Bachelor thesis

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High resolution differential phase contrast scanning X-ray microscopy with a single photon counting 2D detector

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Aware of criminal liability for making untrue statements I declare that the following thesis was written personally by myself and that I did not use any sources but the ones mentioned in the dissertation itself.
Supervisor’s review
Reviewer’s review
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1 Introduction

A number of X-rays applications is known beginning from the everyday life, through industry and medicine to the sophisticated scientific problems. The recent decades have brought new ideas of obtaining the bright X-ray source of tunable energy. The third generation synchrotron light sources have such features allowing for variety of experimental methods. Amongst them there is the X-ray microscopy which allows scientists to make one step farther in unwrapping the mystery of matter. Over the years the X-ray absorption was the leading technique in imaging. Yet, with rapid increase of computational power, other solutions have come into play competing with those so far developed. The capability of resolving the phase problem and the need for other methods allowing for imaging also the weakly absorbing samples caused the phase contrast imaging to develop.

The goal of this thesis is to investigate the differential phase contrast scanning X-ray microscopy by presenting its theoretical background and applying it experimentally. An important feature of an experiment will be utilization of the single photon counting detector PILATUS 6M. Data collected from this 2D detector will be then processed by self-developed software. The thesis consists of three parts.

In the first part the X-rays properties and applications in imaging are discussed. The fundamentals of X-rays propagation in free space are given including the derivation of the scanning image formation. One can find introductions both to the phase contrast imaging method as well as to the coherent diffractive imaging with its novel technique, ptychography.

The second part of the thesis gives an overview of reconstruction algorithms: the extended Ptychographical Iterative Engine and the Differential Phase Contrast Analysis Tool. The latter has been already implemented in C++ programming language and applied in the data analysis in experimental part.

In the third part of the thesis, the experiment of the differential phase contrast scanning transmission X-ray microscopy is presented. It was conducted at beamline P11 of PETRA III synchrotron light source at Deutsches Elektronen-Synchrotron in Hamburg, Germany. Two samples were measured: the zone plate and the Siemens star. The description of the experiment is followed by the differential phase contrast analysis of these scans, utilizing the developed DPC Analysis Tool.
2 X-rays properties and applications in imaging

In 1895 Wilhelm Conrad Röntgen discovered X-rays [1]. In his further experiments, he showed that the high-energy electrons impinging on a material object (target) are responsible for production of this invisible and so far unknown kind of radiation. This discovery was undoubtedly a real milestone which has influenced not only many branches of science but also medicine and industry. Over following years, a significant number of scientists have laid the foundations for theory of X-rays propagation proposing the broad variety of future applications. Surely, the X-rays have become an invaluable probe of the structure of matter, giving the opportunity to reveal the mysteries of nature beginning from the simple compounds through to more complex and prominent examples, such as DNA [2]. The goal of this chapter is to outline the most important X-rays characteristics and shed a light on the basics of the interaction of X-rays with matter.

2.1 X-rays propagation and their interaction with matter

X-rays are part of the electromagnetic radiation spectrum with wavelengths in range from 10 to 0.01 nm or respective energies of 100 eV to 100 keV [3]. In many cases, one is interested in the monochromatic X-ray beam propagating along the \( z \)-axis.

Considering the X-rays application in imaging, a common classification with respect to X-rays energy ranges has been done as follows

- Soft X-rays: \( E \lesssim 1 \text{ keV} \),
- Medium-energy X-rays: \( 1 \text{ keV} \lesssim E \lesssim 5 \text{ keV} \),
- Hard X-rays: \( 5 \text{ keV} \lesssim E \lesssim 20 \text{ keV} \).

Energies presented above are the most frequently used in terms of X-ray microscopy applications, hence those above 20 keV are not considered here.

Now, we shall briefly discuss the ways X-rays interact with matter. Among several interaction mechanisms, these in the range of photon energies regarded in this thesis are absorption, elastic and inelastic scattering.

Considering absorption, the fully absorbed X-ray photon causes the ejection of the photoelectron from an atom in the material, thus the atom becomes ionized. The result of the consecutive filling of this hole is a photoemission of a characteristic wavelength.

In case of elastic scattering, the incident photon is deflected while its energy is conserved. In other words, the wavelength of the scattered wave is equal to the incident one.

In inelastic scattering, finally, the energy and the momentum of the system consisting of the photon and the scattering electron is preserved. The impinging photon energy and momentum
are partly transferred to the electron which scatters the photon off at an angle with reduced energy. This scattering process is known as the Compton effect. Yet, the elastic scattering of X-rays is the main process that is exploited in investigations of the structure of materials [2].

The discussion about the interaction of X-ray photons with matter can be held at the atomic level. However, since X-rays are also electromagnetic waves, it should be expected to observe some kind of refraction phenomena at boundaries between different media. Description of such refractive processes requires the media of interest being homogeneous with sharp interfaces between them. Each of them is supposed to have its own refractive index \( n \). In vacuum the refractive index is defined to be equal to one. For X-rays the deviation of \( n \) from unity of \( n \) is very small. Generally for X-rays, the refractive index is expressed as follows

\[
n = 1 - \delta + i\beta
\]  

(2.1)

where \( \delta \) is of order of \( 10^{-5} \) in solids and only around \( 10^{-8} \) in air. The imaginary part \( \beta \) is usually much smaller than \( \delta \) [2]. These parameters \( \delta \) and \( \beta \) are sometimes called the refractive index decrement and absorption index, respectively, and may be derived from the scattering and absorption properties of the considered medium [4].

Another important property of X-rays is their coherence. Undoubtedly, considering an X-ray beam in a perfect plane wave state is an idealization. Thus, the concept of the coherence length of the real beam has been introduced, since the real beam may deviate from an ideal plane wave in two ways: not only is it imperfectly monochromatic but also the direction of its propagation is not defined precisely. Taking this into account, one can consider two, propagating in exactly the same direction, plane waves with slightly different wavelengths, \( \lambda \) and \( \lambda - \Delta\lambda \). In regarded case one can define the longitudinal coherence length \( L_L \) equal to the closest distance between two points where these two waves are in phase and out of phase:

\[
L_L = \frac{1}{2} \frac{\lambda^2}{\Delta\lambda}
\]

(2.2)

Alternatively, two waves of the same wavelength can are regarded with slightly different directions of propagation, differing by an angle of \( \Delta\theta \). The similar question may arise, what is the closest distance between two point in phase and out of phase? In reply to this, the transverse coherence length \( L_T \) is defined. Suppose that the different directions of propagation are caused by the fact that the two waves originate from two different points on the source, say a distance \( D \) apart. Then, if the distance from the observation point to the source equals \( R \), we have \( \Delta\theta = D/R \) which leads to the following formula

\[
L_T = \frac{1}{2} \frac{\lambda}{D/R} = \frac{\lambda}{2} \frac{R}{D}
\]

(2.3)

Regarding typical values of the coherence lengths, one can consider a third generation synchrotron light source of the vertical source size around 20 \( \mu \text{m} \) and the experiment performed
some 100 m away, especially for coherence applications. Given 1Å X-rays, the transverse coherence length is approximately equal to 250 µm in the vertical plane. In order to calculate the longitudinal coherence length one needs to make an additional assumption about the device used to monochromate the beam. The quality of the crystal or grating used in the monochromator defines $\Delta \lambda/\lambda$. In crystal monochromators this can exceed $10^6$, hence $L_L$ is much more than 10µm also for 1Å X-rays.

The crucial consequence of a finite coherence length is that it establishes the upper limit on the separation of two objects if they are supposed to give rise to interference effects. Several modern imaging methods which utilize such coherent X-ray beams will be mentioned in following sections [2].

The propagation of X-rays may be described considering a scalar wave field $\psi$. One can formulate an equation for a plane wave with amplitude $\psi_0$ propagating in free space along the $z$-direction. In the stationary case it can be written as

$$\psi(z) = \psi_0 \exp(-ikz) \quad (2.4)$$

where $k = 2\pi/\lambda$ is the wave number.

Regarding X-rays propagation in a homogeneous material with refractive index $n$, the wave equation is given as

$$\psi(z) = \psi_0 \exp(-inkz) \quad (2.5)$$

Bearing in mind the definition of the complex refractive index from Eq. [2.1], this can be rewritten explicitly as

$$\psi(z) = \psi_0 \exp(-ikz) \exp(+i\delta kz) \exp(+\beta kz) \quad (2.6)$$

In Eq. (2.6) one can identify the first term as the free space propagation, the second term is responsible for the phase shift imposed on the wave field by the material and the third term is the amplitude attenuation. This leads us to a conclusion that X-rays traversing a material with index of refraction $n$ and the lateral thickness profile function $\Delta z(r)$ are attenuated and phase shifted in comparison to propagation in vacuum. Thus, it is worth describing the X-rays interaction with a sample using a multiplicative transmission function $O(r)$ which operates on an incoming wave field $\psi_{in}$, hence the outgoing wave field $\psi_{out}$ is produced as

$$\psi_{out} = O(r)\psi_{in} \quad (2.7)$$

where the specimen (transmission) function is given by

$$O(r) = \exp(+i\delta k\Delta z) \exp(+\beta k\Delta z) \quad (2.8)$$

This introductory survey on X-ray propagation and their interaction with matter shall serve as foundations for the consecutive sections within this thesis, once the sample influence on X-rays will be introduced.
2.2 Synchrotron radiation characteristics

Progress in both understanding of X-rays physics and their experimental exploitation was constant from their discovery till the mid 1970s. The main limitation then appeared to be the source remaining unchanged since about 1912. Though, in the 1970s it was realized that the synchrotron radiation emitted from charged particles circulating at relativistic speeds along the storage rings (constructed for high energy nuclear physics experiments) was potentially much more intense and universal in comparison to those sources considered until then.

Although the synchrotron radiation is named after a particular type of particle accelerator, it describes generally radiation from charged particles travelling at ultra-high speeds in applied magnetic fields that force them to follow bended paths. In storage rings the synchrotron radiation is produced because electrons or positrons are kept circulating at constant energy. It may be emitted either in a bending magnet necessary for keeping the particles in a closed orbit, or in so-called insertion devices such as wigglers and undulators which are located in the straight sections of the storage ring. The major feature of these devices is an alternating magnetic field which forces electrons (or positrons) to follow oscillating paths rather than moving along a straight line. The main difference between them stems from distinct amplitudes of the oscillation which for a wiggler is rather large, so the radiation from different wigglers add incoherently, whereas in case of undulators the small-amplitude oscillations from the passage of a single electron result in a coherent addition of radiation produced in each oscillation [2].

Among variety of parameters defining storage ring properties one can consider the total emitted power by the circulating electrons

\[ P_{\text{TOT}} = \frac{2q^2 c^7 E^4}{3m_0^3 R^2} = \frac{2q^2 c^3 R^2}{3\gamma^4} \]  \hspace{1cm} (2.9)

where by \( q \) charge of the circulating particle is denoted, \( m_0 \) represents rest mass of circulating particle, \( c \) speed of light, \( R \) is a radius of the storage ring, \( E = \gamma m_0 c^2 \) refers to energy of particle circulating at relativistic speed and \( \gamma \) is the Lorentz factor.

Another important quantity which allows to determine the quality of the X-ray beam from different sources is called the brilliance. It incorporates the following properties: the number of photons emitted per second, the collimation of the beam (usually in milli-radian), the source size (in \( \text{mm}^2 \)) and the spectral distribution. Concerning the latter, some X-ray sources produce very smooth spectra, while others have peaks at certain photon energies. Therefore it matters, when comparing them, what range of photon energies contributes to the measured intensity. The convention is to define the photon energy range as a fixed relative energy bandwidth (BW), chosen to be 0.1% [2]. Eventually, the brilliance is then defined as

\[ \text{Brilliance} = \frac{\text{photons/second}}{(\text{mrad})^2(\text{mm}^2 \text{ source area})(0.1\% \text{ BW})} \]  \hspace{1cm} (2.10)
and is therefore the flux per unit source area and unit solid angle. Taking into account even slight imperfections of the optics of a beamline, the brilliance at the experimental station will fall short of theory. The brilliance of a third generation beamline is about $10^{20}$ photons/s/mrad$^2$/mm$^2$/0.1%BW and this is the main reason why synchrotrons have become such important research tools.

One can also meet another twin quantity, called the peak brilliance which, comparing to the brilliance definition (Eq. (2.10)), includes the inverse proportionality to the electron bunch length.

Brilliance $\propto \frac{\text{Flux}}{\text{source area}}$ (2.11)

Peak Brilliance $\propto \frac{\text{Flux}}{(\text{source area})(\text{electron bunch length})}$ (2.12)

In Fig. 2.1, the peak brilliance with respect to the photon energy is presented for a few X-ray radiation facilities. Considering the third generation of synchrotron light sources, the upper limit for the peak brilliance reaches the order of magnitude of $10^{23}$ photons/s/mrad$^2$/mm$^2$/0.1%BW. However, this limit is significantly exceeded by the emerging fourth-generation facilities, free electron lasers, operating in far ultraviolet and X-ray regime.

Summarizing, the most important characteristics of synchrotron radiation are:

- broad spectrum: from infrared ($10^5\text{Å}$) to hard X-rays ($10^{-3}\text{Å}$),
- extremely high intensity,
- linear horizontal polarization,
- strong collimation due to relativistic effect,
- pulsed time structure,
- small source dimensions,
- beam stability,
- clear environment,
- source of tunable radiation.
2.3 X-ray Microscopy

Microscopy exploiting X-rays constitutes a complementary technique to imaging with visible light and electrons. Its distinctive merits open a broad field of applications. This section is meant to provide a brief overview of major capabilities of this technique.

Generally speaking, X-ray microscopy allows the imaging of structures on length scales between those probed by optical (< 1µm) and electron (∼ 1Å) based techniques [2]. Thus, in comparison to visible light imaging, X-ray microscopes have the potential for higher spatial resolution (due to shorter wavelength). Common to all microscopies is the need for efficient focusing optics. Optical instruments applicable in case of X-rays are Kirkpatrick-Baez mirrors, arranged to focus successively in orthogonal planes, and lenses such as compound refractive lenses (working best at energies above 10 keV) or Fresnel zone plates (FZP). The latter may be designed to operate over a substantially wide range of photon energies, including the soft part of X-rays spectrum, crucial in imaging of biological tissue [2]. The essential information about the optics of a Fresnel zone plate one can find in section 2.4.

The Fresnel zone plate may be utilized in X-ray microscopes in two distinct ways. Either it can be used to focus a beam down to a small focus, or it can magnify an image. This

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1 Image source: http://hasylab.desy.de/images/content/e208/e209/index_eng.html
defines two different techniques: the scanning transmission X-ray microscopy (STXM) and the full-field transmission X-ray microscopy (TXM), respectively. The basic experimental setup of both methods is depicted in Fig. 2.2.

**STXM: scanning transmission x-ray microscope**

- undulator source with monochromator
- sample
- bright field cone
- objective zone plate
- detector

**TXM: transmission x-ray microscope**

- bending magnet source
- condenser zone plate
- sample
- objective zone plate
- pixelated detector

Fig. 2.2: The schematic view of the experimental setup for the STXM and the full-field TXM microscopes. Taken from [3].

In STXM the sample is placed on a mechanical stage which allows it to be raster scanned through the focused beam [2]. Different signals such as the transmitted intensity or the emitted X-ray fluorescence are collected for each scan point and form the image. In order to achieve high spatial resolution, one needs to make the FZP create a small spot. Therefore it needs to be illuminated coherently which explains why STXM is commonly operated at undulator beamlines. Although image acquisition is slower due to the raster scan approach, the field of view is easily variable and the dose delivered to the sample is significantly reduced [3]. The image in STXM can be formed either from absorption or phase contrast in a transmission geometry, or by collecting the fluorescence (scanning X-ray fluorescence microscopy).

Moving to the alternative geometry of using the FZP as a magnifying lens, a full-field image of the sample is collected in a single exposure on a two-dimensional pixelated X-ray detector. Direct detection with high spatial resolution is only possible below 1 keV, otherwise indirect detection with a fluorescence screen must be performed. The full-field TXM has an advantage of speed over the STXM and it does not require highly parallel (i.e. transversely coherent) incident beam [2]. It can be operated at bending magnet source beamlines. Due to fast acquisition of images (exposure times in the range of seconds), this technique is well suited for tomographic imaging applications. By contrast to STXM, however, the full-field TXM has the limited field of view and the radiation dose delivered to the sample is high for images of good quality. The
latter is caused by an inefficiency in collecting the sample information by the objective lens [3].

Within this thesis the scanning transmission X-ray microscopy is exploited and the foundations for diffraction based microscopy as an emerging type of lensless imaging will be given.

2.4 Optics of Fresnel Zone Plate

In section 2.3 we learnt about the application of a Fresnel zone plate (FZP) in two X-ray microscopy techniques. Let us focus for a while on the principles underlying the operation of this lens.

A Fresnel zone plate is a circular diffraction grating. The aim is to achieve a constructive interference at the lens focus, which is determined by the path difference to the focus from radiation passing through adjacent open zones. The radius \( r_m \) of a FZP is defined in such a way that the waves radiated from a particular zone and arriving at the focus have an integrated phase shift of \( m\pi \), equivalent to \( m\lambda/2 \), relative to the incident beam. Yet, waves radiated from successive zones tend to interfere destructively. What brings a large increase in the radiation arriving at the focus is applying the material with alternate zones which either completely absorbs the incident beam, or introduces the phase shift of \( \pi \). These two alternative approaches are usually referred to as absorption and phase Fresnel zone plate, respectively [2]. From now on the term Fresnel zone plate will refer to its absorption version.

The Fig. 2.3 shows a FZP formed from a series of concentric rings with zones between these rings altering from transparent to opaque. We can derive an equation for the radius of the \( m \)'th Fresnel zone. According to Pythagoras’ theorem

\[
r_m^2 + f^2 = \left(f + \frac{m\lambda}{2}\right)^2
\]

from which the solution for the radius of the \( m \)'th zone can be approximated as

\[
r_m \approx \sqrt{m\lambda f}
\]

where \( f \) is a focal length of the lens.

![Fig. 2.3: The scheme of focusing of the parallel beam by the Fresnel zone plate. The radius of the \( m \)'th zone, dependent on the focal length \( f \), is marked. Taken from [2].](image)
Another quantity of interest is the width $\Delta r_M$ of the outermost zone (assuming that the total number of Fresnel zones equals $M$) which can be derived consecutively

$$\Delta r_M = \sqrt{\lambda f \left( \sqrt{M} - \sqrt{M-1} \right)} = \sqrt{\lambda f} \left( \sqrt{M} - \sqrt{M} \sqrt{1 - \frac{1}{M}} \right) \approx \frac{\sqrt{\lambda f}}{2\sqrt{M}} \quad (2.15)$$

According to Eqs (2.14) and (2.15), one can easily find the Fresnel zone plate’s diameter $D$ as

$$D = 2r_M = 2\sqrt{Mf\lambda} = 4M\Delta r_M \quad (2.16)$$

Combining preceding equations together, it is possible to derive the expression for the focal length of the FZP

$$f = 4M \frac{(\Delta r_M)^2}{\lambda} = \frac{D\Delta r_M}{\lambda} \quad (2.17)$$

An important parameter in microscopy is the numerical aperture $NA$, which for the Fresnel zone plate is equal to a half-angle of the maximum cone of light that exits the lens:

$$NA = \frac{\lambda}{2\Delta r_M} \quad (2.18)$$

One can ask about the resolution of FZP which obviously limits the final resolution of a microscope. Recalling the Rayleigh criterion, a perfect lens can resolve the smallest detail given by

$$\Delta x = 1.22 \frac{\lambda f}{D} \quad (2.19)$$

and if substituting $\Delta r_M = f\lambda/D$ derived from Eq. (2.17), one obtains the resolution of the Fresnel zone plate operating at X-ray wavelengths

$$\Delta x = 1.22\Delta r_M \quad (2.20)$$

An explicit conclusion from the last equation is that the best transverse resolution is achieved by minimizing the width of the outermost zone and is independent of the wavelength as long as $\Delta r_M \gg \lambda$.

One can also think of the FZP as a variable period diffraction grating. So far, only the first, positive diffraction order has been considered in this section and all derived formulae apply to that case. Other diffraction orders exist and this fact has to be taken into account in any practical implementation of an absorption Fresnel zone plate [2]. Furthermore, one should point out that due to diffractive nature, the largest part of the illumination passes through the FZP undiffracted. To isolate the first order diffracted focus, one usually uses an arrangement of a central stop (CS) and an order-sorting aperture (OSA). The minimum OSA diameter is determined by its required distance from the focus. So is the diameter of the central stop, which ought not to cover more than a half of the zone plate’s diameter. Otherwise, the focal spot quality becomes degraded [3].
2.5 Mathematical definitions and theorems

The aim of this section is to systematize mathematical definitions and theorems used in subsequent part of this thesis.

Firstly, the forward Fourier transform of a function \( g(r) \) is given by

\[
\mathcal{F}\{g(r)\} = G(\omega) = \int d^2r \, g(r)e^{i\omega r}
\]  

(2.21)

The corresponding inverse Fourier transform

\[
\mathcal{F}^{-1}\{G(\omega)\} = g(r) = \int d^2\omega \, G(\omega)e^{-i\omega r}
\]  

(2.22)

Secondly, the convolution \( c(r) \) of two functions \( g(r) \) and \( h(r) \) with respect to the variable \( r \) is defined as follows

\[
c(r) = g(r) \otimes h(r) = \int d^2r' \, g(r')h(r - r') = \int d^2r' \, g(r - r')h(r')
\]  

(2.23)

The convolution integral is sometimes referred to as the folding integral.

Finally, the convolution theorem is recalled. It states that the Fourier transform of the convolution of two functions \( g(r) \) and \( h(r) \) is equal to the multiplication of their individual Fourier transforms

\[
C(\omega) = \int d^2r \, c(r)e^{i\omega r} = \int d^2r \, g(r) \otimes h(r) \, e^{i\omega r} = G(\omega)H(\omega)
\]  

(2.24)

Note that by notation \( r \) and \( \omega \), the real space and the Fourier space coordinates, respectively, have been defined. They refer to two dimensions, which will be the case in following sections.

2.6 X-ray wave field propagation in free-space

Let us consider the problem of diffraction of light by an aperture \( \Sigma \) in an infinite opaque screen. As shown in Fig. 2.4 a wave disturbance \( \psi_1 \) impinges on the screen and the aperture from the left and the wave field \( \psi_2 \) in the plane \([r_2]\) is to be calculated. The successive formulae will be derived provided that the wave field is monochromatic.

To propagate the wave field from plane \([r_1]\) to \([r_2]\) the Fresnel-Kirchhoff diffraction formula is used \[2\]

\[
\psi_2(r_2) = \int_\Sigma \, d^2r_1 \, \frac{e^{-ikr}}{i\lambda r} \cos \angle(n, r) \psi_1(r_1)
\]  

(2.25)

where \( r \) is the distance between a particular point in the aperture and the plane \([r_2]\]

\[
r = \sqrt{z^2 + (r_2 - r_1)^2}
\]  

(2.26)

and \( \angle(n, r) \) is an angle between the normal to the surface \( \Sigma \) and the distance vector \( r \). The integral is considered with infinite limits, whereas \( \psi_1 \) is identical to zero except for the aperture. The maximum linear extend of the aperture is equal to \( a \).
If it is assumed that the distance $z$ between aperture and the observation plane is much greater than $a$ ($z \gg a$), the above diffraction formula (Eq. (2.25)) may be initially approximated. Moreover, provided that in the observation plane only a finite region around the optical axis is of interest, then $z$ is much greater than the maximal linear dimension in this region. Therefore, the vectors $n$ and $r$ may be considered as nearly parallel which implies the approximation $\cos \angle(n, r) \approx 1$. Similarly, following these conditions, the distance $r$ in the denominator does not differ much from $z$. All these approximations altogether may be expressed as follows

$$e^{-ikr} \cos \angle(n, r) \approx e^{-ikr}$$

(2.27)

The distance $r$ in exponent cannot be replaced since it is multiplied by the wave number $k$ of a large value and this could cause the rapidly varying phase errors [3].

Simplifying the diffraction formula even further, the specific approximation to the quantity $r$ in the exponent can be considered. Transforming Eq. (2.26) and performing a binomial expansion of the square root, one obtains

$$r = \sqrt{z^2 + (r_2 - r_1)^2} = z \sqrt{1 + \left(\frac{r_2 - r_1}{z}\right)^2} \approx z \left[1 + \frac{1}{2} \left(\frac{r_2 - r_1}{z}\right)^2\right]$$

(2.28)

under the necessary assumption that $\left(\frac{r_2 - r_1}{z}\right)^2 < 1$.

This is referred as to the Fresnel approximation (near-field regime of diffraction). The resulting expression of the wave field propagating in the Fresnel regime is given by

$$\psi_2(r_2) = \frac{e^{-ikz}}{i\lambda z} \int_{\Sigma} d^2 r_1 \psi_1(r_1) \exp\left(-\frac{ik}{2z}(r_2 - r_1)^2\right)$$

(2.29)
Rewriting the above equation more explicitly,
\[
\psi_2(r_2) = \frac{e^{-ikz}}{i\lambda z} \exp \left( -\frac{ik}{2z} r_2^2 \right) \int_{\Sigma} d^2 r_1 \psi_1(r_1) \exp \left( -\frac{ik}{2z} r_1^2 \right) \exp \left( i \frac{k}{z} r_2 r_1 \right)
\]  
(2.30)

one can see that, leaving out the integral pre-factors, the wave field \(\psi_2\) is proportional to the following Fourier transform
\[
\Psi_2(\omega_2) \propto \mathcal{F}_{\omega_2} \left\{ \psi_1(r_1) \exp \left( -\frac{ik}{2z} r_1^2 \right) \right\}
\]  
(2.31)

with frequency \(\omega_2 = (k/z)r_2\)

Analyzing Eq. (2.30), it is feasible to make further simplifications once more rigorous restrictions are to be introduced. Stronger than the Fresnel assumption is the Fraunhofer assumption

\[
z \gg k \max \{ r_1^2 \}
\]  
(2.32)

and when satisfied, the quadratic phase factor in Eq. (2.30) may be neglected as \(\exp \left( -\frac{ik}{2z} r_1^2 \right) \approx 1\). That yields the Fraunhofer approximation, known also as the far-field regime.

\[
\psi_2(r_2) = \frac{e^{-ikz}}{i\lambda z} \exp \left( -\frac{ik}{2z} r_2^2 \right) \int_{\Sigma} d^2 r_1 \psi_1(r_1) \exp \left( i \frac{k}{z} r_2 r_1 \right)
\]  
(2.33)

Similarly to the near-field case, one can see that not taking into account the pre-factors, the resulting wave field \(\psi_2\) is the forward Fourier transform of the wave field \(\psi_1\) within the aperture

\[
\Psi_2(\omega_2) \propto \mathcal{F}_{\omega_2} \{ \psi_1(r_1) \}
\]  
(2.34)

with frequency \(\omega_2 = (k/z)r_2\).

The quantity called the Fresnel number \(F\) has been defined to provide a measure to identify the regime of the observed diffraction:

\[
F = \frac{a^2}{\lambda z}
\]  
(2.35)

where \(a\) is a maximal linear extent of the aperture. The classification is done as in Tab. 2.1.

| Fresnel number | Regime               |
|---------------|----------------------|
| \(F \gg 1\)   | Contact              |
| \(F \approx 1\)| Fresnel (near-field) |
| \(F \ll 1\)   | Fraunhofer (far-field)|

Tab. 2.1: Classification of different diffraction regimes with respect to the Fresnel number.

Let us consider the example depicted in Fig. 2.5 where the scattering calculated from a simple model discs of diameter 5\(\mu\)m is presented. There are two types of idealised discs: perfect absorbers (red) and perfect phase objects (blue).
Fig. 2.5: Simulated X-ray radiographs for illustration of the transition from pure absorption contrast (in the contact regime) via edge-enhancing phase contrast (the near field) and towards the far-field regime (though the Fraunhofer image is not given). An ensemble of small disc-like objects is simulated, some of which are ideal absorbers (red, zero transmission) and the rest is ideal phase objects (blue, no absorption, phase shift $\pi$). The diameter of each discs is $5 \, \mu m$. The gray scale images show simulated radiographs of the phantom object for the illumination by a monochromatic X-ray plane wavelength $1 \, \AA$ at different sample-to-detector distances: 0.1, 1, 10, 100, 1000 mm. Taken from [2].

In contact regime the imaging arises from absorption contrast only. Moving to the near-field (Fresnel) regime and beyond, the phase objects become to be more and more visible. Once the distance from the object to detector is far beyond the Fresnel region (towards the far-field regime), the shape of the diffraction image does not change any more, even though it gets obviously weaker in intensity per unit area of detector [2].

2.7 STXM - scanning image formation

Having discussed the propagation of X-rays in free-space (section 2.6), we turn our attention to the image formation process in scanning transmission X-ray microscope. The Fig. 2.6 shows how such a microscope may be realized.

Since it is requested to obtain a diffraction limited spot size and resolution of the microscope, the source must be sufficiently small and far away from the focusing zone plate. Then the lens is illuminated with a high degree of spatial coherence. If this is fulfilled, the lens focuses then the radiation to a small spot through which the sample is raster scanned. A collected dataset is the transmitted intensity for each scan displacement $R_s$ recorded in a far-field plane from
the focus with the area sensitive detector. In front of the zone plate the central stop is placed. The Fresnel zone plate (FZP) is characterized by the diameter $D_o$ and the outermost zone width $\Delta r_M$. Thus, the cut-off frequency: $\omega_o = \pi/r_M$. As stated in section 2.4 in order to isolate the first order focus, a central stop has to be exploited, which has a diameter $D_{\text{stop}}$.

Similarly to the FZP, the central stop’s corresponding cut-off in the frequency space is then given by $\omega_{\text{stop}} = \frac{D_{\text{stop}}}{D_o} \omega_o$. According to assumed parameters, the objective aperture is described by having a real pupil function

$$p_o(\omega) = \begin{cases} 0, & |\omega| \leq \omega_{\text{stop}} \\ 1, & \omega_{\text{stop}} < |\omega| \leq \omega_o \\ 0, & |\omega| > \omega_o \end{cases} \quad (2.36)$$

The Fresnel zone plate is supposed to be operated in its first diffraction order. Assuming, that the zone plate consists of sufficient number of zones, its influence on the wave field may be regarded as that of a thin lens and therefore is described by a quadratic phase factor [6]. Consequently, the total transmission function of the FZP has the form

$$t(r_1) = p_o(r_1) \exp\left(\frac{ik}{2f_o}r_1^2\right) \quad (2.37)$$

where $f_o$ is the focal length of the first order focus of the zone plate and $r_1$ (according to notation from the Fig. 2.6) expresses coordinates in FZP’s plane.

Now, the comprehensive derivation of the wave propagation to the detector plane will be given. Depending on the considered case, either Fresnel (Eq. (2.29)) or Fraunhofer (Eq. (2.33)) approximation will be used. It should be denoted that constant phase factors, which precede the integrals, will be left out because they do not matter in terms of the final image.
The monochromatic point source of amplitude $\psi_0$ is situated at a point on the optical axis a distance $d_0$ (see Fig. [2.6]) away from the FZP. One can express its spatial distribution by a Dirac delta function [2], defined as follows

$$\delta(r - r_0) = \begin{cases} 
0, & r \neq r_0 \\
\infty, & r = r_0 
\end{cases} \quad (2.38)$$

$$f(r_0) = \int d^2r \ f(r) \delta(r - r_0) \quad (2.39)$$

for an arbitrary function $f(r)$.

Considering propagation of the wave field from this source point to the objective lens and multiplication with the FZP’s transmission function (Eq. (2.37)), one can formulate the wave field $\psi'_1(r_1)$ exiting the zone plate

$$\psi'_1(r_1) = \frac{1}{i\lambda d_0} \int d^2r_0 \ \psi_0 \ \delta(r_0) \exp\left(-\frac{ik}{2d_0}(r_1 - r_0)^2\right) t(r_1) \quad (2.40)$$

Using the Fresnel approximation, one can find the wave field $\psi_2(r_2)$ in the focal plane a distance $d_1$ away

$$\psi_2(r_2) = \frac{1}{i\lambda d_1} \int d^2r_1 \ \psi'_1(r_1) \exp\left(-\frac{ik}{2d_1}(r_2 - r_1)^2\right) \quad (2.41)$$

Substituting Eq. (2.40) into Eq. (2.41), one obtains

$$\psi_2(r_2) = -\frac{1}{\lambda^2 d_0 d_1} \int d^2r_1 \ \int d^2r_0 \ \psi_0 \ \delta(r_0) \exp\left(-\frac{ik}{2d_0}(r_1 - r_0)^2\right) t(r_1) \ \exp\left(-\frac{ik}{2d_1}(r_2 - r_1)^2\right) \quad (2.42)$$

In regarded problem, the lens law must be satisfied

$$\frac{1}{d_0} + \frac{1}{d_1} = \frac{1}{f_o} \quad (2.43)$$

Thus, Eq. (2.37) can be rewritten as

$$t(r_1) = p_o(r_1) \exp\left(\frac{ik}{2} \left(\frac{1}{d_0} + \frac{1}{d_1}\right) r_1^2\right) \quad (2.44)$$

By inserting the above equation into Eq. (2.42), the more explicit form of $\psi_2(r_2)$ is derived

$$\psi_2(r_2) = -\frac{1}{\lambda^2 d_0 d_1} \exp\left(-\frac{ik}{2d_1} r_2^2\right) \int d^2r_1 \ \int d^2r_0 \ \psi_0 \ \delta(r_0) p_o(r_1) \exp\left(-\frac{ik}{2d_0} r_0^2\right) \ \exp\left(\frac{ik}{2} \left(\frac{r_0}{d_0} + \frac{r_2}{d_1}\right) r_1\right) \quad (2.45)$$

Performing integral over $r_0$ via Dirac delta function (according to Eq. (2.39)), one gets

$$\psi_2(r_2) = -\frac{1}{\lambda^2 d_0 d_1} \psi_0 \exp\left(-\frac{ik}{2d_1} r_2^2\right) \int d^2r_1 \ p_o(r_1) \exp\left(\frac{ik}{d_1} r_1 r_2\right) \quad (2.46)$$

The quadratic phase term in $r_2$ may be left out, because there is no noticeable optical amplitude at distances exceeding several Rayleigh resolution lengths. Within this area, the considered
phase factor is of order of $10^{-3}$ that is much less than unity, hence may be neglected [3].

The wave field (without preceding factors) in the focal plane of the microscope is usually referred to as the probe function $P(r)$ as follows

$$P(r) = \int d^2r_1 p_o(r_1) \exp \left( \frac{i k}{d_1} r_1 r_2 \right)$$  \hspace{1cm} (2.47)

One can notice that the probe function is the Fourier transform of the objective pupil function given by Eq. (2.36)

$$P(r_2) \propto \mathcal{F}_{r_2} \{ p_o(\omega_1) \}$$  \hspace{1cm} (2.48)

with a frequency $\omega_1 = (k/d_1) r_1$.

Thus, by rewriting Eq. (2.46) one obtains

$$\psi_2(r_2) = -\frac{1}{\lambda^2 d_0 d_1} \psi_0 \int d^2r_1 p_o(r_1) \exp \left( \frac{i k}{d_1} r_1 r_2 \right) = -\frac{1}{\lambda^2 d_0 d_1} \psi_0 P(r_2)$$  \hspace{1cm} (2.49)

The interaction of the probe function with the specimen function (Eq. (2.8)) in the focal plane causes the wave field exiting the specimen to be expressed as follows

$$\psi'_2(r_2) = -\frac{1}{\lambda^2 d_0 d_1} \psi_0 O(r_2) P(r_2 - R_s)$$  \hspace{1cm} (2.50)

where $R_s$ is a scan displacement of the probe with respect to the specimen.

Since the sample-to-detector distance $d_2$ is a multiple of the FZP-to-sample distance $d_1$ and the far-field image of the specimen is considered, one should use the Fraunhofer approximation to find the wave field in the detector plane. Thereby:

$$\psi_3(r_3; R_s) = \frac{i}{\lambda d_2} \exp \left( -\frac{ik}{2d_2} r_3^2 \right) \int d^2r_2 \psi'_2(r_2) \exp \left( \frac{ik}{d_2} r_3 r_2 \right)$$  \hspace{1cm} (2.51)

As, eventually, one is interested in the intensity of the wave field in the detector plane, the quadratic phase term in $r_3$ preceding the integral may be disregarded.

Substituting Eq. (2.50), one obtains

$$\psi_3(r_3; R_s) = \frac{iv_0}{\lambda^3 d_0 d_1 d_2} \int d^2r_2 O(r_2) P(r_2 - R_s) \exp \left( \frac{ik}{d_2} r_3 r_2 \right)$$  \hspace{1cm} (2.52)

which is proportional to the forward Fourier transform of the wave field exiting the specimen

$$\Psi_3(\omega_3; R_s) \propto \mathcal{F}_{\omega_3} \{ O(r_2) P(r_2 - R_s) \}$$  \hspace{1cm} (2.53)

with frequency defined as follows $\omega_3 = \frac{k}{d_2} r_3$ [3].

The detector can measure only the intensity which is defined as a modulus squared of the wave field reaching the detector plane and thence also proportional to the modulus squared of the corresponding Fourier transform

$$I(\omega_3; R_s) = |\Psi_3(\omega_3; R_s)|^2 \propto |\mathcal{F}_{\omega_3} \{ O(r_2) P(r_2 - R_s) \} |^2$$  \hspace{1cm} (2.54)
With this in mind, one can apply the convolution theorem (Eq. (2.24)) to obtain the final result for the intensity in the detector plane

\[ I(\omega_3; R_s) \propto |O(\omega_3) \otimes \left( P(\omega_3) e^{i R_s \omega_3} \right) |^2 \]  

(2.55)

This means that the measured intensity is proportional to the convolution of the Fourier transformed object and probe functions [9]. A considerable number of imaging methods takes the advantage of this folding.

### 2.8 Phase contrast imaging

Using X-ray absorption relies on ray optics to describe and interpret image formation. This approach, however, ignores potentially more useful source of contrast: phase information. By contrast to absorption-based methods, the phase-sensitive techniques may be understood using wave optics rather than ray optics. In phase contrast imaging, small phase shifts in different parts of an X-ray beam passing through the sample are converted into contrast in the projected image [4]. One can argue that since the parameter \( \delta \) by which the refractive index \( n \) differs from unity has almost negligibly small value, the X-rays undergo only an extremely small angular deviation \( \alpha \) when refracted. Yet, it proves feasible to determine \( \alpha \) with considerably great accuracy using a variety of methods [4].

Let us consider for simplicity a pure phase object with no absorption towards the transmitted intensity. The object (specimen) function then has the form

\[ O(r) = \exp (i \delta(r) k \Delta z(r)) \equiv \exp (i \phi(r)) \]  

(2.56)

The phase contrast imaging, exploiting the refractive properties of materials, is based on the fact that the angular deviation \( \alpha \) is directly proportional to the gradient of the phase of the refracted beam. Concluding from Eq. (2.56), the phase of refracted beam is defined as

\[ \phi(r) = \delta(r) k \Delta z(r) = k' \cdot r \]  

(2.57)

where \( k' \) is the wave vector of the refracted beam. To justify this statement, let us focus on the direction of the refracted beam which is specified by the unit vector

\[ \hat{n} = \frac{k'}{|k'|} = \frac{\lambda}{2\pi} \nabla \phi(r) \]  

(2.58)

The angular deviation is a function of the coordinates \((x, y)\) in the plane perpendicular to the direction of the propagating incident beam. Therefore it is given by

\[ \alpha_x = \frac{\lambda}{2\pi} \frac{\partial \phi(x, y)}{\partial x} \quad \alpha_y = \frac{\lambda}{2\pi} \frac{\partial \phi(x, y)}{\partial y} \]  

(2.59)

Within this thesis the phase contrast imaging method utilizing propagation in free-space will be considered. Its basic experimental realization is depicted in Fig. 2.7. A finely focused
X-ray beam is incident on a sample with negligible absorption. As mentioned before, refraction causes the beam to be deflected by an angle of $\alpha$ (see Eq. (2.59)). The deflection of the refracted ray is recorded on a position sensitive area detector located a distance $L$ downstream of the sample and is equal to $\alpha L$. The blue circular spot corresponds to the direct beam if there was no sample on a scanning stage. Whereas the red spot indicates the deflected beam when the sample is at a fixed position $(x, y)$ relative to the focal point of the incident beam. This method relies on scanning the sample in the $(x, y)$ plane allowing a map of the phase gradient (deflection as a function of $x$ and $y$) to be created. From such collected dataset, an image of refractive index decrement $\delta(x, y)$ may be computed [2].

This shall serve as a theoretical background in calculation of the differential phase contrast signals presented in section 3.1.

### 2.9 Coherent X-ray diffractive imaging

It has been explained in former chapters how the diffraction pattern collected in the far-field regime is related to the Fourier transform of the complex X-ray optical transmission function of the specimen. Clearly, under normal circumstances it is not viable to recover an image of the electron density by performing the inverse Fourier transform of the diffraction pattern due to the phase problem. Yet, if a small but finite sized object is illuminated with coherent radiation, it turns out that the phases may be retrieved. A real space image of the sample can be then reconstructed by Fourier inversion. This type of imaging is called the coherent X-ray diffraction imaging (CXDI or CDI). Its most prominent feature is that it does not utilize lenses, becoming an example of a lensless imaging technique (see Fig. 2.8). This makes the final images free from aberration and resolution limited ultimately by the illumination wavelength. Another benefit is related to the fact that CDI has proven its capability of imaging both
crystalline and non-crystalline materials [2]. Concerning the resolution, CDI bridges its gap between the optical microscopy (hundreds of nanometres) and electron microscopy (nanometre or subnanometre). It should be also pointed out that the ultimate resolution of the obtained image improves linearly with the size $D$ of the detector (Fig. 2.8). Moreover, because of the considerable transparency of matter to X-rays, the internal structure of objects may be probed.

Fig. 2.8: The scheme of lensless imaging (CDI). To obtain the specimen image, the inverse Fourier transform of the far-field diffraction pattern recorded by detector is needed. From [4].

In order to obtain the conclusive results in coherent diffraction imaging, two conditions must be fulfilled: the sample must be entirely illuminated by the coherent part of the beam and the maximum path-length difference must be less than the longitudinal coherence length. In the case of the third generation synchrotron sources, the longitudinal coherence is limited by the monochromaticity of the beam, whereas the transverse coherence is determined by the source size and the distance of the source from the sample (see Eq. (2.3)). The degree of coherence may be improved by incorporating into experimental setup micronsized pinholes to be situated in the beam path and by making the source-to-sample distance as long as realizable.

As it was mentioned before, CDI is applicable to both noncrystalline and crystalline specimen. In the first case, one is interested in signal close to the forward scattering direction around the direct beam. In the second case, one can investigate the details of the scattering around the Bragg peaks associated with the crystal [4].

Concerning the ptychography method, introduced in section 2.10, the schematic of Fresnel coherent diffractive imaging experiment is presented in Fig. 2.9. The sample is illuminated with a nominally coherent beam of X-rays and the resulting diffraction pattern is captured on a suitable detector placed downstream of the sample. The first pinhole is used to select a coherent patch of the beam. A beam stop (central stop) is introduced to prevent the direct beam from the synchrotron damaging the detector. A zone plate is here used to create a diverging beam to be incident on the sample. Assembly of an order sorting aperture (OSA) allows to remove other diffracting orders from the zone plate and also, in conjunction with a beam stop (central
What should be emphasized is a fundamental drawback of diffraction-based imaging: the phase problem. The recorded real-valued data contains only amplitude information and the equally important knowledge of the phase values are lost [13].

One should therefore analyze how the coherent X-ray diffractive imaging solves the phase problem. It must be emphasized that the CDI relies foremost on the concept of oversampling of a diffraction pattern. The diffraction pattern from extended, three-dimensional crystal consists of a series of Dirac delta functions, $\delta(\omega - \Delta \omega)$, in reciprocal space. Hence, it can be sampled only when $\omega = \Delta \omega$. The frequency of the sampling in reciprocal space equals to $\Delta \omega = 2\pi/a$ for $a$ being equal to unit cell parameter [2]. However, from a signal processing point of view, sampling intensity only at the reciprocal lattice points undersamples the data by a factor of 2. For simplicity, the Young’s slits experiment is considered. The diffracted amplitude pattern is depicted in the top of Fig. 2.10.

If one samples at the peaks of intensity (shown as circles), then from a computerized representation of the data one would conclude that the diffraction pattern has a constant numerical intensity (represented by the horizontal dotted line) at points in the diffraction plane. In contrast, the Nyquist condition requires a function to be sampled at twice a frequency of the highest frequency component contributing to that function. In case of intensity (rather than amplitude), the Nyquist sampling is $\Delta \omega/2$, not $\Delta \omega$. This appropriate sampling of the intensity of the diffraction pattern is called oversampling [9]. For oversampling to be possible, the diffraction pattern itself must be extended in reciprocal space. This occurs for any finite-sized single objects, either non-crystalline or crystalline in nature. For instance, for a single molecule, there are no Bragg peaks and the diffraction pattern is a continuous function which thus can
be sampled at any $\omega$. For crystalline material oversampling is also feasible if the crystal itself is finite.

While oversampling enables an unambiguous solution for the phase problem, it does not suggest any practical method of finding it. For different fields of diffraction and microscopy, various solutions have been suggested to retrieve the phase information. The certain number of iterative phase retrieval methods involves algorithms which lead to unambiguous phase regain by the alternations between real and Fourier space with certain boundary constraints. These computer algorithms cleverly incorporate the oversampling as real-space constrains.

Thus far, in many numerical implementations of phase retrieval algorithms these constrains have been applied in different ways [2]. For one of such a brief overview is given in section 3.2 concerning the iterative phase retrieval in ptychography which derives from CDI.

2.10 Concepts of ptychography

Ptychographical coherent diffractive imaging (PCDI) is an experimental method originally developed in the 1970s for electron microscopy [7, 8]. Recently, it has been successfully applied also to X-rays. PCDI is also called ptychography, which origins from the Greek word "πτνξ" meaning "to fold". This refers to the overlap between diffraction orders via the convolution of the Fourier transform of a localized aperture or illumination function in the object plane [9]. Modern X-ray ptychography can be thought of as a combination of the coherent diffractive imaging (section 2.9) and scanning transmission X-ray microscopy (section 2.3). The crucial difference between STXM and the ptychography is that in the latter, the coherent X-ray radiation is used in such a way that the transmitted signal is spatially resolved using an area
detector, rather than recording just the integrated signal (as in STXM). Therefore, ptychography has been recently referred to as scanning X-ray diffraction microscopy (SXDM) [4].

In Fig. 2.11 the basic configuration of ptychography is shown. The beam which is incident on the sample is either so tightly focused that the beam divergence is larger than the angular separation of adjacent diffraction maxima, or is defined by a miconsized pinhole.

If a crystalline sample is considered, the diffraction pattern on an area detector contains diffraction discs as a result of the beam divergence, which partially overlap. In these regions of overlap, the diffraction maxima interfere with one another and the intensity depends on their relative phases.

In the case of noncrystalline samples, the same overlapping information is obtained by keeping the scanning shift of the sample between recording images less than the extend of the localized illumination [4]. Given this simple lateral shift of the illumination (or of the object), ptychography then allows a large number of interference patterns (as many as required) to be processed in order to obtain an image of a nonperiodic structure of unlimited size [9].

What should be emphasized, is that this "folding" of diffraction features contains the extra information allowing to extract the phases required for reconstructing the object. Obviously, there remains an ambiguity regarding the sign of the phase for any given diffraction pattern, but this is resolved by observing how the overlap intensity changes as one translates the sample [4]. In Fig. 2.12 an example of several illumination positions is depicted.

What is quite important, is that with the term overlap a linear distance offset is meant, not area overlap. For two circles of radius $r$ and the centre-to-centre distance $a \in [0, 2r]$ one
Fig. 2.12: Scanning electron micrograph of the test sample with gold nanostructures. The circles indicate positions for which diffraction patterns were recorded. Taken from [12].

defines the absolute overlap $o_{\text{abs}}$ as

$$o_{\text{abs}} = 2r - a$$

Normalized by the diameter of the circles the relative overlap $o$ is given by

$$o = 1 - \frac{a}{2r}$$ \hspace{1cm} (2.60)

It has been analyzed which value of the overlap of the different illumination positions is optimal. An instructive following example is taken from [13]. The simulated object shown in Fig. 2.13 was scanned with 5 x 5 positions of different overlaps.

Fig. 2.13: Sample image (a) and diffraction patterns (b-e) for the positions indicated in the image (a). Taken from [13].
The result is shown in Fig. 2.14. In Fig. 2.14(a) the relative overlap of the positions (see Eq. (2.60)) is only 10%. The reconstructed image hardly resembles the original one. When the relative overlap is increased to 60%, the retrieved image cannot be distinguished by eye from the original one (Fig. 2.13(a)). With 100% relative overlap the reconstruction algorithm degenerates to a conventional iterative phase retrieval, resulting in again poor image quality. Generally, it is advised in ptychographical experiments to keep the illuminated areas of the specimen overlap by around 70%.

Since ptychography derives from the coherent diffractive imaging, one should not forget about appropriate sampling approach. The ptychography technique relies on sampling between conventional diffraction orders. To fulfill this condition, the oversampling of diffracted intensity has to be utilized whose description has been presented in section 2.9. Oversampling the diffraction pattern of nonperiodic specimen with extension $D$ by a factor of $\sigma$ along a particular dimension requires a certain physical detector pixel width $\Delta x_2$ \[10\]. Thus, the oversampling ratio is defined as follows

$$\sigma = \frac{\lambda z}{\Delta x_2 D} \quad (2.61)$$

where $z$ is a sample-to-detector distance and $\lambda$ is radiation wavelength. This parameter is usually taken into account while designing the experimental setup.

In conventional STXM resolution is limited by the size of the focal spot. By contrast, in case of ptychography, the extra information obtained by using coherent radiation and an area detector means that the resolution is significantly improved. If one considers the square region

Fig. 2.14: Reconstruction for different relative overlaps. The relative overlap of the object position: (a) 10%, (b) 60%, (c) 100%. Taken from [13].
of interest of recorded diffraction pattern whose side consists of $N$ detector pixels, then the ultimate resolution of the ptychographical experiment is described by the pixel width $\Delta x_1$ in the object (sample) plane. It is determined by the parameters of experiment, which have been already mentioned, and given by the formula

$$\Delta x_1 = \frac{\lambda z}{N \Delta x_2}$$

(2.62)

As far as cited literature is concerned, the resolution of the final ptychographical image may be of tens of nanometres. What is more, because the sample is scanned, large fields of view can be obtained, even of order of several hundred square microns or more [4].

Once the ptychographical dataset is captured, the reconstruction method needs to be chosen to regain the sample image. The solution strategy is to iteratively reconstruct and refine a single projection of the sample which is consistent with all the recorded diffraction patterns [12]. Within the scope of this thesis, the extended Ptychographical Iterative Engine is presented as such iterative phase retrieval method (section 3.2).
3 Data analysis software overview

3.1 Differential Phase Contrast Analysis Tool

Phase contrast imaging methods are doubtless of significant interest in measurement of the phase structure of a specimen. Especially for investigation of the soft-matter samples, a quantitative evaluation of three- or two-dimensional map of refractive index (more precisely, the refractive index decrement $\delta$) is mostly desired. There are several different approaches, originating in phase contrast imaging, to obtain this information by converting phase differences into intensity contrast. An introduction to phase contrast imaging has been made in section 2.8. Within this section the technique of differential phase contrast will be presented including two approaches of its numerical implementation.

Firstly, we will focus more rigorously on the behaviour of the far-field intensity in the presence of a specimen according to the Fourier optics treatment. Combining Eqs (2.52) and (2.54), one can derive explicit formula for the intensity formed by the wave field in the detection plane of a scanning microscope

$$I_3(r_3; R_s) = |\psi_3(r_3; R_s)|^2 = \left| C \int d^2r_2 \, O(r_2) P(r_2 - R_s) \exp \left( i \frac{k}{d_2} r_3 r_2 \right) \right|^2$$

(3.1)

The detector is supposed to be located in the far-field, the specimen is described by the object function $O(r_2)$, the probe by $P(r_2 - R_s)$ and the scan displacement vector by $R_s$. The object function (Eq. (2.8)) can be rewritten as follows

$$O(r_2) = \exp (i \delta(r_2) k \Delta z(r_2) + \beta(r_2) k \Delta z(r_2)) \equiv \exp (\eta(r_2))$$

(3.2)

The terms in the exponential may be expanded in the Taylor series around the point illuminated by the probe (chosen to be 0). This yields

$$\eta(r_2) = \eta|_0 + \eta'|_0 \cdot r_2 + \mathcal{O}(r_2^2)$$

$$= i(\delta k \Delta z)|_0 + (\beta k \Delta z)|_0 \cdot r_2 + \nabla(\delta k \Delta z)|_0 \cdot r_2 + \nabla(\beta k \Delta z)|_0 \cdot r_2 + \mathcal{O}(r_2^2)$$

(3.3)

By inserting Eq. (3.3) into Eq. (3.1) and neglecting the second order terms, one gets

$$I_3(r_3; R_s) = \exp (2(\beta k \Delta z)_0) \left| C \int d^2r_2 \, P(r_2 - R_s) \exp \left( i \nabla(\delta k \Delta z)_0 \cdot r_2 + \nabla(\beta k \Delta z)_0 \cdot r_2 \right) \exp \left( i \frac{k}{d_2} r_3 r_2 \right) \right|^2$$

(3.4)

The factor which precedes the modulus squared denotes an absorption contrast. The gradient terms inside the integral represent differential phase contrast (DPC) and differential absorption contrast (DAC), respectively. It is often assumed, that DAC may be neglected as long as the
ratio $\delta/\beta$ is large and the probe small. Since for hard X-rays this ratio is in the range of 10 to 1000 (or even larger), the above equation can be rewritten as follows

$$I_3(r_3; R_s) = \exp (2(\beta k \Delta z)_0) \left| C \int d^2 r_2 \ P(r_2 - R_s) \exp \left( i \frac{k}{d_2} r_2 \left( r_3 + \frac{d_2}{k} \nabla (\delta k \Delta z) \right) \right) \right|^2 \quad (3.5)$$

Not taking into account the leading absorption contrast factor, it is visible that Eq. (3.5) is equivalent to the intensity in the absence of the specimen ($O(r) = 1$) and with a shift in the coordinate

$$r_3 \rightarrow r_3 + \frac{d_2}{k} \nabla (\delta k \Delta z)_0$$

Therefore, the resulting function can be identified as the non-specimen intensity shifted by an amount proportional to the phase gradient and attenuated through the specimen absorption.

$$I_3(r_3; R_s) = \exp \left( 2(\beta k \Delta z)_0 \right) I_3^{O(r)=1} \left( r_3 + \frac{d_2}{k} \nabla (\delta k \Delta z)_0 \right) \quad (3.6)$$

It should be pointed out, that the resulting angular deflection through the shift is in perfect agreement with angles given by Eq. (2.59).

One can conclude that through keeping the first order term of the Taylor expansion, the intensity gets shifted and therefore is not a symmetric function with respect to the origin any longer. This means that its centre of mass and centre of symmetry are shifted which is motivating conclusion in succeeding differential phase contrast definition. Within the scope of this thesis two approaches of calculating the DPC signals are implemented. The first utilizes the quadrant detector, the second applies the explicit formula for the diffraction pattern’s centre of mass. In both cases the single photon counting 2D detector is used.

### 3.1.1 Quadrant detector

As stated before, differential phase contrast signals are proportional to the angular deflections resulting from the phase gradient. In order to quantify arbitrarily these deflections of the intensity distribution, one can use a quadrant detector. A schematic of such detector, consisting of four square segments, is depicted in Fig. 3.1.

It is advised to symmetrically align the detector in the absence of a specimen with respect to the illumination. As it has been shown in Fig. 3.1 the arbitrary deflection $D$ may be decomposed into two orthogonal components $D_x$ and $D_y$ which relate to the intensity distribution shift in horizontal and vertical directions, respectively. Therefore, a complete definition of differential phase contrast signals for the each scan point are expressed as follows

$$DPC_x = \frac{I_L - I_R}{I_{total}} = \frac{(I_2 + I_3) - (I_1 + I_4)}{I_1 + I_2 + I_3 + I_4} \propto \frac{\partial \phi(x,y)}{\partial x} \quad (3.7)$$

$$DPC_y = \frac{I_T - I_B}{I_{total}} = \frac{(I_1 + I_2) - (I_3 + I_4)}{I_1 + I_2 + I_3 + I_4} \propto \frac{\partial \phi(x,y)}{\partial y} \quad (3.8)$$
Normalizing DPC signals by the total detected intensity \( I_{\text{total}} \) allows to remove the effect caused by source intensity variations and may refer to the local specimen absorption \[3\]. Although in a single photon counting 2D detector no such segments are defined, it is possible to create them numerically. The pixel which refers to the direct beam position is set to be the origin of the coordinate system. Then the size of segments is chosen, hence DPC signals from differing in size virtual quadrant detector may be calculated. An intensity from each segment \( I_i, i = 1, 2, 3, 4 \) is a sum over all pixels within a particular segment.

### 3.1.2 Determining the centre of mass

Another approach of evaluating differential phase contrast signals, which has been used in this thesis, is based on an explicit calculation of the diffraction pattern’s centre of mass. Let us consider \( N_{\text{PIE}} \) scans of the specimen. The subtraction of vectors \( \mathbf{r}_2 - \mathbf{R}_s^{(j)} \) denotes the position of the beam centre at the sample in the \( j \)-th scan, \( j = 1, \ldots, N_{\text{PIE}} \). Accordingly, the centre of mass of the diffracted intensity distribution is defined as follows

\[
CM_x(\mathbf{r}_2 - \mathbf{R}_s^{(j)}) = \frac{\sum_{r_3} x_3 I_j(r_3)}{\sum_{r_3} I_j(r_3)} \tag{3.9}
\]

\[
CM_y(\mathbf{r}_2 - \mathbf{R}_s^{(j)}) = \frac{\sum_{r_3} y_3 I_j(r_3)}{\sum_{r_3} I_j(r_3)} \tag{3.10}
\]

for \( r_3 = (x_3, y_3) \) being the real space coordinates of a detector pixel and \( I_j \) denoting the recorded intensity distribution for the \( j \)-th scan point \[10\]. If then the coordinates of the pixel at which the direct beam points equal \((x_3^{\text{ref}}, y_3^{\text{ref}})\), differential phase contrast signals are defined as written below:

\[
DPC_x = CM_x - x_3^{\text{ref}} \tag{3.11}
\]

\[
DPC_y = CM_y - y_3^{\text{ref}} \tag{3.12}
\]
Implementation of this approach in the case of a single photon counting 2D detector allows to use by default all the available pixels, however, specific masks can be applied to select desired regions of interest.

3.2 Ptychographical Iterative Engine

Iterative phase retrieval is one method for recovering a complex-valued reconstruction of a sample from its diffraction patterns. This is achieved by iteratively enforcing a set of constraints corresponding to the measurements taken and a priori knowledge of the system [14].

The crucial step for the application to non-crystalline samples was to draw from concepts of iterative phase retrieval for the analysis of a ptychographical dataset. Such implementation led to an algorithm that was named the Ptychographical Iterative Engine (PIE) [10]. However, a considerable drawback of the PIE is that it requires an accurate model of the localised wavefront which illuminates the target object. With this requirement the quality of the object function may never be better than the quality of the guessed illumination. Therefore, an extension of the original PIE has been developed to introduce the simultaneous reconstruction of both the object function and the complex illumination (the probe). In this scope, I shall refer to this extended Ptychographical Iterative Engine (ePIE) as a more efficient solution to the ptychographical problem bringing a major improvement in reconstruction process. For more detailed description, see [14].

The operation of the ePIE is depicted in Fig. 3.2. It is assumed that the interaction between the sample and the probe can be modelled by a complex multiplication. The wavefront exiting the sample is termed the exit wave and the further assumption is that its propagation to the plane of the detector can be modelled by a Fourier transform (high degree of coherence required from the probe at the plane of the sample). Therefore, the intensity of the wavefront incident at the detector is given by the formula

\[ I_j(u) = |\mathcal{F}[O(r)P(r - R_j)]|^2 \]  \hspace{1cm} (3.13)

where \(r\) and \(u\) are real-space and reciprocal-space coordinate vectors and \(O(r)\) and \(P(r)\) denote the object and the probe wavefronts, respectively. The vector \(R_j\) encodes the relative shift which is introduced between the object and probe before the intensity of the \(j\)th diffraction pattern is recorded.

Initial guesses \((j = 0), O_0(r)\) and \(P_0(r)\), are required to begin the algorithm. Generally, the initial object guess is taken as free-space and the initial probe wavefront is taken as a support function roughly the size of the intense region of the probe wavefront. The diffraction patterns are addressed in a random sequence \(s(j)\). Beginning with pattern \(s(0)\), a guess at the exit wave is formed by multiplying the current object guess by the appropriately shifted probe guess,
Previous guesses:
\[ P_j(r), O_j(r) \]

The exit-wave
\[ \psi_j(r) = O_j(r) \cdot P_j(r - R_s(j)) \]

Forward Fourier Transform
\[ \mathcal{F} \]

Replace *modulus* with measurement

Known intensity
\[ I_j(u) \]

Inverse Fourier Transform
\[ \mathcal{F}^{-1} \]

The updated exit-wave
\[ \psi'_j(r) \]

Update object guess using update function 1
\[ O_{j+1}(r) \]

Update object guess using update function 2
\[ P_{j+1}(r) \]

Fig. 3.2: Flowchart of the ePIE method.
which gives:

$$\psi_j(r) = O_j(r)P_j(r - R_{s(j)})$$

(3.14)

The next step is substituting the positive square-root of the $s(j)$th diffraction pattern recording for the modulus of the Fourier transform of this exit wave, so that

$$\Psi_j(u) = \sqrt{I_{s(j)}(u)} \frac{\mathcal{F}[\psi_j(r)]}{|\mathcal{F}[\psi_j(r)]|}$$

(3.15)

An updated exit wave is then calculated with a use of an inverse Fourier transform

$$\psi'_j(r) = \mathcal{F}^{-1}[\Psi_j(u)]$$

(3.16)

Finally, updated object and probe guesses are extracted from this result using two update functions. Eq. (3.17), update function 1, updates the current object guess and is given by

$$O_{j+1}(r) = O_j(r) + \alpha \frac{P'_j(r - R_{s(j)})}{\max(|P_j(r - R_{s(j)})|^2)} (\psi'_j(r) - \psi_j(r))$$

(3.17)

Whereas the following equation describe the update of the probe (update function 2)

$$P_{j+1}(r) = P_j(r) + \beta \frac{O'_j(r + R_{s(j)})}{\max(|O_j(r + R_{s(j)})|^2)} (\psi'_j(r) - \psi_j(r))$$

(3.18)

where $\alpha, \beta \in [0, 1]$ are constants adjusted to alter the step-size of the update.

The process presented above is continued with diffraction pattern $s(1), s(2), \ldots, s(J)$ until each of the $J$ diffraction patterns have been used to update the object and probe guesses. At this point a single ePIE iteration is completed. Depending on a chosen implementation, 20 to 30 iterations are needed to obtain final reconstruction image.
4 Measurement

The second part of this thesis is focused on experimental realization of the scanning transmission X-ray microscopy. The experiment was conducted at beamline P11 of PETRA III synchrotron light source at Deutsches Elektronen-Synchrotron in Hamburg, Germany. The images of two samples have been captured: a zone plate and a Siemens star.

4.1 Experimental setup

The schematic view of the STXM experiment is shown in Fig. 4.1. The well-defined and collimated undulator beam was used as a source of illumination. Its polarization was linear and in the horizontal plane. Details of the utilized setup are depicted in Fig. 4.2 and Fig. 4.3. The probe was formed using: beam defining pinhole (BDP) and a central stop (CS), mounted upstream of the Fresnel zone plate (FZP) and the order sorting aperture (OSA). The sample was mounted on a computer-controlled piezomotorized cartesian x/y stage. The PILATUS 6M detector was collecting the data situated in the far-field distance downstream of the sample stage.

To prevent diffracted X-rays from absorption in air on their quite long way to the detector, a flight tube was installed between the sample stage and the detector (Fig. 4.3). The flight tube was evacuated to reduce signal loss. Two Kapton windows were mounted at the flight tube’s ends because of their high X-ray transmittance and insensitivity to radiation damage.

It should be pointed out that this experimental setup is also applicable to the ptychographical coherent diffractive imaging, since the single photon counting area detector has been implemented. The PILATUS detector itself is a novel type of an X-ray detector, which has...
been developed at the Paul Scherrer Institut (PSI) for the Swiss Light Source (SLS). PILATUS detectors are two-dimensional hybrid pixel array detectors, which operate in single photon counting mode. A hybrid pixel, that features single photon counting, comprises a preamplifier, a comparator and a counter. The preamplifier amplifies the charge generated in the sensor by the incoming X-rays; the comparator produces a digital signal if the incoming charge exceeds a predefined threshold and thus, together with the counter, one obtains a complete digital storage and read-out of the number of detected X-rays per pixel without any read-out noise or dark current.

PILATUS detectors are characterized by several advantages compared to current state-of-the-art CCD and imaging plate detectors. The main features include: no readout noise, superior signal-to-noise ratio, read-out time of 5 ms, a dynamic range of 20bit, high detective quantum efficiency and the possibility to suppress fluorescence by an energy threshold that is set individually for each pixel. The short readout and fast framing time allow to take diffraction data in a continuous mode without opening and closing the shutter for each frame [15].
Fig. 4.4: Single view from all modules of PILATUS 6M detector (one projection from the scan of the sample zone plate). Decimal logarithm of the measured intensity is plotted.

In Tab. 4.1 the spatial characteristics of the PILATUS 6M detector have been summarized. An example of a single view from this detector, captured during the STXM experiment, is depicted in Fig. 4.4.

| PILATUS 6M                  |
|-----------------------------|
| **Module arrangement**      | 5 x 12                        |
| **Detector size**           | 424 x 435 mm²                 |
| **Format**                  | 2463 x 2527 pixels            |
| **Pixel size**              | 172 x 172 µm²                 |
| **Intermodule gap**         | x: 7 pixels                   |
|                             | y: 17 pixels                  |
|                             | 8.4% of total area            |

Tab. 4.1: Selected parameters of PILATUS 6M single photon counting 2D detector [15].
As it has been mentioned before, two samples, the zone plate and the Siemens star, have been scanned during two beamtimes. The Tab. 4.2 shows the most important parameters of the experiment in which the scan of the zone plate was performed. The diameter of the sample zone plate was equal to 150 µm, whereas its outermost zone width was 25 nm.

| Beam energy                         | 7.5 keV  |
|-------------------------------------|----------|
| Beam defining pinhole (diameter)    | 150 µm   |
| Central stop (diameter)             | 40 µm    |
| Focusing Fresnel Zone Plate         |          |
| diameter                            | 150 µm   |
| outermost zone width                | 25 nm    |
| focal length                        | 23.4 mm  |
| Order sorting aperture (diameter)   | 10 µm    |
| Sample-to-detector distance         | 636 cm   |

Tab. 4.2: The most important experiment parameters of the zone plate scan.

The second measurement was carried out to collect the STXM dataset for the Siemens star of diameter 20 µm. The Tab. 4.3 contains all important parameters determining geometry of that setup.

| Beam energy                         | 6.2 keV  |
|-------------------------------------|----------|
| Central stop (diameter)             | 50 µm    |
| Beam defining pinhole (diameter)    | 100 µm   |
| Focusing Fresnel Zone Plate         |          |
| diameter                            | 100 µm   |
| outermost zone width                | 25 nm    |
| focal length                        | 12.5 mm  |
| Order sorting aperture (diameter)   | 10 µm    |
| Sample-to-detector distance         | 395 cm   |

Tab. 4.3: The most important experiment parameters of the Siemens star scan.

In both tables, the optical instruments are ordered in such a way as they were mounted along the X-ray beam propagation direction.
4.2 Data analysis

In this section results of the differential phase contrast (DPC) analysis of the zone plate and the Siemens star scans will be presented. The scan of the zone plate consists of 21 x 21 projections (views) shifted by the step size equal to 100 nm. Hence, the field of view of a single final image covers 2.1 x 2.1 $\mu$m$^2$ area. The number of projections for the Siemens star scan is in turn equal to 80 x 80 with the step size of 250 nm which yields the field of view covering 20 x 20 $\mu$m$^2$.

The aim of this thesis was also to develop the DPC Analysis Tool. The program was implemented in C++ with support of the ROOT Data Analysis Framework [16]. It is an independent part of the software which is meant to provide tools for a comprehensive analysis of imaging dataset, i.e. detector raw data control, visualization of the probe positions in the sample plane, the DPC Analysis Tool and the ptychography reconstruction based on the ePIE algorithm. The ultimate and documented version will be available for the users of beamline during imaging experiments. Within this thesis the results obtained mostly with the DPC module are discussed.

As it has been mentioned in section 3.1, two algorithms of calculating the differential phase contrast signals were implemented: the quadrant detector DPC and the centre of mass DPC. In the first approach, one can take advantage of the tunable size of the virtual segments. Shrinking of the detector’s dimensions leads to substantial reduction of the computational time. But it could potentially cause data loss. The second approach, the centre of mass DPC, uses information from all valid pixels calculating centres of mass in $x$ and $y$ directions (Eqs (3.9), (3.10)).

Except for the differential phase contrast signals, the transmission intensity has been calculated as an indicator of transmitted photons which were collected on a 2D detector. It has been performed by summing the intensity values over all pixels of requested region of interest (ROI) and then dividing by the highest total intensity of given scan.

It is very important, when using detector such as PILATUS 6M, to control if detected intensity by a particular pixel has reasonable value. Hence, the detector bad pixels mask, specified for this model of detector, has been used to mask all unreliably behaving pixels. Nevertheless, it is advised to monitor measured intensity values, since even one wrong pixel value may cause the systematic error in data analysis. Therefore, intensity distributions have been plotted for single projections both for the zone plate and the Siemens star scans. Fig. 4.5 shows examples of such histograms for the zone plate scan (left) and the Siemens star scan (right). One can see that reasonable intensity values do not exceed the order of $10^6$, hence those above this threshold should be cut off. According to this result, in both DPC algorithms one extra condition was introduced that the intensity value was read in only when it was smaller than $10^6$. This analysis allows also to detect new badly behaving pixels which are not included in the detector bad pixels mask. In the case of both scans, there are 10 such new bad pixels which intensity value exceeds $10^9$ (Fig. 4.5).

Let us focus on the results obtained from the DPC Analysis Tool. Images in Fig. 4.7 to Fig. 4.9 concern the zone plate scan. Figures 4.7 and 4.8 depict the quadrant detector DPC
results for two different sizes of the virtual quadrant detector (ROI): 512 x 512 pixels and 2350 x 2350 pixels, respectively. In Fig. 4.9 one can see the output from the centre of mass DPC approach taken for all valid detector pixels. The same follows for the images of the Siemens star scan: Fig. 4.10 and Fig. 4.11 refer to the quadrant detector DPC approach for regions of interest: 512 x 512 pixels and 2350 x 2350 pixels, respectively, whereas Fig. 4.12 is the centre of mass DPC result for this scan.

The zone plate scan images depict several zones whose width may be estimated to around 0.4 \( \mu m \). The transmission intensity signal as well as the DPC\(_x\) clearly correspond to the zone plate’s structure, yet the DPC\(_y\) signal becomes more conclusive only in Fig. 4.9. Images of the Siemens star appear to be rather distorted and blurred on a micrometre level. This could have been caused by the motor instabilities. Images from the quadrant detector DPC analysis contain two pixels which appear in the same positions and have values that stand out from the others. This may be one cause of a lower contrast of these images. Yet, one can read from them the Siemens star’s diameter equal approximately to 20 \( \mu m \) which is consistent with its real dimension.

It is possible now to evaluate whether virtual shrinking of the detector’s dimensions in the quadrant detector DPC approach affects quality of images. From comparison of results in Fig. 4.7 to Fig. 4.8 (the zone plate scan) and Fig 4.10 to Fig. 4.11 (the Siemens star scan) one may conclude that choosing the smaller ROI does not cause any substantial loss of information. This may be explained by the fact that in this experiment the radiation scatters at narrow angles, therefore one needs not so large segments to regain the image of specimen.

One can compare quality of images from the centre of mass DPC approach, Fig. 4.9 (the zone plate scan) and Fig. 4.12 (the Siemens star), to the images obtained with the quadrant
detector DPC by taking the corresponding ROI of 2350 x 2350 pixels (neglecting pixels far from the centre does not matter due to weak scattering). It is possible to determine which approach reaches higher contrast. In order to quantify this, profiles along the same horizontal line for both DPC approaches have been compared for scan of the zone plate and the Siemens star separately.

In Fig. 4.13 one can see results for the zone plate scan: comparisons of DPC<sub>x</sub> profiles (on left) and of DPC<sub>y</sub> profiles (on right). Profiles have been plotted for a line y = 0.9 µm. Differences between minima and maxima of the quadrant detector DPC (plotted in black) are smaller than those of the centre of mass DPC (plotted in red). The same follows for the Siemens star scan. Fig. 4.14 shows the comparison of respective profiles along the line y = 5.25 µm. Differences between maxima and minima in the case of DPC<sub>x</sub> profiles prove higher contrast achieved by the centre of mass DPC approach. Yet, the comparison of DPC<sub>y</sub> profiles does not show such tendency. This may be caused by the general lower quality of the Siemens scan star. A possible reason for this is that the central stop used in this scan had too large diameter comparing to parameters of the focusing Fresnel zone plate (see Tab. 4.3) and they were a little misaligned. This fact can be seen in the enlarged central part of one single view from detector (Fig. 4.6): the direct beam position (the only red pixels) is not exactly in the centre of the far field image of the focusing zone plate. Nevertheless, one can conclude that, generally, in comparison to the quadrant detector DPC results, the higher contrast is achieved for the centre of mass DPC approach.

![Enlarged central part of a single view from detector from the Siemens star scan.](image)

Fig. 4.6: Enlarged central part of a single view from detector from the Siemens star scan. Central stop and the focusing Fresnel zone plate are not precisely aligned, because the direct beam position (the only red pixels) is not exactly in the centre of the far field image of the focusing FZP.
Fig. 4.7: The quadrant detector DPC analysis of the zone plate scan. Number of positions: 21 x 21. The step size: 100 nm. ROI (size of the virtual quadrant detector): 512 x 512 pixels.
Fig. 4.8: The quadrant detector DPC analysis of the zone plate scan. Number of positions: 21 x 21. The step size: 100 nm. ROI (size of the virtual quadrant detector): 2350 x 2350 pixels.
Fig. 4.9: The centre of mass DPC analysis of the zone plate scan. Number of positions: 21 x 21. The step size: 100 nm. All valid pixels from detector are taken into account.
Fig. 4.10: The quadrant detector DPC analysis of the Siemens star (diameter: 20 μm). Number of positions: 80 x 80. The step size: 250 nm. ROI (size of the virtual quadrant detector): 512 x 512 pixels.
Fig. 4.11: The quadrant detector DPC analysis of the Siemens star (diameter: 20 µm). Number of positions: 80 x 80. The step size: 250 nm. ROI (size of the virtual quadrant detector): 2350 x 2350 pixels.
Fig. 4.12: The centre of mass DPC analysis of the Siemens star (diameter: 20 μm). Number of positions: 80 x 80. The step size: 250 nm. All valid pixels from detector are taken into account.
Fig. 4.13: Comparison of profiles from the quadrant detector DPC analysis (in black) and the centre of mass DPC analysis (in red). Profiles have been taken along the horizontal line $y = 0.9 \, \mu m$. Larger differences between minima and maxima in the centre of mass DPC approach account for the higher contrast obtained by this method in comparison to the quadrant DPC.

Fig. 4.14: Comparison of profiles from the quadrant detector DPC analysis (in black) and the centre of mass DPC analysis (in red). Profiles have been taken along the horizontal line $y = 5.25 \, \mu m$. In the case of DPC$_x$ profiles, a gain in contrast is achieved by the centre of mass DPC approach. Yet, a comparison of DPC$_y$ profiles does not confirm this. It may be caused by the general lower quality of the Siemens star scan.
5 Conclusions

The aim of this thesis was to investigate the differential phase contrast scanning X-ray microscopy and apply this technique experimentally with the single photon counting 2D detector.

In the first part of this thesis, the fundamental information about physics of X-rays has been given as well as their applications in imaging. Three techniques have been discussed: scanning transmission X-ray microscopy (STXM), coherent diffractive imaging (CDI) and ptychography, deriving both from CDI and STXM. The second part of the thesis has provided essential information about two reconstruction methods: the differential phase contrast analysis and the extended Ptychographical Iterative Engine.

The final part of the thesis has been focused on the experiment conducted at the beamline P11 of PETRA III synchrotron light source at Deutsches Elektronen-Synchrotron in Hamburg, Germany. Experimental setups for scans of two samples, the zone plate and the Siemens star, as well as the parameters of single photon counting detector PILATUS 6M have been described. The collected data has been analyzed using the Differential Phase Contrast Analysis Tool, the program written in C++ programming language with support of the ROOT Data Analysis Framework library. An emphasis has been laid on controlling values of measured intensity. Histories of intensity distributions have shown that reasonable pixel values is from 0 to $10^6$. By introducing such intensity threshold to the analysis software, several unreliable pixels have been discarded which were not included in a detector bad pixels mask.

The final images differ in contrast depending on the approach of calculating DPC signals. The comparison of images from the quadrant DPC approach prove that there is no substantial data loss while taking smaller region of interest due to the weak scattering of radiation.
References

[1] Eine neue Art von Strahlen, W. Röntgen, in Sitzungberichte der Physikalischmedizinischen Gesellschaft zu Würzburg, 1895; Eine neue Art von Strahlen (in English), W. Röntgen, Nature 53 (1896) 274; Eine neue Art von Strahlen (in English), W. Röntgen, Science 3 (1896) 3.

[2] Elements of Modern X-ray Physics, Second Edition, J. Als-Nielsen, D. McMorrow, Wiley, 2011.

[3] Hard X-ray Phase Contrast Microscopy - Techniques and Applications, Ch. Holzner, Ph.D. Thesis, Stony Brook University, 2010.

[4] An Introduction to Synchrotron Radiation, Techniques and Applications, Ph. Willmott, Wiley, 2011.

[5] Introduction To Fourier Optics, J.W. Goodman, Roberts and Company Publishers, 2005.

[6] Soft X-rays and Extreme Ultraviolet Radiation, D. Attwood, Cambridge University Press, 2000.

[7] Beugung im inhomogenen Primärstrahlwellenfeld, III. Amplituden- und Phasenbestimmung bei unperiodischen Objekten, W. Hoppe, Acta Cryst. A 25 (1969) 508-514.

[8] Dynamische Theorie der Kristallstrukturanalyse durch Elektronenbeugung im inhomogenen Primärstrahlwellenfeld, R. Hegerl, W. Hoppe, Ber. Bunsen-Ges. 74 (1970) 1148-1154.

[9] Ptychography and Related Diffractive Imaging Methods, J.M. Rodenburg, Advances in Imaging and Electron Physics, Vol. 150, 2008.

[10] A study on new approaches in coherent x-ray microscopy of biological specimens, K. Giewekemeyer, Ph.D. thesis, Göttingen University, 2011.

[11] Imaging cellular architecture with X-rays, C. A. Larabell, K. A. Nugent, Current Opinion in Structural Biology (2010) 20(5) 623–631.

[12] Ptychography and lensless X-ray imaging, M. Dierolf et al., Nature 467 (2010) 436.

[13] Influence of the overlap parameter on the convergence of the ptychographical iterative engine, Oliver Bunk et al., Ultramicroscopy 108 (2008) 481–487.

[14] An improved ptychographical phase retrieval algorithm for diffractive imaging, A. M. Maiden, J. M. Rodenburg, Ultramicroscopy 109 (2009) 1256.

[15] PILATUS Project website, SLS Detector Group, http://pilatus.web.psi.ch/pilatus.htm

[16] The ROOT Data Analysis Framework website, http://root.cern.ch/drupal/