the aberration-corrected microscope by factors of $1.7$ and $2.5$ compared to the standard case for energy spreads of $\Delta E = 3 \text{ eV}$ and $\Delta E = 1.5 \text{ eV}$, respectively. That means that the increase in transmission can be as large as a factor of 6.

3.4.2 X-RAY PEEM

Incident X-rays create secondary, photo and Auger electrons which can be used for imaging. The energy distribution of emitted secondary electrons can be well approximated by a model function of the form $f \propto E_0/(E_0 + \omega)^4$ with the work function of the sample $\omega$ and the starting energy of the electrons $E_0$ [29]. We use here the normalized energy distribution function $f(E_0) = 6\omega^2 E_0/(E_0 + \omega)^4$ with a typical work function of $\omega = 4 \text{ eV}$. The maximum of that function is located at $\omega/3 \approx 1.33 \text{ eV}$. For practical reasons we consider the function only in the range between $E_0 = 0 \text{ eV}$ and $E_0 = 20 \text{ eV}$ containing $93 \%$ of the total intensity.
For the IBM LEEM instrument [30] the radius in mm of the Ewald sphere in the back-focal plane (bfp) of the objective lens for energy \( E_0 \) in eV is given by
\[
r(E_0) = 60 \sqrt{E_0}.
\]
The radius in mm of the aperture in the bfp of the objective lens is given by \( a(\alpha) = 9.6 \times 10^3 \alpha \) where \( \alpha \) is the aperture angle. Let us assume that for a given energy \( E_0^1 \) the corresponding radius of the Ewald sphere is \( r^1 \). If the radius of the aperture \( a \) is larger than \( r^1 \) the entire Ewald sphere corresponding to \( E_0^1 \) is transmitted. However, if the radius of the aperture becomes smaller than \( r^1 \) the Ewald sphere corresponding to \( E_0^1 \) is only partially transmitted. That means if the following condition is true the Ewald sphere is only partially transmitted:
\[
\frac{a^2}{r^2} = 2.5 \times 10^4 \frac{\alpha^2}{E_0} \leq 1
\]

Therefore, we have to consider two conditions of the energy distribution function which are given by
\[
f(E_0) = \begin{cases} 
\frac{6 \omega^2 E_0}{(E_0 + \omega)^4} & \text{if } 2.5 \cdot 10^4 \frac{\alpha^2}{E_0} > 1 \text{ and } E_0 \geq 0 \\
\frac{1.5 \times 10^5 \omega^2 \alpha^2}{(E_0 + \omega)^4} & \text{if } 2.5 \cdot 10^4 \frac{\alpha^2}{E_0} \leq 1 \text{ and } E_0 \geq 0
\end{cases}
\]

(3.34)

Using this expression for the energy distribution function we calculated the resolution similar to the case of UV-PEEM discussed above. Figure 3.16a shows resolution plots as a function of the aperture angle for standard (dotted-dashed and dashed lines) and aberration-corrected (dotted and solid lines) X-ray PEEM, respectively. The resolution was calculated using the energy distribution function \( f(E_0) \) with \( E_0 \) ranging from \( E_0 = 0 \) eV to \( E_0 = 20 \) eV. The resolution limit is \( R = 9.8 \) nm and \( R = 8.2 \) nm for a standard and an aberration-corrected microscope (black dotted-dashed and dotted lines). We also calculated the resolution obtained by energy filtering with an energy window of \( \Delta E = 2 \) eV. In practice the slit of the energy filter in the IBM LEEM [30] might impose a maximum acceptance angle in the bfp of the objective lens and therefore limit the resolution to a value above the theoretical minimum. It is clear that energy filtering improves the resolution limit significantly to \( R = 7.4 \) nm and \( R = 3.5 \) nm for standard and aberration-corrected LEEM.

Figure 3.16b shows plots of the resolution as a function of the transmission. The transmission is given by
\[
T = \frac{1}{C_{20}} \sum_{m}^{i=1} f(E_{0i})
\]

(3.35)

where \( E_{01} \) and \( E_{0m} \) are the energy at the start and the end point of the corresponding energy window and \( C_{20} = \sum_{n}^{i=1} 6 \omega^2 E_{0i} / (E_{0i} + \omega)^4 \) with \( E_{01} = 0 \) eV and
3.4. PEEM

\[ E_0 = 1 \text{ eV} \]

Transmission at optimum aperture angle is improved for the aberration-corrected instrument compared to the standard microscope. The higher optimum resolution in the case of energy filtering comes with lower transmission.

In Fig. 3.17a we compare the behavior of the ultimate resolution at optimum aperture angle for PEEM and LEEM. The graph shows resolution of amplitude objects in different PEEM modes and resolution of amplitude and phase objects in LEEM with \( E_0 = 10 \text{ eV} \) and \( \Delta E = 0.25 \text{ eV} \) at zero defocus for non-aberration-corrected and for aberration-corrected microscopes. The LEEM data closely follows a \( R_{\text{LEEM}}^{\text{opt}} = 0.42\lambda/\alpha \) behavior. In the case of PEEM, however, a \( R_{\text{PEEM}}^{\text{opt}} = 0.61\lambda/\alpha \) behavior is observed. For aberration-corrected LEEM the resolution falls closer at the 0.42\( \lambda/\alpha \) limit compared to the data for standard LEEM because additional limiting effects due to chromatic and spherical aberrations are more dominant in the latter case. In PEEM the dominant limiting factor is a broad energy spread in combination with chromatic aberrations. We conclude that the resolution for coherent imaging, i.e. LEEM, is limited by the Gaussian width of the central Airy disk \( 2\sigma = 0.42\lambda/\alpha \) (see discussion in Section 3.3.3 on resolution). The resolution limit for PEEM is defined by \( 2\sigma = 0.61\lambda/\alpha \), that is the width of a Gaussian fitted as envelope to the maxima of the entire PSF, i.e. the Airy pattern with chromatic and spherical aberrations. Such a Gaussian envelope neglects the oscillations in the...
3. Image Formation in LEEM and PEEM

Figure 3.17: (a) Optimum resolution of different objects as a function of optimum aperture angle for a standard (full symbols) and an aberration-corrected (empty symbols) microscope in LEEM and PEEM. The starting electron energy is $E_0 = 10$ eV for LEEM and $E_0 = 1$ eV for PEEM, all at in-focus condition. The energy spread for LEEM is $\Delta E = 0.25$ eV. The resolution limit for LEEM (dashed line) and for PEEM (dotted line) are also indicated. (b) Ultimate resolution and optimum aperture angle of a $1 : 1/\sqrt{2}$ amplitude object as a function of starting electron energy for standard (full symbols) and aberration-corrected (empty symbols) LEEM at zero defocus. The solid curves are fits to the resolution data and the dotted curves are fits to the aperture angle data (see text for details).

PSF appropriate for incoherent imaging. Hence, a good estimate for the ultimate resolution for a given aperture angle and starting electron energy of 10 eV can be obtained from $R_{\text{LEEM}}^{\text{opt}} = 0.42 \lambda / \alpha$ for LEEM and from $R_{\text{PEEM}}^{\text{opt}} = 0.61 \lambda / \alpha$ for PEEM without the need of a full CTF image calculation.

Figure 3.17b shows the energy-dependence of ultimate resolution and optimum aperture angle of an amplitude object in standard (full symbols) and aberration-corrected (empty symbols) LEEM. We find that the resolution behaves according to power laws of the form $R_{\text{nac}}(E) = 0.83 E^{-0.49} + 1.62$ and $R_{\text{ac}}(E) = 0.34 E^{-0.74} + 0.49$ (solid curves in Fig. 3.17b) for standard and aberration-corrected LEEM, respectively. We know from the discussion above that $R \propto 1 / \alpha$. We fit the energy-dependent optimum aperture angle data with functions of the form $\alpha(E) = a / R(E)$ and obtain $\alpha_{\text{nac}}(E) = 0.0042 / R_{\text{nac}}(E)$ and $\alpha_{\text{ac}}(E) = 0.0039 / R_{\text{ac}}(E)$ (dotted curves in Fig. 3.17b) for standard and aberration-corrected LEEM, respectively. The fits reproduce the data very well and can therefore be used as good estimates for ultimate resolution and optimum aperture angle in LEEM for a given starting electron energy without the need of a full-wave-optical image calculation.

3.5 Conclusions

We have introduced an extended Contrast Transfer Function (CTF) approach for the calculation of image formation in Low Energy Electron Microscopy (LEEM). This approach considers chromatic and spherical aberrations up to fifth order,
appropriate for image formation in state-of-the-art aberration-corrected LEEM. We have derived a set of four Scherzer defocus values for weak phase objects, and for strong phase and amplitude objects, in both non-aberration-corrected and aberration-corrected LEEM. Using this extended CTF formalism, we have calculated contrast and resolution of one-dimensional and two-dimensional pure phase, pure amplitude, and mixed phase and amplitude objects in LEEM. We show that adjusting defocus causes a change in resolution and contrast. In the case of weak phase objects choosing defocus is a trade-off between optimum resolution and optimum contrast. For strong phase and amplitude objects, the relevant Scherzer defocus setting optimizing resolution without negatively affecting contrast. The CTF approach was also adapted to consider the case of incoherent imaging in Photo Electron Emission Microscopy (PEEM). Based on these calculations, we show that the ultimate resolution in aberration-corrected LEEM is about 0.5 nm, and in aberration-corrected PEEM about 3.5 nm. The aperture sizes required to achieve these ultimate resolutions were precisely determined with the CTF method. We conclude that the commonly employed geometric optics model to estimate optimum resolution and aperture size underestimates both parameters by about 50 % and 30 %, respectively, compared to the more accurate results determined with the CTF approach. Furthermore, we find that the energy-dependent resolution in LEEM behaves according to $R_{nac}(E) = 0.83E^{-0.49} + 1.62$ and $R_{ac}(E) = 0.34E^{-0.74} + 0.49$ for a standard and an aberration-corrected microscope, respectively. A good estimate for the energy-dependent optimum aperture angle is given by $\alpha_{nac}(E) = 0.0042/R_{nac}(E)$ and $\alpha_{ac}(E) = 0.0039/R_{ac}(E)$ for standard and aberration-corrected LEEM, respectively. A good estimate for the resolution in PEEM can be obtained from $R_{PEEM} = 0.61\lambda/\alpha$, independent of starting electron energy and aberration-correction.
REFERENCES


