



Deep Learning for Bragg Coherent Diffraction Imaging: Detector Gap Inpainting and Phase Retrieval

Thesis

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0.1 Preface

In this manuscript, the use of Deep Learning methods, and more generally of GPU accelerated optimizations, for the advance of data analysis in Bragg Coherent Diffraction Imaging (BCDI) will be presented. However, before delving into the study's developments, I would like to share with the reader a reflection that has taken shape over the course of this PhD, serving as a kind of preface. In particular, I have come to observe that, unlike other more fundamental scientific investigations, this work originates from the practical limitations of the technique in question. It is indeed because the detectors are unable to record flawless images due to gaps, or incapable of capturing phase information because its oscillations are too rapid—that one is compelled to manipulate the available data with sophisticated algorithms. And, as often happens in science, compensating for these technical shortcomings leads to the development of tools rooted in the most abstract realms of mathematics and information theory. How much information can one extract from a signal? How can it be extracted, and under what conditions? In which circumstances is it easier, and why? Thus, a fascinating world opens up not when we directly investigate the foundations of matter, but when we examine *how* we go about investigating them.

Although this manuscript is ultimately focused on the specific cases of BCDI gap inpainting and phase retrieval, I hope to convey at least a bit of the wonder and awe that comes from knowing that such applications draw their roots from far deeper, more general, complex, and abstract themes.

0.1.1 Context, PhD objectives and manuscript outline

Over the past two decades the field of Coherent X-ray Diffraction Imaging (CXDI or CDI) has combined the study of microscopic structures enabled by X-rays with the imaging world. This powerful connection has been enabled by the availability of high-brilliance and coherent X-ray sources and the developments of computer algorithms for Phase Retrieval (PR) [Luke2004, 1, 2]. In fact, unlike conventional imaging methods, instead of relying on high-quality optics to form an image, CDI records only the intensity of the diffracted waves on a detector and then reconstructs the object by iterative algorithms. This approach removes the limitations imposed by imperfect lenses, enabling spatial resolutions set primarily by the wavelength and the numerical aperture of the scattering geometry. Since the first work by Miao [3] the CDI technique has been widely employed in material science for characterization at nanometric scale of different functional materials. [4–7]

When performed in Bragg geometry (Bragg CDI or BCDI), the technique enables precise 3D visualization and investigation of the internal atomic structure of single-crystal nanoparticles. Since its first demonstration in 2001 by Robinson et al. [8] for imaging gold nanoparticles, BCDI has proven to be a powerful tool for studying strain distributions [9–11], defect populations [12–14], and particle morphologies under different physico-chemical conditions [15–18]. Over the years, it has been applied to a broad range of technologically relevant systems, including nanotechnology and electronics [12], Li-ion and Na-ion batteries [19, 20], catalysis [21], and, more recently, biological materials [22].

In typical experiments, the recorded diffraction patterns are processed by PR softwares

[23] returning 3D complex arrays, the modulus of which represents the electron density of the sample, while the phase is associated to a component of the strain distribution inside the sample. Unlike other X-ray diffraction techniques, BCDI enables the study of isolated single particles, down to a few tens of nanometers in size [24, 25], with spatial resolution on the electron density of the order of 10 nm [26] and sensitivity to strain in the order of few picometers [13].

BCDI experiments are carried out at beamline laboratories of synchrotron facilities or free electron lasers (FELs) capable of delivering intense X-ray beams with exceptional coherence properties. Some examples are the ID01 beamline at the European Synchrotron Radiation Facility (ESRF) [27], the 34ID beamline at Advance Photon Source (APS) [28], the P10 beamline at PETRA III, NanoMax at MAX IV [24] or the CARNAÚBA beamline at SIRIUS [29].

Chapter 1 introduces the fundamental theoretical background of this technique and describes the practical conditions for a BCDI experiment at ID01.

The successful application of BCDI critically depends on computational algorithms that transform measured diffraction patterns into real-space reconstructions of particle shape and strain fields. From the early developments, significant efforts have been devoted to improving the speed, robustness and reliability of these algorithms. This field has gained further momentum with the advent of machine learning (ML). In parallel, the recent upgrade of numerous synchrotron facilities to fourth-generation light sources — including MAX IV (2017), ESRF-EBS (2020), Sirius (2020) and APS (2025) — has dramatically increased coherent flux, boosting the potential of crystalline nano-imaging techniques such as BCDI and Bragg ptychography [27, 30–32]. These advances also pose new challenges, particularly the need for faster and more efficient data processing pipelines capable of handling the rapidly growing volume of experimental data.

This PhD project was conceived in this context, with the goal of exploring how ML algorithms can address specific computational challenges in BCDI. **Chapter 2** provides the necessary theoretical background on conventional phase retrieval algorithms and **Chapter 3** introduces key ML concepts tailored to BCDI data analysis conducted throughout the PhD. Namely, two central problems are addressed in this work.

First, due to manufacturing constraints, detectors often contain grid-like regions that result in missing intensity data within the diffraction patterns. **Chapter 4** discusses how convolutional neural networks (CNNs) can be employed to restore these missing data, thereby improving the reliability of the reconstructed images.

Second, X-ray detectors cannot directly measure the phase of the scattered wavefield, making its retrieval an inherently challenging computational task. **Chapters 5 and 6** present how ML-based approaches developed during this PhD can assist or complement conventional phase retrieval methods.

CHAPTER 1

BRAGG COHERENT DIFFRACTION IMAGING

In this chapter some basic theoretical insights about the BCDI technique are provided, with the aim to highlight the key concepts, assumptions and physical interpretations. More thorough descriptions can be found in papers, textbooks and PhD manuscripts. I will adopt the formalism of Als-Nielsen and McMorrow in [33] but similar derivations and complementing observations can be found in [34, 35] as well as some more recent papers [36] and PhD thesis [37–39].

1.1 Foreword on typical assumptions and approximations in BCDI

In order to keep the dissertation short and targeted to the BCDI case, I will start considering some observations on this technique that will lead to some preliminary assumptions and simplifications. First, the word “Bragg” suggests that crystalline specimens are involved. As discussed later in the text, Bragg’s law applies to periodic structures, therefore we will limit our discussion to this specific case.

The word “Coherent” implies that samples are probed with coherent beams (in our case X-rays). This fundamental property of electromagnetic radiation will be briefly discussed later on. For the moment, this ingredient enables us to approximate the probing radiation with plane electromagnetic waves.

The word “Diffraction” refers to the type of mechanism describing the interaction between the X-rays and the samples. Paraphrasing [34] at page 4, this mechanism can be divided into two main phenomena, namely (i) the scattering of the radiation by each individual atom in the sample and (ii) the interference between the waves scattered by these atoms. The interference mechanism, in turn, is enabled because these scattered waves are coherent with the incident radiation and therefore between themselves. In other words, the information of each scatterer is shared with the other scatterers as the diffracted waves “talk to each other”. The complete mathematical description of these two phenomena without approximations is prohibitive, hence some simplifications usually adopted:

- **No refraction, no absorption:** Scattering is the only mechanism considered. Because of their short wavelengths (0.5 - 2.5 Å), X-rays are practically never deviated by refraction. Moreover, we assume the absence of any absorption effect. While inherently present, it

can be neglected from small enough samples.

- **Elastic scattering:** The interaction between the incoming X-rays and the atom is considered only elastic, meaning that no energy is transferred to the atom, which bounces off instead the photons with unaltered energy and momentum. This is again necessary for the scattered waves to interfere, as any difference in wavelength would prevent any coherent interaction. This description, also called Thomson scattering, considers the interaction with a free charge, and it also shows that cross-section of the scattering of electrons is much higher than the one of protons ($\sim 3 \times 10^6$ larger). For this reason, only the scattering from electrons is considered.
- **Weak diffraction (Born approximation):** This assumption implies that each scattered wave does not interact further with the sample, therefore neglecting any possible multiple scattering event. The consequence of this assumption is that the overall diffracted wave can be approximated by the linear superposition of the contributions of each scattering site. This approximation, in crystallography, is called *kinematical approximation*. Dealing with crystalline samples, this assumption breaks for relatively thick samples ($> 1\mu\text{m}$) in which the light travels through the sample for longer distances before exiting, therefore bouncing off several atoms. Diffraction of larger samples requires more complex theory of the so-called *dynamical regime* [40–42]. However, in our case, the size of the typical samples studied with BCDI hardly exceeds $1\mu\text{m}$ size, making the kinematical approximation suitable.
- **Far-field approximation:** Here, the distance between the scattering atoms and the detector is assumed to be much larger than the distance among the scatterers themselves. One can intuitively see that this approximation turns the spherical waves created by the scatterers, interfering with each other, into plane waves when these are evaluated far from the sources (in this case the atoms). This assumption is always respected in the BCDI technique as the sample-detector distance is in the order of tens of centimeters (practically from 30 cm to 2 m).

The last word “*Imaging*” tells us that the format of the data is by nature, multidimensional (2D - 3D). It will be shown later in the chapter that 3D diffracted signal is recorded stacking 2D images captured by the detector, and therefore the results after the data analysis are 3D images of the sample.

Given this set of assumptions and approximations we can proceed with our simplified derivation of the equation governing the coherent X-ray scattering from a crystal and its interpretation.

1.2 Coherent X-ray scattering from crystalline structures

Let us consider an X-ray beam, represented by a perfectly monochromatic plane wave with linear polarization in the horizontal plane, scattering with a single free electron (see Fig. 1.1). In this simple case we can imagine the X-ray electromagnetic field exerting a force onto the electron placed at the origin of the reference frame. In turn, this force will accelerate the electron accordingly, therefore inducing an oscillation motion generating an electromagnetic wave. Being the scattering assumed to be elastic, the radiation produced by the oscillating

electron (*electric dipole approximation*) will have the same wave-vector as the incoming X-ray. Moreover, the solution of Maxwell equations for this specific case shows that this dipole radiation propagates in the form of a spherical wave. At this point we ask ourselves what is the amplitude of this scattered wave when evaluated in a generic point \mathbf{r} on the vertical plane, far from the origin (*far-field approximation*). The result was achieved by Thomson in 1906 and is here reported without the full detailed derivation which can be found in the cited textbooks [33, 34, 43].

$$\mathbf{E}_{\text{dip}}(\mathbf{r}, t) = -r_0 \frac{e^{ikr}}{r} e^{-i\omega t} E_0 \hat{\mathbf{z}} \quad (1.1)$$

where r_0 is the classical radius of the electron, or Thomson scattering length, k is the outgoing wave-vector, ω is the pulsation of the X-ray beam (incoming and outgoing), E_0 is the electric field of the incoming radiation.

In this case we cannot talk about diffraction as there is no interference of the outgoing wave with other scattered waves. In order to have a diffraction pattern we need to have at least a second charge scattering, from which a phase delay with respect to the first one can be calculated. For instance, if we consider N electrons being illuminated by the same radiation, each of them placed in \mathbf{r}'_n we could evaluate the contribution to the overall scattering wave-field for each electron. The simplest way is to make use of the *kinematical approximation* and sum linearly all the contributions. However, we must take into account the phase delays between the scattering from different positions in space. This phase delay can be calculated, and it turns out to be $\Delta\phi(r) = (\mathbf{k} - \mathbf{k}_0) \cdot \mathbf{r}' = \mathbf{Q} \cdot \mathbf{r}'$ where we have expressed the difference between the wave vectors with \mathbf{Q} often called *scattering vector*. The equation can thus be rewritten like:

$$\mathbf{E}_{\text{atom}}(\mathbf{r}, t) = r_e \frac{e^{ikr}}{r} e^{-i\omega t} E_0 \hat{\mathbf{z}} \sum_{n=1}^N e^{i\mathbf{Q} \cdot \mathbf{r}'_n} \quad (1.2)$$

In the continuum limit, replacing the N point-like charges with an overall electron density distribution $\rho(\mathbf{r})$ the above equation takes the form:

$$\mathbf{E}_{\text{atom}}(\mathbf{r}, t) = r_e \frac{e^{ikr}}{r} e^{-i\omega t} E_0 \hat{\mathbf{z}} \int_{\mathbb{R}^3} \rho_a(\mathbf{r}') e^{i\mathbf{Q} \cdot \mathbf{r}'} d^3 \mathbf{r}' \quad (1.3)$$

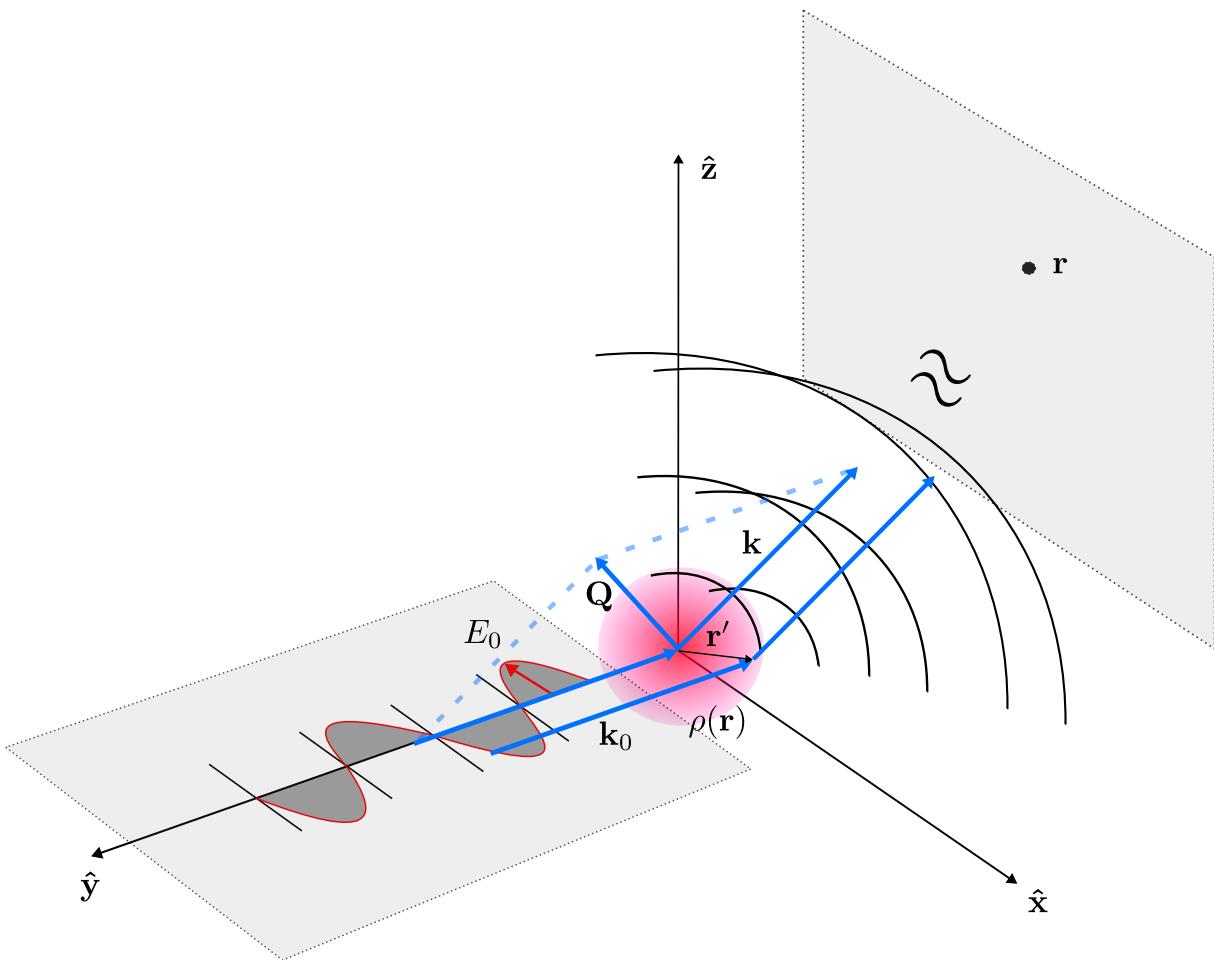


Figure 1.1: Sketch of the scattering process evaluated in the far-field on the vertical plane for an electron density irradiated by a monochromatic X-ray beam with linear polarization along the \hat{x} direction.

It is clear now that the information regarding the physical system of interest is embedded in the integral term. In fact, this is often called “*form factor*” - $F(\mathbf{Q})$ - and it plays an important role in the interpretation of the scattering equations.

$$F(\mathbf{Q}) = \int_{\mathbb{R}^3} \rho(\mathbf{r}') e^{i\mathbf{Q} \cdot \mathbf{r}'} d^3 \mathbf{r}' \quad (1.4)$$

$F(\mathbf{Q})$ represents the Fourier transform of the electron density, and it is the main result of this paragraph as it links the charge distribution of the sample in real space with the quantity measured, in reciprocal space.

To continue, we should bear in mind that X-ray photon counting detectors are sensitive to the time-averaged intensity of the signal as their time response is much slower than the oscillating frequency of X-rays ($\sim 10^9$ Hz for typical read-out limited frame rates of the Maxipix [44] against the $\sim 10^{18}$ Hz for X-rays at 10 keV). This limitation is also at the core of the “Phase Problem” that we will see later on, for which the phase information of the complex-valued wave-field is lost in the measurement. In order to do so, the time-averaged Poynting vector is calculated.

$$\langle \mathbf{S}(\mathbf{r}) \rangle = r_e^2 \frac{1}{r^2} J_0 \left| \int \rho(\mathbf{r}') e^{i\mathbf{Q} \cdot \mathbf{r}'} d^3 r' \right|^2 \hat{\mathbf{r}} \quad (1.5)$$

where $J_0 = |\mathbf{E}_0|^2 / 2\mu_0 c$ is the incident intensity. To conclude we consider the power delivered on the detector. For a pixel with area $d\mathbf{a} = r^2 d\Omega \hat{\mathbf{r}}$ the radiation power is equal to:

$$P(\mathbf{Q}) = r_e^2 J_0 \left| \int \rho(\mathbf{r}') e^{i\mathbf{Q} \cdot \mathbf{r}'} d^3 r' \right|^2 d\Omega \quad (1.6)$$

Eq.1.6 shows that the signal captured by the detectors is now in \mathbf{Q} space, and it is proportional to the square modulus of the Fourier transform of the electron density of the sample. The square modulus operation also shows how the phase of the Fourier transformed scattering complex-amplitude is lost. For this reason, in order to reconstruct the electron density, it is not possible to simply use an inverse Fourier transform, but iterative methods are required.

1.2.1 One atom

If now we were to consider an atom, far from resonance, we could assume the electron density being the main responsible for the scattering. It is known indeed that protons, because of the larger mass, have a much smaller cross-section for the scattering with photons. Using Eq.1.4 we would therefore have the *atomic form factor* - $f_l(\mathbf{Q})$ being defined as:

$$f_l(\mathbf{Q}) = \int_{\mathbb{R}^3} \rho_l(\mathbf{r}) e^{i\mathbf{Q} \cdot \mathbf{r}} d^3 \mathbf{r} \quad (1.7)$$

We now need to study the specific case in which the collection of atoms is ordered into a periodic structure.

1.2.2 Ensemble of ordered atoms: a Crystal

Perfect crystals are constructed by a basic structural arrangement of atoms (*motif*) repeated periodically on a *lattice* of one or more dimensions. The regularity of the lattice is such that, for the 3D case, any of its nodes can be located in space by the formula:

$$\mathbf{R}_n = n_1 \mathbf{a}_1 + n_2 \mathbf{a}_2 + n_3 \mathbf{a}_3 \quad (1.8)$$

where $\mathbf{a}_1, \mathbf{a}_2, \mathbf{a}_3$ constitutes the basis vectors of the primitive unit cell and n_1, n_2, n_3 are integer numbers. It follows that the information can be condensed in the unit cell, i.e. the orientation in space of the basis vectors as any region of the lattice can be seen as the same unit cell, translated from the origin by the amount given by \mathbf{R}_n .

The overall crystal is then constructed positioning on each of the nodes of the lattice the same motif, or basis.

In another more elegant way, we could say that, being the lattice $\mathcal{L}(\mathbf{r})$, the basis $\mathcal{B}(\mathbf{r})$, the crystal $\mathcal{C}(\mathbf{r})$ is given by the convolution of $\mathcal{L}(\mathbf{r})$ with $\mathcal{B}(\mathbf{r})$:

$$\mathcal{C}(\mathbf{r}) = \mathcal{L}(\mathbf{r}) * \mathcal{B}(\mathbf{r}) \quad (1.9)$$

For simplicity, we will consider from now on a single atom basis. At this point, when evaluating the scattering amplitude of the crystal we have to deal to an assembly of atoms, and we may want to exploit the regular structure we have just described. First, we can assume that, similarly to the case of many scattering electrons, in the kinematical approximation the overall scattering factor is given by the sum of the contributions of each atom, weighted by a phase factor that accounts for their positions in space.

$$F_{\text{crystal}}(\mathbf{Q}) = \sum_{l=1}^{\text{All atoms}} f_l(\mathbf{Q}) e^{i\mathbf{Q} \cdot \mathbf{r}_l} \quad (1.10)$$

Secondly, observing that the position of each atom is given by the sum of the position of the atom inside the unit cell and the lattice vector $\mathbf{r}_l = \mathbf{R}_n + \mathbf{r}_j$, we can separate Eq.1.10 in two terms:

$$F_{\text{crystal}}(\mathbf{Q}) = \sum_{\mathbf{R}_n + \mathbf{r}_j}^{\text{All atoms}} f_l(\mathbf{Q}) e^{i\mathbf{Q} \cdot (\mathbf{R}_n + \mathbf{r}_j)} = \underbrace{\sum_n e^{i\mathbf{Q} \cdot \mathbf{R}_n}}_{\text{Lattice}} \underbrace{\sum_j f_j(\mathbf{Q}) e^{i\mathbf{Q} \cdot \mathbf{r}_j}}_{\text{Unit cell}} \quad (1.11)$$

The first summation extends over all lattice points, while the second covers all atoms within the unit cell. Here we can already wrap the second sum into a more practical term expressing the *unit cell form factor*

$$F_{\text{crystal}}(\mathbf{Q}) = F_{\text{u.c.}}(\mathbf{Q}) \sum_n e^{i\mathbf{Q} \cdot \mathbf{R}_n} \quad (1.12)$$

The term $F_{\text{u.c.}}(\mathbf{Q})$ can be easily calculated as typical unit cells contain a small number of elements. On the contrary we need to exploit the properties of the lattice periodicity to evaluate the large summation over all lattice points.

1.2.3 Laue condition and Bragg's Law

The term we want to calculate is the sum of complex exponential, meaning that if the phases $\mathbf{Q} \cdot \mathbf{R}_n$ are misaligned the sum will be *incoherent* and the resultant will be very small, in the order of unity. On the contrary, when phase offsets are equal to an integer multiple of 2π the *phasors* will add *coherently*. The problem is thus to find those \mathbf{Q} values for which

$$\mathbf{Q} \cdot \mathbf{R}_n = 2\pi \times \text{integer} \quad \forall n \quad (1.13)$$

In order to do than we need to construct a reciprocal space lattice with a set of basis $\mathbf{a}_1^*, \mathbf{a}_2^*, \mathbf{a}_3^*$ which fulfill:

$$\mathbf{a}_1 \cdot \mathbf{a}_1^* = 2\pi h \quad \mathbf{a}_2 \cdot \mathbf{a}_2^* = 2\pi k \quad \mathbf{a}_3 \cdot \mathbf{a}_3^* = 2\pi l \quad (1.14)$$

where h, k, l known as Miller indices, are integer. Having a set of basis vectors and the Miller indices, the resulting reciprocal space lattice lies in those points found by the vector \mathbf{G}_{hkl}

$$\mathbf{G}_{hkl} = h\mathbf{a}_1^* + k\mathbf{a}_2^* + l\mathbf{a}_3^* \quad (1.15)$$

This result is telling us that the scattering amplitude of a diffracting crystal is detectable only in those points of in space for which the wave-vector \mathbf{Q} coincides with a point (hkl node) of the reciprocal lattice, hence $\mathbf{Q} = \mathbf{G}_{hkl}$. These isolated points are called *Bragg peaks*.

This is known as the Laue condition for diffraction as it was discovered by Max von Laue in 1912 [45].

A different but equivalent interpretation of the diffraction of a crystal was given by William Lawrence Bragg in 1913 [46]. Here, the crystal lattice is seen as a stack of parallel planes (see Fig.1.2) and the condition for constructive interference of the waves scattered by planes of the same family is found as follows. Let us consider an X-ray beam of wavelength λ and propagation vector \mathbf{k}_i impinging with an angle θ on a crystal. We call d_{hkl} the distance between the planes of the crystal, along a specific hkl direction. The scattered beam is leaving the crystal with the same angle θ and with a propagation vector \mathbf{k}_f equal in magnitude to the incident one (*elastic scattering*). At this point one can find the relationship between θ, d_{hkl}, λ that allows for a constructive interference of the waves diffracted from the series of planes by evaluating the optical path length difference induced by the spacing. Reminding that $|k_i| = |k_f| = 2\pi/\lambda$ and with the help of Fig. 1.2 we can observe that this difference is $\Delta l = 2d_{hkl} \sin(\theta)$ and therefore the phase offset between two waves is $\Delta\phi = |k| \Delta l = 4d_{hkl}\pi \sin(\theta)/\lambda$. We have seen above that the condition for constructive interference requires the phase differences to be equal to a multiple of 2π , therefore:

$$2d_{hkl} \sin(\theta) = n\lambda \quad \text{where } n \in \mathbb{Z} \quad (1.16)$$

Moreover, it can be shown that the family of planes is defined by the vector G_{hkl} which points at the reciprocal space node that is collecting the scattering from those planes, and defines as well the spacing d_{hkl} between each of these planes.

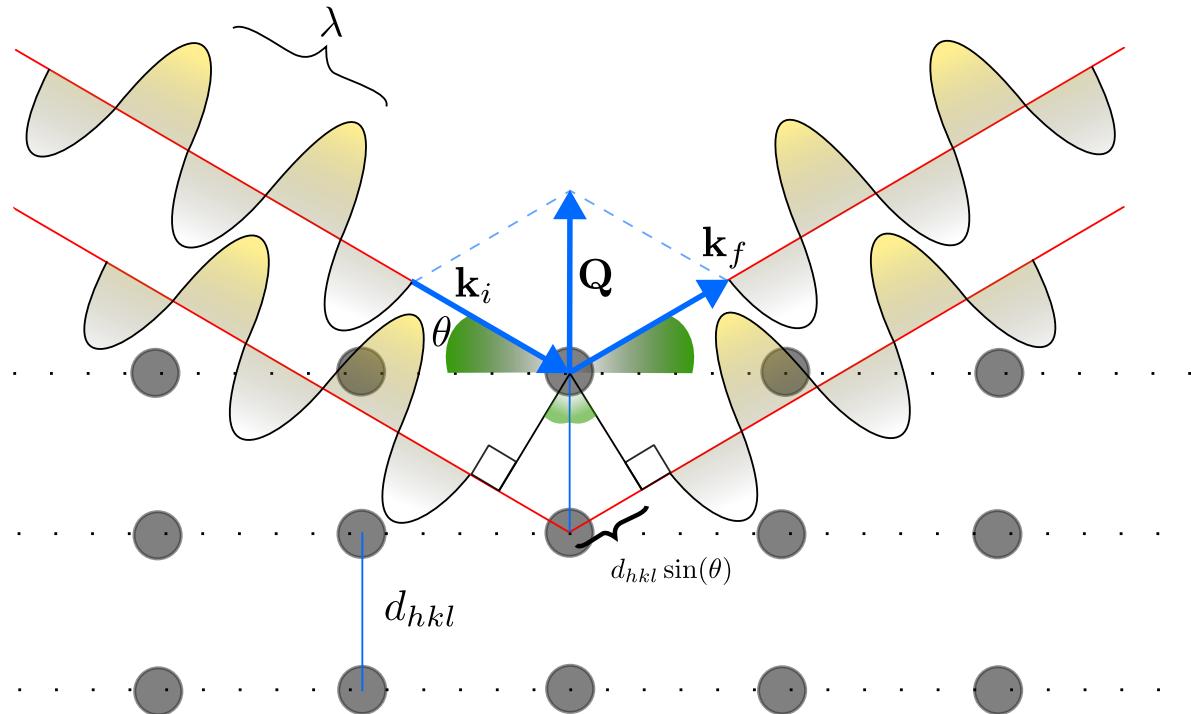


Figure 1.2: Illustration of Bragg's law. The family of crystal planes given separated by d_{hkl} produces constructive interference when the incident radiation illuminates it with an angle θ given by Eq.1.16.

Equation 1.16 is known as Bragg's law, and it can be derived from the Laue condition.

We can now rewrite Eq.1.12 for the case of an infinitely extended perfect 3D crystal as:

$$F_{\text{crystal}^\infty}(\mathbf{Q}) = F_{\text{u.c.}}(\mathbf{Q}) \sum_h \sum_k \sum_l \delta(\mathbf{Q} - \mathbf{G}_{hkl}) \quad (1.17)$$

where The term relative to the lattice is expressed as the sum of Dirac deltas in Q space centered in each hkl node.

1.2.4 Finite size crystals

We are now ready to treat the case of finite size crystals. In the above description the lattice was assumed to extend infinitely along all the 3 dimensions. This, we have seen, results in a scattering signal that lives on a perfect reciprocal lattice made of point-like nodes. Because of the Fourier transformation that links the real space scattering object and the reciprocal space diffracted signal, one can intuitively deduce that if for an infinite crystal we obtain a point-like Bragg peaks, we could expect a broadening of the peaks as the crystal size is reduced.

Mathematically we can derive the result for crystal of shape $S(\mathbf{r})$ considering the function S a window cropping a finite portion of the 3D infinite crystal. This function can also be referred to as Ewald function [47] or *support*. In real space this corresponds to the product:

$$\rho_{\text{fin}}(\mathbf{r}) = \rho_\infty(\mathbf{r})S(\mathbf{r}) \quad (1.18)$$

where

$$S(\mathbf{r}) = \begin{cases} 1, & \text{inside the crystal region,} \\ 0, & \text{otherwise.} \end{cases} \quad (1.19)$$

For the convolution theorem we have now that the Fourier transform $\mathcal{F}\{\rho_{\text{fin}}(\mathbf{r})\}$ is equal to the convolution $\mathcal{F}\{\rho_{\infty}(\mathbf{r})\} * \mathcal{F}\{S(\mathbf{r})\}$. Using Eq.1.12 for the Fourier transform of the infinite crystal and calling $\widehat{S}(\mathbf{Q}) = \mathcal{F}\{S(\mathbf{r})\}$ we can therefore write:

$$F_{\text{fin. crystal}}(\mathbf{Q}) = \left[F_{\text{u.c.}}(\mathbf{Q}) \sum_h \sum_k \sum_l \delta(\mathbf{Q} - \mathbf{G}_{hkl}) \right] * \widehat{S}(\mathbf{Q}) \quad (1.20)$$

For the commutative property of convolutions, we can now rearrange the terms as follows:

$$\begin{aligned} F_{\text{fin. crystal}}(\mathbf{Q}) &= \widehat{S}(\mathbf{Q}) * \left[F_{\text{u.c.}}(\mathbf{Q}) \sum_h \sum_k \sum_l \delta(\mathbf{Q} - \mathbf{G}_{hkl}) \right] \\ &= \int \widehat{S}(\mathbf{Q} - \mathbf{Q}') F_{\text{u.c.}}(\mathbf{Q}') \sum_h \sum_k \sum_l \delta(\mathbf{Q}' - \mathbf{G}_{hkl}) d^3 \mathbf{Q}' \end{aligned} \quad (1.21)$$

At this point, we can bring the integral inside the summation and calculate:

$$F_{\text{fin. crystal}}(\mathbf{Q}) = \sum_h \sum_k \sum_l \int \widehat{S}(\mathbf{Q} - \mathbf{Q}') F_{\text{u.c.}}(\mathbf{Q}') \delta(\mathbf{Q}' - \mathbf{G}_{hkl}) d^3 \mathbf{Q}' \quad (1.22)$$

We observe now that the Dirac delta function is non-zero only in the hkl nodes defined by \mathbf{G}_{hkl} . This leads to the final equation:

$$F_{\text{finite}}(\mathbf{Q}) = \sum_h \sum_k \sum_l \widehat{S}(\mathbf{Q} - \mathbf{G}_{hkl}) F_{\text{u.c.}}(\mathbf{G}_{hkl}) \quad (1.23)$$

Here we see that the Bragg peaks, from point-like spots in reciprocal space are subjected to a broadening that shapes them according to the structure of the Fourier transform of the crystal shape, centered at \mathbf{G}_{hkl} .

Let us now consider the specific case of a cubic crystal aligned with the direct-lattice basis $\mathbf{a}_1, \mathbf{a}_2, \mathbf{a}_3$. The crystal contains N cells along each basis vector \mathbf{a} . Each edge vector is therefore $\mathbf{L}_j = N\mathbf{a}_j$. Being the center of the cubic crystal placed in the origin of the reference frame we can now write the support function as:

$$S(\mathbf{r}) = \prod_{j=1}^3 \mathcal{R}\left(\frac{\mathbf{r} \cdot \hat{\mathbf{a}}_j}{L_j}\right), \quad \mathcal{R}(x) = \begin{cases} 1, & |x| \leq 1/2, \\ 0, & \text{else.} \end{cases} \quad (1.24)$$

where $\hat{\mathbf{a}}_j = \mathbf{a}_j/|\mathbf{a}_j|$ and $L_j = \mathbf{L}_j/|\mathbf{L}_j|$, and the 1D rectangular function $\mathcal{R}(x)$ has been defined. Here we have defined the cube as the product of the three 1D rectangular function

on each of the 3 dimensions. We can therefore calculate the Fourier transform of $S(\mathbf{r})$ as the product of the Fourier transform of each $\mathcal{R}\left(\frac{\mathbf{r} \cdot \hat{\mathbf{a}}_j}{L_j}\right)$.

It follows that:

$$\widehat{S}(\mathbf{Q}) = \prod_{j=1}^3 L_j \operatorname{sinc}\left(\frac{\mathbf{Q} \cdot \hat{\mathbf{a}}_j L_j}{2}\right), \quad (1.25)$$

where we made use of the cardinal sine function defined as $\operatorname{sinc}(x) = \frac{\sin x}{x}$. Putting this result in Eq. 1.23 we obtain:

$$F_{\text{cube}}(\mathbf{Q}) = \sum_h \sum_k \sum_l F_{\text{u.c.}}(\mathbf{G}_{hkl}) \prod_{j=1}^3 L_j \operatorname{sinc}\left(\frac{(\mathbf{Q} - \mathbf{G}_{hkl}) \cdot \hat{\mathbf{a}}_j L_j}{2}\right), \quad (1.26)$$

At this point of the derivation we can introduce another simplification, directly linked to the BCDI technique. Up to now we have been considering the scattering amplitude in the full reciprocal space. This means that Eq. 1.26 is encompassing all the hkl nodes in which we could find the diffracted signal. However, in typical BCDI experiments only a small region around a single Bragg peak is measured. This means that we can from now on focus on one \mathbf{G}_{hkl} vector and drop the summation. Moreover, defining $\mathbf{q} = \mathbf{Q} - \mathbf{G}_{hkl}$ and recalling the result obtained in Eq. 1.6 one can find that the intensity of the diffraction pattern of the hkl Bragg peak is :

$$I_{\text{cube}}(\mathbf{q}) \propto F_{\text{u.c.}}(\mathbf{G}_{hkl}) \left| \prod_{j=1}^3 L_j \operatorname{sinc}\left(\frac{\mathbf{q} \cdot \hat{\mathbf{a}}_j L_j}{2}\right) \right|^2, \quad (1.27)$$

The diffracted signal for a cubic crystal in the vicinity of the hkl Bragg peak has therefore the shape of a *squared* 3D cardinal sine function as represented in Fig. 1.3

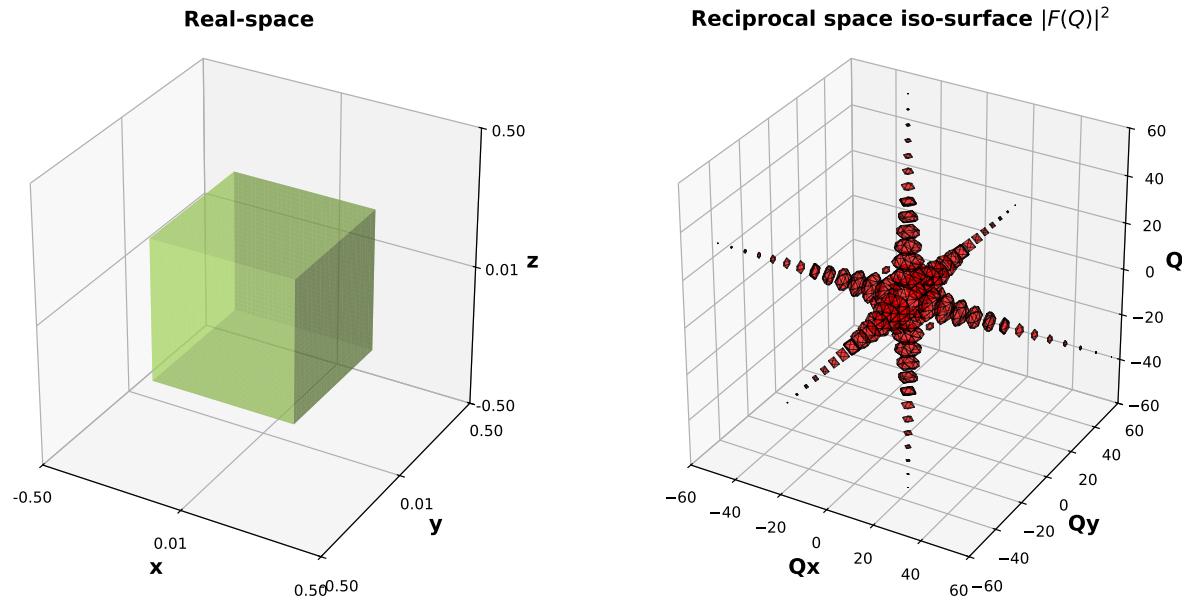


Figure 1.3: Illustration of the squared modulus of the Fourier transform of cube as representation of the diffracted intensity around a Bragg peak for a cubic crystal.

An interesting observation concerns the spacing between the fringes of the interference pattern. Studying Eq.1.27 we can infer that:

- The maximum of the peak is concentrated at $\mathbf{q} = 0 \Rightarrow \mathbf{Q} = \mathbf{G}_{hkl}$ as expected.
- The intensity decays as $\sim 1/q^2$
- The intensity drops to zero every time the argument $(\mathbf{q} \cdot \hat{\mathbf{a}}_j L)/2 = n\pi$ with n integer. This results in the presence of fringes that have a thickness as a function of the \mathbf{q} direction given by: $\Delta q_j = 2\pi/L$.
- Given the above relationship between the thickness of the fringes and the size of the crystal, one can intuitively imagine extending L to infinity, thus narrowing the fringes to zero width and the central lobe to a Dirac delta in $\mathbf{q} = 0$ as in the infinite lattice case.

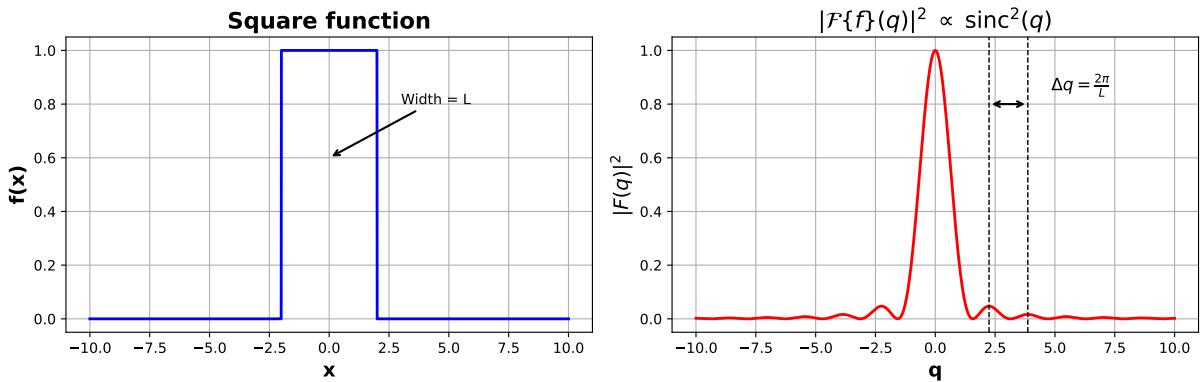


Figure 1.4: 1D example of the squared modulus of the Fourier transform of a square function. The sharp intensity decay outside the central peak is evident. The spacing between fringes of constructive interferences is also related to the width of the square.

For the understanding and visualization of Fourier transforms in crystallography I recommend the pedagogical work of Aubert and Lecompte [48].

1.2.5 Real crystals

We are now ready to deal with non-perfect crystals. In the most general case the deformation is modeled by a displacement field $\mathbf{u}(\mathbf{r})$ that shifts the position of each atom of the crystal off the position of the perfect lattice. However, for simplicity the displacement field is often assumed to act on the whole unit cell rather than each single atom. One can therefore write the scattering amplitude as the sum over the unit cells. Defining $\mathbf{u}(\mathbf{R}_n)$ the displacement of the $n - th$ cell with respect to the perfect lattice position we can calculate the structure factor of the infinite crystal summing over all the unit cells the form factor of each cell weighted by their displaced positions:

$$F_\infty^{\text{strain}}(\mathbf{Q}) = \sum_n F_n(\mathbf{Q}) e^{i\mathbf{Q}\cdot(\mathbf{R}_n + \mathbf{u}(\mathbf{R}_n))} \quad (1.28)$$

To lighten up the notation we can introduce another vector $\mathbf{R}'_n = \mathbf{R}_n + \mathbf{u}(\mathbf{R}_n)$ that points at each displaced unit cell. Being this lattice no longer perfectly periodic we cannot any more simplify the sum into a sum of delta functions calculated in the hkl nodes of the reciprocal lattice. At this point we can however repeat the calculations for the case of a finite size crystal of shape $S(\mathbf{r})$. For the convolution theorem we can write:

$$F_{\text{fin}}^{\text{strain}}(\mathbf{Q}) = \sum_n F_n(\mathbf{Q}) e^{i\mathbf{Q}\cdot\mathbf{R}'_n} * \widehat{S}(\mathbf{Q}) \quad (1.29)$$

Developing the convolution integral we obtain:

$$\begin{aligned} F_{\text{fin}}^{\text{strain}}(\mathbf{Q}) &= \int \sum_n F_n(\mathbf{Q} - \mathbf{Q}') e^{i(\mathbf{Q}-\mathbf{Q}')\cdot\mathbf{R}'_n} \widehat{S}(\mathbf{Q}') d^3\mathbf{Q}' \\ &= \sum_n \int F_n(\mathbf{Q} - \mathbf{Q}') e^{i(\mathbf{Q}-\mathbf{Q}')\cdot\mathbf{R}'_n} \widehat{S}(\mathbf{Q}') d^3\mathbf{Q}' \end{aligned} \quad (1.30)$$

where, being the sum over a finite number of unit cells converging, we moved the integral sign inside the sum. Now, we can assume to restrict the field of view in the vicinity of the \mathbf{G}_{hkl} where the unit cell form factor varies slowly compared to the $\widehat{S}(\mathbf{Q}')$. Intuitively we could understand this step considering that typical BCDI crystals contain a large number of unit cells, therefore in Fourier space, the features relative to the crystal shape vary much faster than the features relative to the unit cell. This approximation allows us to write: Hence:

$$\begin{aligned}
F_{\text{fin}}^{\text{strain}}(\mathbf{Q}) &= \sum_n F_n(\mathbf{G}_{hkl}) \int e^{i(\mathbf{Q}-\mathbf{Q}') \cdot \mathbf{R}'_n} \widehat{S}(\mathbf{Q}') d^3 \mathbf{Q}' \\
&= \sum_n F_n(\mathbf{G}_{hkl}) \int e^{i\mathbf{Q} \cdot \mathbf{R}'_n} e^{-i\mathbf{Q}' \cdot \mathbf{R}'_n} \widehat{S}(\mathbf{Q}') d^3 \mathbf{Q}' \\
&= \sum_n F_n(\mathbf{G}_{hkl}) e^{i\mathbf{Q} \cdot \mathbf{R}'_n} \int e^{-i\mathbf{Q}' \cdot \mathbf{R}'_n} \widehat{S}(\mathbf{Q}') d^3 \mathbf{Q}' \\
&= \sum_n F_n(\mathbf{G}_{hkl}) e^{i\mathbf{Q} \cdot \mathbf{R}'_n} S(\mathbf{R}'_n)
\end{aligned} \tag{1.31}$$

where we have recognized the inverse Fourier transform of the shape function calculated in \mathbf{R}'_n . If we now assume that the displacement affecting each unit cell does not influence the form factor, we can replace $F_n(\mathbf{G}_{hkl})$ with $F_{\text{u.c.}}(\mathbf{G}_{hkl})$ and take it out of the sum. Additionally, considering the relations $\mathbf{Q} = \mathbf{G}_{hkl} + \mathbf{q}$ and $e^{i\mathbf{G}_{hkl} \cdot \mathbf{R}_n} = 1$ we can rearrange into:

$$\begin{aligned}
F_{\text{fin}}^{\text{strain}}(\mathbf{Q}) &= \sum_n F_n(\mathbf{G}_{hkl}) e^{i\mathbf{Q} \cdot \mathbf{R}'_n} S(\mathbf{R}'_n) \\
&= F_{\text{u.c.}}(\mathbf{G}_{hkl}) \sum_n e^{i(\mathbf{G}_{hkl} + \mathbf{q}) \cdot (\mathbf{R}_n + \mathbf{u}(\mathbf{R}_n))} S(\mathbf{R}'_n) \\
&= F_{\text{u.c.}}(\mathbf{G}_{hkl}) \sum_n e^{i\mathbf{G}_{hkl} \cdot \mathbf{R}_n} e^{i\mathbf{G}_{hkl} \cdot \mathbf{u}(\mathbf{R}_n)} e^{i\mathbf{q} \cdot (\mathbf{R}_n + \mathbf{u}(\mathbf{R}_n))} S(\mathbf{R}'_n) \\
&= F_{\text{u.c.}}(\mathbf{G}_{hkl}) \sum_n S(\mathbf{R}'_n) e^{i\mathbf{G}_{hkl} \cdot \mathbf{u}(\mathbf{R}_n)} e^{i\mathbf{q} \cdot \mathbf{R}'_n}
\end{aligned} \tag{1.32}$$

The formula we obtained tells us that scattering amplitude for the finite strained crystal is proportional to the sum over all the unit cells of the shape function evaluated in each displaced unit cell times a two phase factors. The first phase factor is given by the projection of the displacement of each unit cell (relative to the perfect lattice) onto the scattering vector of the given hkl node. This term is relevant in BCDI experiments as it is directly linked to the strain and more general information on the internal lattice displacements of the particle. The second complex exponential evaluates the phase delay associated to each displaced unit cell with respect to the center of Bragg peak.

This formula is also known as “*kinematical sum*” and it is implemented

Despite the close similarity to a classical discrete Fourier transform (DFT), one should notice that the variable \mathbf{R}'_n) is not uniform as the displacements disrupt the regular periodicity of the grid, required for a standard Fourier transform. It is therefore common to approximate this non-uniform discrete Fourier transform with a standard discrete one by assuming:

- $\mathbf{q} \cdot \mathbf{u}(\mathbf{R}_n) = 0$ which means that the projection of the displacements on the vector \mathbf{q} originating in the hkl and exploring the vicinity of the Bragg peak, is small. Given the relatively reduced volume of \mathbf{q} -space around the Bragg peak that is usually probed in BCDI this approximation is also reasonable. This is also known as the Takagi approximation [40]. When this approximation is applied Eq.1.33 takes the form of the so-called *kinematic*

sum and it is implemented in software for simulation of diffraction patterns starting from atomic positions like, for instance, the scattering module of PyNX [49].

- $S(\mathbf{R}'_n) = S(\mathbf{R}_n)$, which means that the shape function is not altered by the internal or superficial displacements. This is the case for displacements much smaller than the lattice parameter, which is easily fulfilled in typical BCDI samples. In practice, this approximation allows to describe the electron density on a periodic grid.

At this point, embedding the $e^{i\mathbf{G}_{hkl} \cdot \mathbf{u}(\mathbf{R}_n)}$ in a more general *complex-valued shape function* \tilde{S} and considering now the intensity of the signal impinging on the detector, we can write:

$$F_{\text{fin}}^{\text{strain}}(\mathbf{q}) = F_{\text{u.c.}}(\mathbf{G}_{hkl}) \sum_n \tilde{S}(\mathbf{R}_n) e^{i\mathbf{q} \cdot \mathbf{R}_n} \quad (1.33)$$

where we have now put the reference frame in the center of the Bragg peak at the hkl position, therefore expressing the scattering factor as function of \mathbf{q} .

This is the most important result of the chapter as later in the manuscript we will always refer to this complex-valued shape function as the “*complex object*” or “*reconstructed particle*” and will often deal with its complex phase. Specifically it will be used for simplicity the following relationship, which drops all the constant terms:

$$I(\mathbf{q}) = |\mathcal{F}\{\rho(\mathbf{r})e^{i\phi(\mathbf{r})}\}|^2 \quad (1.34)$$

Eq.1.34 has the advantage of expressing the diffraction pattern with a DFT, which allows for fast computations via efficient algorithms, rather than a sum evaluated in each individual unit cell position. This represents a key ingredient for the most used phase retrieval algorithms in BCDI (as we will see in the next Chapter), due to the large number of DFT operations required. Interesting works have evaluated the validity of these approximations [50] and the discrepancy between diffraction patterns calculated with the DFT and kinematic sum [51, 52] of

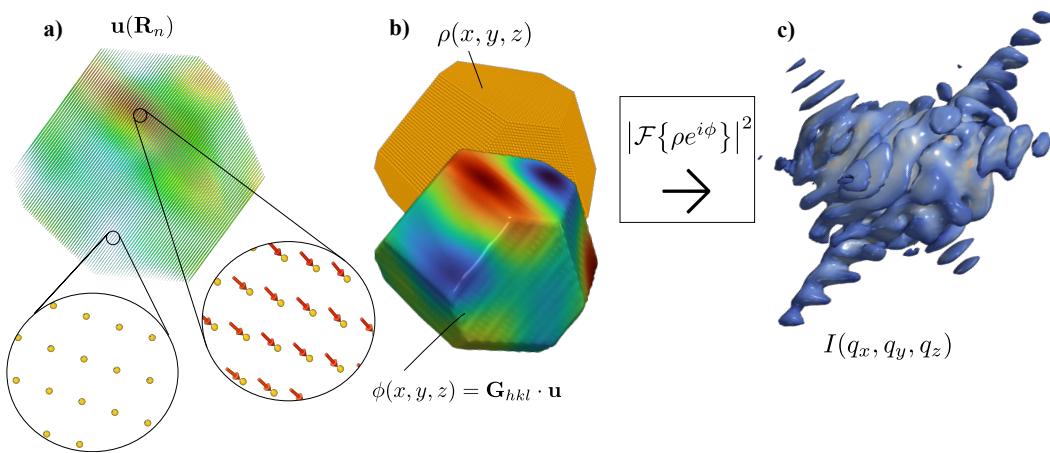


Figure 1.5: Schematic of the relationship between displacement field and diffraction pattern. **a)** Simulated displacement field obtained from the subtraction of the atomic positions of a simulated strained gold crystal with the relative perfect lattice copy. Close-up on two different regions where the displacement is small (left) and large (right). **b)** Formulation of the scattering object as a complex object. The modulus being interpreted as the electronic density and the phase being the projection of the displacement on the scattering vector pointing at the probed hkl node. **c)** Corresponding diffraction pattern proportional to the square modulus of the Fourier transform of the complex object.

It is interesting here to analyze what the effect of the displacement is on the shape of diffraction pattern. Intuitively we can think that the complex phase term is altering the interference of the scattered waves. If, for a perfect lattice, we had Dirac deltas broadened and modulated by the Fourier transform of the shape function, here we have to consider an additional broadening due to the strain. Moreover, we know from Friedel's law [53] that the diffraction pattern of a real valued function (shape function for perfect crystals) is always centro-symmetric while here, having introduced a complex term, we expect this symmetry to be broken. Fig.1.6 shows the effect of the displacement field on the diffraction pattern.

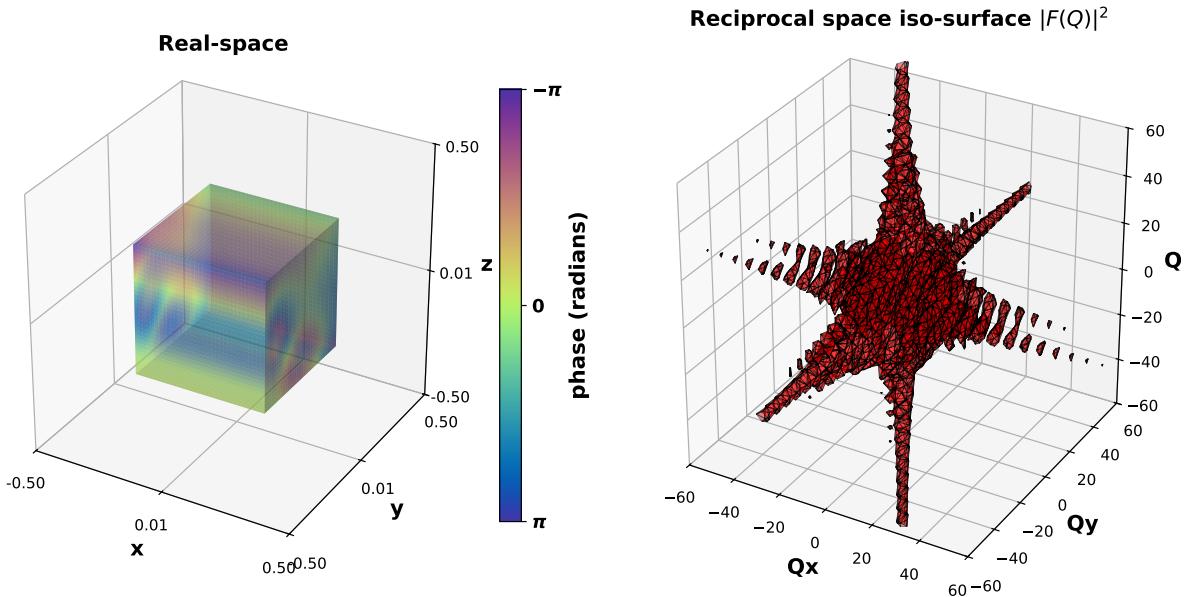


Figure 1.6: Illustration of the squared modulus of the Fourier transform of a cube with an applied displacement field. The phase derived from the projection of the displacement on the scattering vector is represented in on the left. On the right the corresponding diffraction pattern. It is visible the deformation of the Bragg peak with respect to the one in Fig.1.3 for the perfect lattice case.

1.2.6 Noise

We should now introduce the last ingredient to our derivation, the presence of noise. When dealing with real life experiment we have to consider that photons are quantum particles, thus their arrivals on the detector surface are random independent events. Here, because of the discrete nature of photons, a statistical approach based on the average arrival rate is required. It follows that an uncertainty is intrinsically associated to the estimate, hence the emergence of a source of noise. The statistical model describing these phenomena is known as Poisson statistic [54]. In particular, given a constant *average* rate of arrival η over a unit of time, the *expected* number of photons after an exposure time τ is $\mu = \eta\tau$, and the probability of detecting N photons over an observing time τ is given by the formula:

$$p(N) = \frac{(\eta\tau)^N e^{-\eta\tau}}{N!} \quad (1.35)$$

also called *Poisson probability distribution*. It can be proven that for the Poisson distribution the variance equals the mean $\mu = \sigma^2 = \eta\tau$. Moreover, we can intuitively think the expected number of photons collected during a time τ by a pixel in position \mathbf{q}_i to be proportional to the intensity in 1.34 evaluated in \mathbf{q}_i and integrated over a time τ . At this point, if we were to make a measurement, in absence of background, we would have at position \mathbf{q}_i an expected integrated intensity $\mu = \bar{I}(\mathbf{q}_i)$ with an uncertainty of $\sigma = \sqrt{\bar{I}(\mathbf{q}_i)}$. Being the Signal-to-Noise-Ratio (SNR) defined as $SNR = \mu/\sigma$ we obtain in our case:

$$SNR = \frac{1}{\sqrt{\bar{I}(\mathbf{q}_i)}} \quad (1.36)$$

Though very simple, this equation is important and deserves some comments. First, we can observe that in the limit of infinitely long integration times or infinite intensity (infinite flux), the $SNR \rightarrow 1$, meaning that we would in principle able to deterministically measure the diffraction pattern, in this case noiseless. Secondly, being most of the intensity of a diffraction pattern concentrated in the center of the Bragg peak, (Fig.1.4) we can deduce from Eq.1.36 that Poisson noise is affecting regions at high \mathbf{q} much more than at the center. During PR, this uncertainty in high \mathbf{q} ranges reflects, in real space, to a lower accuracy on fine resolution features of the reconstructed objects.

1.3 BCDI at ESRF - ID01

We have so far discussed the theoretical foundations for the understanding of the BCDI technique. It is now time to see the practical aspects.

Starting from the source of the probing radiation downstream to the detectors we will focus on the most important stages, necessary to envision the experimental conditions as well as typical numbers (resolution, energies, sizes).

BCDI experiments require a *coherent* X-ray beam focussed on a volume of few cubic microns. Moreover, such beam also needs to deliver a sufficient amount of photons to the sample which, being in the micrometer range as well possesses a limited scattering power. Such properties today can only be achieved in large scale facilities like synchrotrons and free electron lasers (FELs).

We know that electromagnetic waves can be generated by accelerating charges [55]. In synchrotron facilities electrons travel in a large circular ring (*storage ring*) at relativistic speeds and X-rays are produced deflecting their trajectories with the help of magnets. Depending on the magnet size, magnetic field and spatial configuration the X-ray beam is generated with different properties. Typically, these configurations are grouped into two main categories, namely: *bending magnets* and the more modern Insertion Devices (ID) like *wigglers* and *undulators*. In synthesis, while bending magnets produce X-rays in a broad energy spectrum with relatively low intensity, wigglers exploit series of smaller magnetic dipoles to increase the intensity of the radiation. Undulators on the contrary leverage the periodicity of the dipoles to stimulate coherent emission, hence sharpening the energy bandwidth around tunable peaks and increasing the intensity as well. The radiation generated by the latter is the most suited for coherent diffraction experiments because of its inherent higher coherence. An illustrative explanation is provided by Fig. 1.7 while an exhaustive description of synchrotron radiation can be found in the notable book from Prof. Giorgio Margaritondo [56].

The X-rays produced in these sections travel along straight lines, tangential to the storage ring, where they go through a number of optical elements dedicated to the spatial and spectral filtering as well as focusing and collimation, down to the experimental hutch where they finally radiate the samples. These are called *beamlines*, and they are usually designed and equipped for a specific class of techniques.

At the European Synchrotron Radiation Facility (ESRF) the ID01 beamline was conceived to combine the lattice parameter resolution provided by X-ray diffraction with spatial resolution of imaging techniques [27] and is able to offer various techniques for the investigation of strain at nanoscale, including BCDI.

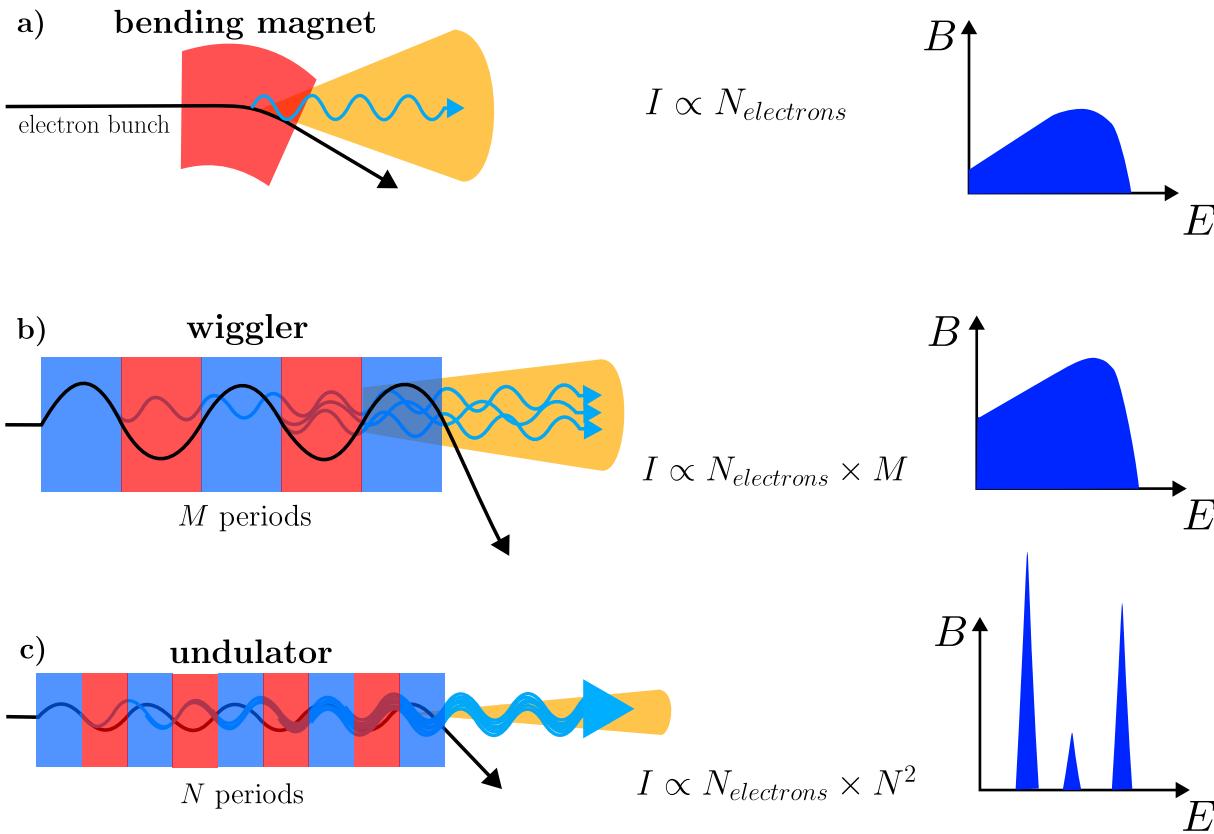


Figure 1.7: Illustration of the three main synchrotron X-ray sources. a) The intensity of the bending magnet radiation scales with the number of electrons being deflected. The beam is emitted with a broad horizontal divergence and the spectrum covers multiple wavelengths. b) The wiggler exploits a series of magnets to increase the intensity of the emitted radiation. c) In the undulator the periodicity of the poles is such that the photons are emitted with higher temporal coherence. The X-ray beam shows narrower emission cone (increased transversal coherence) as well as higher intensity and narrower bandwidths.

1.3.1 Beam size

Typical BCDI experiments are focused on single nanoparticles. The diffraction pattern is the result of the scattering of a single crystal in Bragg condition. It is therefore necessary to have a beam size of the order of the size of the crystal. At ID01 the beam size can scale down to $35 \times 35\text{nm}$ [57], but typical sizes are around $1\mu\text{m}^2$. A too large beam could shine some neighbor particles which, if in Bragg condition, could contaminate the diffraction pattern. These often appear as isolated bright spots, called *aliens* [58]. On the contrary, regions of the same particle that are not illuminated do not contribute to the diffraction pattern, therefore are not imaged in the reconstructed objects.

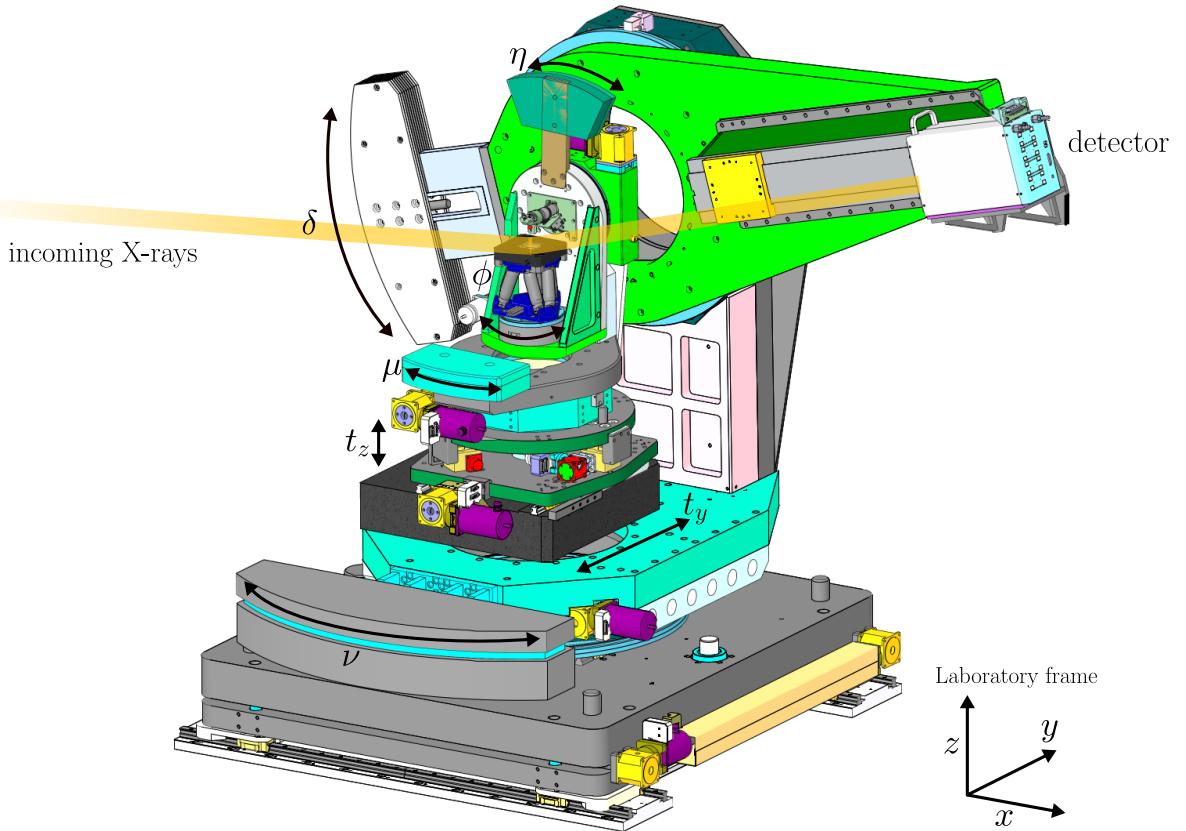


Figure 1.8: Computer Aided Design of the diffractometer at the ID01 beamline with the corresponding degrees of freedom. Adapted from [39]

1.3.2 Energy and flux

An important figure of merit for the characterization of synchrotron radiation is *brilliance*. It measures the number of photons populating the beam per unit of time, area and solid angle within a 0.1 % fraction of bandwidth relative to a certain energy. It is therefore defined as:

$$B = \frac{\text{nb photons}}{dt \cdot dA \cdot d\Omega \cdot 0.1\%dE/E} \quad (1.37)$$

ID01's radiation is generated by up to three undulators, one of which is tuned to deliver the optimum brilliance in the range of 6 - 11.5 keV. This energy range is particularly suited for coherent diffraction experiments on crystals . In fact, at lower energy absorption effects from optical elements and air start being important, hence reducing the net flux available on the sample. On the other hand, at higher energies other problems arise, namely: (i) the degree of coherence drops significantly (see next paragraph) and (ii) the diffraction patterns, for fixed detector distance, shrink in a lower number of pixels causing problems for PR.¹

¹This is related to the concept of the *oversampling condition* discussed in the next chapter

1.3.3 Coherence

This fundamental property of electromagnetic radiations is probably the most important for CDI. In the derivation of the scattering equation Eq.1.1 and following, we have always assumed the incident radiation to be described by a plane wave, perfectly monochromatic ($\Delta\lambda \neq 0$) and with no angular spreading ($\Delta k = 0$). In the ideal case, the wave-field is perfectly coherent, meaning that the relative phase between any two points in space is completely well-defined. In reality, electromagnetic waves have finite temporal duration and therefore, by Fourier transform relations, a finite spectral bandwidth. Likewise, they are produced by extended sources and observed at finite distances, which implies a finite angular spread of the wave-vectors. These limitations introduce phase decorrelation between fields evaluated at two different points, either *along* the propagation direction (due to bandwidth) or *across* it (due to angular dispersion). For this reason, it is useful to introduce quantities that characterize the “degree of coherence” of the radiation. An intuitive approach is to define a *longitudinal coherence length*, associated with phase delays from spectral bandwidth, and a *transverse coherence length*, associated with phase delays from angular spread.

Longitudinal Coherence Length L_l : is defined as the distance after which two beams originated in the same point and characterized by a wavelength difference $\Delta\lambda = 0$ have a phase difference of π , being therefore in phase opposition. This means that after a distance $2L_l$ the two waves are back in phase. If the first wave of wavelength λ needs N cycles to cover $2L_l$ it follows that the second wave with wavelength $\lambda + \Delta\lambda$ will need $N - 1$ cycles. Therefore:

$$\begin{aligned} N\lambda &= (N - 1)(\lambda + \Delta\lambda) \\ \text{thus, } \Delta\lambda(N - 1) &= \lambda \end{aligned} \tag{1.38}$$

In case of small $\Delta\lambda/\lambda$ we can approximate $N - 1 \approx N$, and obtain $N \approx \lambda/\Delta\lambda$. This leads us to the expression for the longitudinal coherence length:

$$L_l \approx \frac{\lambda^2}{2\Delta\lambda} \tag{1.39}$$

As expected the longitudinal coherence increases with the monochromaticity of the beam. On the contrary it significantly worsens for high energies, as anticipated above.

Transversal Coherence Length L_t is defined as the distance between two points A and B sitting on a plane at distance D from a source of size S for which both A and B are out of phase. Fig. shows geometrically that this distance, when $S \ll D \rightarrow \theta \approx 0 \rightarrow \cos(\theta) \approx 1$, the transversal coherence length is given by:

$$L_t \approx \frac{\lambda D}{2S} \tag{1.40}$$

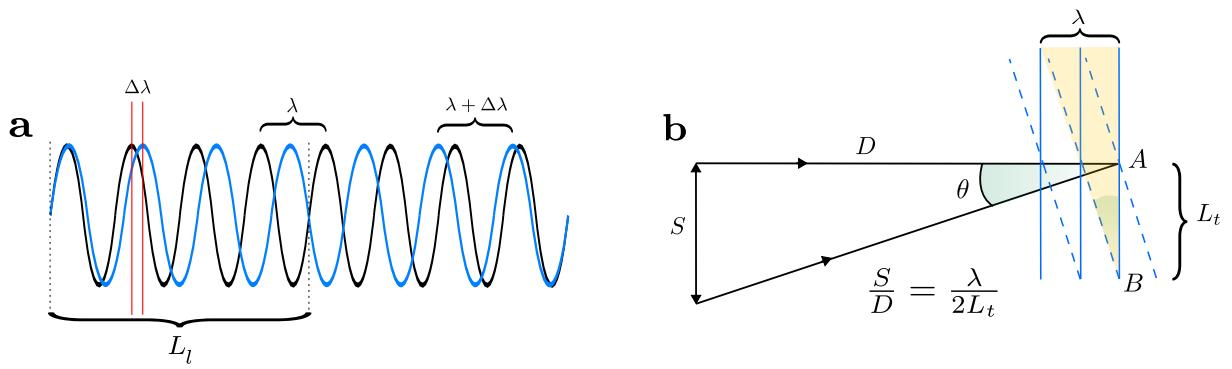


Figure 1.9: **a** Sketch of the longitudinal coherence length L_l . Two plane waves with slightly different wavelength originating in phase are out of phase after propagating over a distance equal to L_l . **b** Sketch of the transverse coherence length L_t . Two plane waves emitted at slightly different angles from an extended source are evaluated on a distant plane. They become out of phase when separated by L_t . Being the highlighted triangle similar to the bigger one in black, an expression for L_t can be derived from simple geometrical considerations

Also in this case we see the coherence decreasing for high energies. Moreover, the dependence on S and D explains the challenges to build long beamlines (118 meters for ID01) and reduce the electron bunches in storage rings. The transverse coherence length can be easily lifted to a coherence surface for a source extended in 2D ($L_t^{hor} \times L_t^{ver}$).

Given these premises we can now calculate the coherence *volume* ($L_l \times L_t^{hor} \times L_t^{ver}$) available ID01 for a typical X-ray energy used in BCDI. Given the source sizes of $60\mu m$ horizontally and $15\mu m$ vertically, at 8 keV ($\lambda = 1.55\text{ \AA}$) we obtain a transverse coherent surface of $152\mu m \times 610\mu m$. The longitudinal coherence length is found to be approximately $0.8\mu m$ considering the $\Delta\lambda/\lambda \sim 10^{-4}$ typical of the double Si (111) crystal monochromator. It follows that often, in BCDI experiments, the limitation on the particle size is given by the longitudinal coherence [59].

The coherence volume specifies the maximum spatial separation between two scatterers for which their scattered waves remain mutually coherent and can interfere. For this reason, while classical X-ray diffraction is limited to the interference of few lattice points, in the order of few \AA , the X-ray beam at ID01 can enable interference from objects separated by the order of few μm . In BCDI, the sample size is usually smaller than the coherence volume, meaning that scattering from all parts of the crystal, even from opposite surfaces, remains mutually coherent. This results in the characteristic interference pattern observed.

1.3.4 Ewald sphere and Rocking curves

At this point of the dissertation we should address the procedure related to data acquisition. In particular, in order to understand the generation of 3D diffraction patterns from the 2D images collected by the detector we need to introduce the concept of Ewald sphere.

We have said that in BCDI we focus on a single hkl node of the reciprocal lattice, and we collect the diffraction pattern of the full diffracting crystal around that Bragg peak. Given the

exit wave-vector \mathbf{k} pointing at the Bragg peak, we could draw a sphere of radius k centered in the sample and find the diffraction pattern of interest at the intersection of such sphere with the reciprocal lattice in the hkl node. Such sphere is called *Ewald sphere* and it is illustrated in Fig.1.10. More generally, one can state that the diffraction from a wave-vector \mathbf{k} occurs only in those reciprocal lattice points that intersect the Ewald sphere of radius k .

Moreover, the surface of the sphere around the Bragg peak tells us the signal captured by the 2D detector. In order to collect the 3D pattern one usually needs to apply a small rotation the sample along the axis that crosses the sample horizontal and perpendicular to \mathbf{k} such that the Bragg condition is found for the same hkl node in a slightly shifted position in space. The Ewald sphere is no longer cutting the diffraction pattern in the precise hkl node but in the extended peak broadened by the finite size effect. When this rotation, called *rocking curve*, is performed along both directions from one extremity of the pattern to the opposite, a 2D image is recorded by the detector at each angular step. The full stack of 2D images creates the full 3D diffraction pattern.

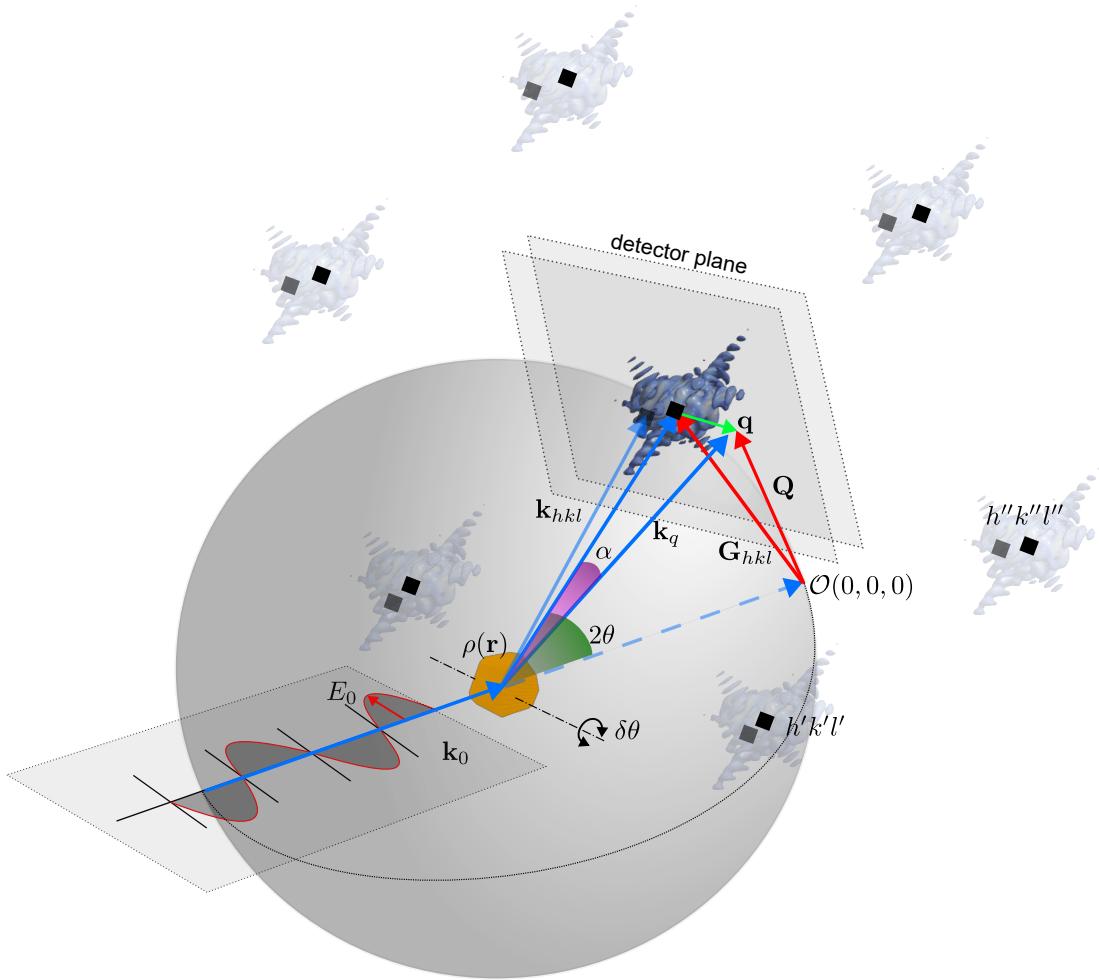


Figure 1.10: Illustration of the typical geometry for a BCDI experiment and the Ewald sphere (not in scale). A monochromatic plane wave approximates a coherent X-ray beam that fully illuminates sample with a wave-vector \mathbf{k}_i . A diffraction pattern given by Eq. 1.33 is formed around each hkl node. Of the different reciprocal lattice nodes a single one is selected for the measurement. The Ewald sphere, of radius k_{hkl} shows that the diffraction condition is met at the intersection with the hkl node. The detector plane, perpendicular to the \mathbf{k}_{hkl} vector, captures a swathe of Ewald sphere extending of an amount $\mathbf{q} = \mathbf{k}_q - \mathbf{k}_{hkl}$ around the hkl node. Different slices of the diffraction pattern are acquired rotating the sample at each angle $2\theta \pm N\delta\theta$. Notice that this angle is equivalent to the η angle in Fig.1.8, typically tuned during the rocking curve.

This approach assumes that the surface of the Ewald sphere in the vicinity of the Bragg peak can be approximated to be flat. In reality onto the detector is projected the slice of diffraction pattern extending over a swathe of Ewald sphere of radius $r = k$. It follows that for higher energies this approximation holds better as the contact angle is decreasing. Additionally, another consequence of the reduction of the contact angle at high energies is that the reciprocal space is shrinking. In order to understand this can consider that: geometrically the contact angle is equivalent to the angle α between the vectors \mathbf{k}_q and \mathbf{k}_{hkl} . Being now $\delta q = |\mathbf{k}_q - \mathbf{k}_{hkl}| = |\mathbf{k}_q| \sin(\alpha)$ one could infer that for higher energies (larger \mathbf{k}_{hkl}) the portion of diffraction pattern explored by $\delta q = |\mathbf{k}_q| \sin(\alpha) \approx |\mathbf{k}_q|\alpha$ is now shrunk into a smaller α .

This effect compresses the whole reciprocal lattice and thus the diffraction pattern as well, around the hkl node. This issue causes constraints in the sampling as will be discussed in the next Chapter.

CHAPTER 2

THE PHASE PROBLEM IN BCDI

This chapter is dedicated to the discussion of the *Phase Problem* in BCDI and the main computational methods that are currently adopted to solve it. This problem arose in the beginning in the field of X-ray crystallography since the first measured diffraction patterns, but similarly affects other domains like astronomical and seismic imaging as well as the coherent diffraction imaging of our interest. As anticipated briefly in the preface, the phase problem arises from a technical limitation. In X-rays, the fast oscillations of the electromagnetic fields induce detectors to only measure a time-averaged intensity (Eq.1.5) with the consequent loss of the phase information in the measurement.

The Fourier phase problem is therefore the impossibility to compute the complex-valued signal $\tilde{\rho}(\mathbf{r})$ from the intensity measurement of $I(\mathbf{q}) = |\mathcal{F}\{\tilde{\rho}(\mathbf{r})\}|^2$ with a simple inverse Fourier transform of the type $\mathcal{F}^{-1}\{\sqrt{I(\mathbf{q})}e^{i\varphi(\mathbf{q})}\}$ because of the lost reciprocal space phase $\varphi(\mathbf{q})$. It is thus necessary to find alternative strategies, often based on iterative algorithms, to perform the Phase Retrieval (PR) and recover the signal. However, as one can suspect, the modulus operation applied to the Fourier transform allows in principle an infinite variety of complex functions to be solution. For this reason the problem is said to be *ill-posed*. Consequently, the modulus operation makes it such that any PR algorithm is seeking the solution (*global minimum*) in a non-convex landscape of possible complex functions, populated by a number of “valleys” (*local minima*) in which the algorithm can get trapped. In this context, the seek of the solution to the problem has fascinated (and still does) scientists for decades, contributing to an extensive production of works in literature.

The first published studies date back to 1951 when Sayre in a comment [60] to the paper by Shannon *Communication in the presence of Noise* [61] in which a condition on the sampling of the diffraction pattern was proposed for the restoration of the unit cell extent. Later in 1972 Gerchberg and Saxton [62] developed an algorithm capable of inverting the diffraction pattern that is nowadays at the basis of currently used standard PR algorithms. However, proof of uniqueness of the solution arrived only later in 1979 by Bruck and Sodin [63]. The authors showed that, for 2D and 3D problems, the phase retrieval has unique solution except for rare cases, therefore conferring the mathematical solidity to the algorithm’s results. Later in 1982 Bates draws the link between uniqueness and the Sayre sampling intuition, as necessary condition for 2D case [64]. A refined version of the Gerchberg - Saxton algorithm was proposed by J.R. Fienup in 1978 [1] who named it Error Reduction (ER). In [2], published in 1982, the same author developed the Hybrid-Input Output (HIO) algorithm, able to outperform ER, and

compared gradient-descent methods as well. In 1987 again Fienup showed the possibility of reconstructing *complex-valued* objects if the constraints on the object support are “tight”, i.e. the shape of the object is known [65]. This result is particularly interesting for BCDI since, as we have seen in Eq.1.34, the object to be retrieved is complex-valued. Based on the suggestion of Sayre in 1991 [66] the works of Miao and coauthors from 1998 opened the X-ray coherent diffraction imaging field addressing the phase retrieval combining the sampling proposed by Sayre and iterative algorithms developed by Fienup [3, 67, 68]. From that moment on several works have corroborated the robustness of Fienup’s algorithms in .. However, the research on the Phase Problem did not stop there and among the many works published later it is worth mentioning the Difference Map algorithm [69], which generalized Fienup’s HIO introducing an additional non-linear term. Later in 2004, the work of Rodenburg and Faulkner [70] showed an improved PR convergence for diffraction patterns obtained illuminating the sample from multiple and partially overlapping regions. It was the birth of ptychography. More recently, Candès and cohautors have developed the PhaseLift algorithm [71] that turns the PR into a *convex* problem, thus improving the stability and convergence guarantees. In 2015, the same Candès [72] theorized the “Wirtinger flow” in which the PR is solved as least square problem with gradient descent using the Wirtinger derivatives [73] for complex functions and a *spectral initialization* method that enables the start of the PR near the global minimum. Further details on the applications of PR algorithms can be found in the review by Shechtman [74] while recent developments and theoretical insights can be found in the work of Fannjiang and Strohmer [75].

Here we will present first the sampling condition and the main alternating projections algorithms

2.1 Oversampling

Let us consider a direct space complex object $O(x)$ extended over a region of space R , and its Fourier transform $\tilde{O}(q) = \mathcal{F}\{O(x)\}$ in 1D defined like:

$$O(x) = \rho(x)e^{i\phi(x)} \quad \tilde{O}(q) = A(q)e^{i\varphi(q)} \quad (2.1)$$

The measurement of the diffracted intensity of the object would be equal to, barring constants, $I(q) = |\tilde{O}(q)|^2$. We should consider now that we are measuring $I(q)$ on a finite size detector made of discretized pixels. It thus follows the question: how finely in space should we sample the signal such that we can recover $O(x)$?

If $O(x)$ is a square of size R , Nyquist theorem states that each point sampling $\tilde{O}(q)$ should have a spacing $\Delta q = 1/R$. In our case we measure $|\tilde{O}(q)|^2$ which, corresponds to the Fourier transform or the so-called *autocorrelation function* of the object $(O(-x) * O(x))$, which extends over a size $2R$. Hence, the sampling should happen every $\Delta q = 1/2R$ to recover the autocorrelation of $O(x)$ without aliasing. This should in principle contains the information necessary to recover $O(x)$. This intuition was proposed by Sayre in 1952.

Following this idea a more rigorous explanation was given by Miao *et al.* in [3] in which the definition of *oversampling condition* is given. The salient ideas can be summarized as follows. With a hypothetical detector of N pixels on a line the extent of reciprocal space measured is $\Delta q = N\delta q$ where δq is the extent of a single pixel. The \mathbf{q} vector of Fig.1.10 is now discretized

in a q_k where $k \in [0, N - 1]$. In the direct space as well the coordinate x is now discretized into N values x_n where $n \in [0, N - 1]$ and the extent of direct space is $\Delta x = N\delta r$. According to the relationship between direct and reciprocal space the pixel size $\delta q = \frac{2\pi}{\Delta x} = \frac{2\pi}{N\delta x}$ which implies a Nyquist sampling. Hence, we can write the diffracted amplitude impinging on the detector as a discrete Fourier transform in each pixel.

$$\tilde{O}(q_k) = \sum_{n=0}^{N-1} O(x_n) e^{i \frac{q_k r_n}{N}} = \sum_{n=0}^{N-1} \rho(x_n) e^{i \phi(x_n)} e^{i \frac{q_k r_n}{N}} \quad (2.2)$$

Observing the above equation we can notice N variables but $N \times 2$ unknowns ($\rho(x_n), \phi(x_n)$), hence making the system under-determined. Using now Sayre condition of sampling at double the frequency $\delta q = \frac{2\pi}{2N\delta x}$ the system becomes solvable. In practice the size of the measured array is fixed by the detector, therefore one can reduce the number of unknown variables in the direct space to ensure a good sampling. In other words the object array is padded with a number of zeros determined by the oversampling condition defined as:

$$\sigma = \frac{\text{total pixel number}}{\text{unknown-valued pixel number}} \quad (2.3)$$

In the 2D or 3D case the same factor 2 needs to be fulfilled in order to have a (over)determined system of equations. However, one can calculate an oversampling ratio along each dimension d , resulting to be $\sigma \geq 2^{1/d}$ [76]. Nevertheless, it is preferable to ensure a larger value for σ along each dimension for better reconstructions [77].

Another interesting remark is that the oversampling condition can vary depending on the energy of the beam and on the distance of the detector with respect to the sample. In fact, we have seen that at high energy the reciprocal space shrinks (Fig.1.10), meaning that the same Δq is compressed into less detector pixels. Considering the detector positioned at distance D with respect to the sample, having a pixel size $p_{ix} \ll D$ we can approximate the angle subtended by the pixel as $\alpha = \frac{p_{ix}}{D}$. This angle is also approximated to be the angle subtended by $\delta \mathbf{q} = \mathbf{k}_q - \mathbf{k}_{hkl}$ as in Fig.1.10. We can therefore write:

$$\delta q = |k_q| \frac{p_{ix}}{D} \quad (2.4)$$

From which we see that to explore the same extent in q whilst fulfilling the oversampling condition, at high energies we need to have smaller pixel sizes or move the detector further away from the sample.

Beside this formal definition of the oversampling condition, it is useful to introduce a more practical one in terms of number of pixels. In particular, one can derive that fulfilling the oversampling condition is equivalent to a sampling in reciprocal space that is smaller than the distance between fringes by a factor of at least two. In other words, in the detector frame, at least two pixels are required between two fringes.

$$\delta q \leq \frac{\delta q_{\text{fringe}}}{2} \quad (2.5)$$

Ensuring a spacing of at least two pixels between fringes along all directions, naturally confines the object in direct space into an array which is at least half the size of the window containing the diffraction pattern in detector space.

2.2 Alternating projections algorithms

In this section the class of algorithms known as “alternating projections” (AP) mentioned above is presented, and the three most used algorithms in BCDI are described in more detail. We invite the reader to refer to the more exhaustive lecture notes by Cegielski [78] or the review written by Marchesini, from which the following paragraphs take inspiration [79].

Before delving into the details of each algorithm is important to clarify some fundamental concepts.

The goal of the Phase Retrieval is to reconstruct the complex object in direct space $O^*(\mathbf{r}) = \rho(\mathbf{r})e^{i\phi(\mathbf{r})}$ given the intensity measurement $I(\mathbf{q}) = |\mathcal{F}\{\rho(\mathbf{r})e^{i\phi(\mathbf{r})}\}|^2 = |A(q)e^{i\varphi(q)}|^2 = |A(q)|^2$.

The solution space is therefore a Hilbert space $\mathcal{H} \in \mathbb{C}^N$ where N is the number of complex-valued pixels, limited by typically two constraint sets \mathcal{C}_s and \mathcal{C}_m , defined as:

- $\mathcal{C}_s = \{O(\mathbf{r}) \in \mathcal{H} : O(\mathbf{r}) = 0 \quad \forall \mathbf{r} \notin \mathcal{S}\}$ Often called “support constraint” is the set containing all objects with zero amplitude outside the *support* \mathcal{S} . This last, in BCDI, coincides with the shape function encountered in Eq.1.18 and it is in principle unknown.
- $\mathcal{C}_m = \{O(\mathbf{r}) \in \mathcal{H} : |\mathcal{F}\{O(\mathbf{r})\}| = m\}$ Often called “modulus constraint” is the set containing all objects with Fourier transform of modulus m . This set is however “non-convex” as $|\mathcal{F}\{O(\mathbf{r})\}| = m$ is fulfilled for any reciprocal space phase. This poses challenges for deriving the convergence criterion of AP operating on this set [80].

The Phase Problem is then formulated as a *feasibility problem*:

$$\text{find the object } O(\mathbf{r})^* \in \mathcal{C}_s \cap \mathcal{C}_m \quad (2.6)$$

Moreover, we can define operators \mathcal{T} which transform the object according to the constraint set they are operating in, and are used to bring the current object estimate closer to the solution at each iteration. More precisely we can define:

- **Projector** onto the set C as $\mathcal{P}_C(x) = \arg \min_{y \in C} \|y - x\|$. It maps x to the nearest point y on the constraint set C in the Euclidean norm. The projector produces a *feasible point*, as the mapped point belongs to the constraint set C .
- **Reflector** with respect to the set C as $\mathcal{R}_C(x) = 2\mathcal{P}_C(x) - x$. It maps x to a point y across the constraint set C by applying two times the projector. Since y does not belong to C the reflector does not produce a feasible point.
- **Identity** \mathcal{I} as the operator that leaves unaltered the operated estimate.

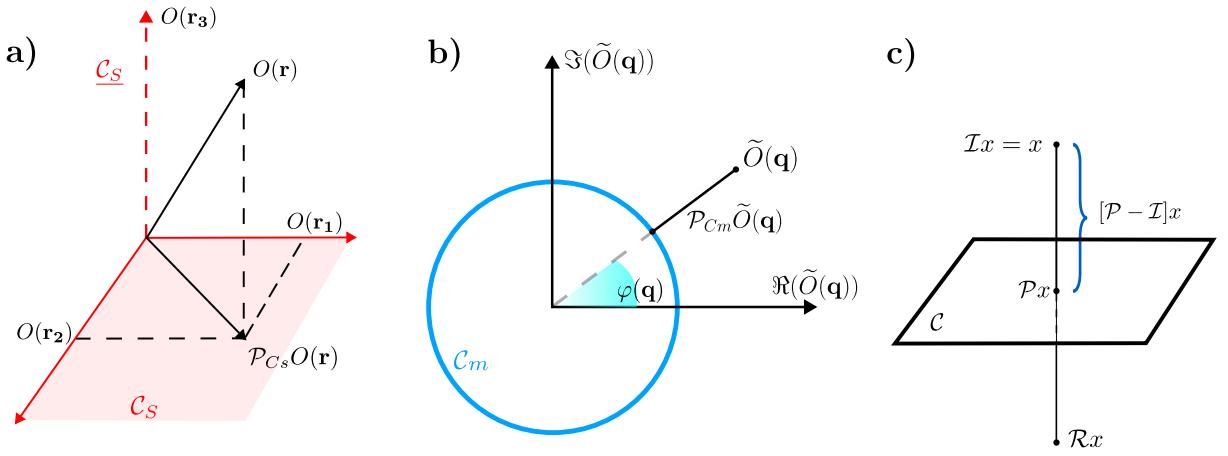


Figure 2.1: **a** Geometrical representation of the projection of the object onto the support constraint set. **b** Geometrical representation of the projection of the object onto the Fourier modulus constraint set. **c** Geometrical representation of the identity, projector and reflector operators. Inspired by [79]

In our case we have that the projector onto C_s applied to the object sets to zero the values outside the support and does not alter the values inside:

$$\mathcal{P}_{C_s}(O(\mathbf{r})) = \begin{cases} O(\mathbf{r}), & \mathbf{r} \in \mathcal{S} \\ 0, & \mathbf{r} \notin \mathcal{S} \end{cases} \quad (2.7)$$

On the other hand the projector onto C_m applied to the object replaces the modulus of its Fourier transform with the squared root of the measured intensity $m = \sqrt{I(\mathbf{q})}$.

$$\mathcal{P}_{C_m}(\tilde{O}(\mathbf{q})) = \mathcal{P}_{C_m}(A(\mathbf{q})e^{i\varphi(\mathbf{q})}) = \sqrt{I(\mathbf{q})}e^{i\varphi(\mathbf{q})} \quad (2.8)$$

This step forces the modulus of the Fourier transform of the object at the iteration k to be exactly the measured magnitude.

Since \mathcal{P}_{C_m} and \mathcal{P}_{C_s} operate in two conjugate spaces (direct-reciprocal), when used in sequence a direct or inverse Fourier transform is implied in between. The symbol will be omitted to simplify the notation.

At this point we have all the necessary ingredients to introduce the three main AP algorithms used for BCDI PR.

2.2.1 Error Reduction (ER)

If we consider as a starting point the object $O^0(\mathbf{r}) = \mathcal{F}^{-1}\{\sqrt{I(\mathbf{q})}e^{i\varphi^0(\mathbf{q})}\}$, obtained by the inverse Fourier transform of the squared root of the measured intensity with a random complex phase array $\varphi^0(\mathbf{q})$, we can express the object at the $k - th$ iteration of the *Error Reduction* (ER) algorithm as:

$$O^{k+1}(\mathbf{r}) = \mathcal{P}_{C_m}\mathcal{P}_{C_s}(O^k(\mathbf{r})) \quad (2.9)$$

This is the simplest and most intuitive AP algorithm as it only projects back and forth the object between the two sets. Although it guarantees linear convergence, ER is not optimal since it only converges to the nearest local minimum, and it is unable to escape it. For this reason in typical BCDI it is used to at the end of the PR, when the current estimate is close enough to the final solution. Additionally, one can define the magnitude error functional $\varepsilon_m(O)$ as:

$$\varepsilon_m(O) = \|\mathcal{P}_{Cm}(O) - O\|_{L2} \quad (2.10)$$

i.e. the Euclidean distance between the current estimate O and its projection onto the Fourier modulus constraint set. Differentiating this functional with respect to O yields the gradient $\nabla \varepsilon_m(O)$, which points in the direction that reduces the Fourier magnitude mismatch, or in other words, the natural descent direction of the error. If this gradient is further restricted to the support constraint set, one obtains a *projected gradient-descent step* $\nabla_s \varepsilon_m(O)$ which is the part of the descent direction that lies inside the feasible object region.

Marchesini shows that the ER step can thus be rewritten as:

$$O^{k+1}(\mathbf{r}) = \mathcal{P}_{Cs}(O^k(\mathbf{r})) - \frac{1}{2} \nabla_s \varepsilon_m^2(O^k(\mathbf{r})) \quad (2.11)$$

which makes explicit the equivalence between ER and steepest descent projected onto the support constraint set, with a fixed step size of $1/2$.

2.2.2 Hybrid Input-Output (HIO)

The Hybrid-Input Output (HIO) algorithm introduces a nonlinear feedback that is essential to escape local minima. Specifically, we can express the object at the $k - th$ iteration as:

$$O^{k+1}(\mathbf{r}) = \begin{cases} \mathcal{P}_{Cm}(O^k(\mathbf{r})), & \mathbf{r} \in \mathcal{S} \\ (\mathcal{I} - \beta \mathcal{P}_{Cm})(O^k(\mathbf{r})), & \mathbf{r} \notin \mathcal{S} \end{cases} \quad (2.12)$$

where β is a positive hyperparameter with value typically around 0.9. Inside the support, the estimate is replaced by its Fourier-modulus projection. Outside the support, instead of being set to zero (as in ER), the estimate is updated with a feedback term proportional to the modulus projection. This subtraction, which can in principle yield negative values as well, prevents stagnation and allows the algorithm to explore solutions that are consistent with both the support and modulus constraints. However, it is worth mentioning that due to the nonlinear feedback outside the support, HIO does not converge to local minima but rather towards *saddle* points of the error functional. For this reason HIO is used in combination with ER: HIO drives the reconstruction away from traps by oscillating near saddle points, and ER then provides stable convergence once the estimate is close to a true solution.

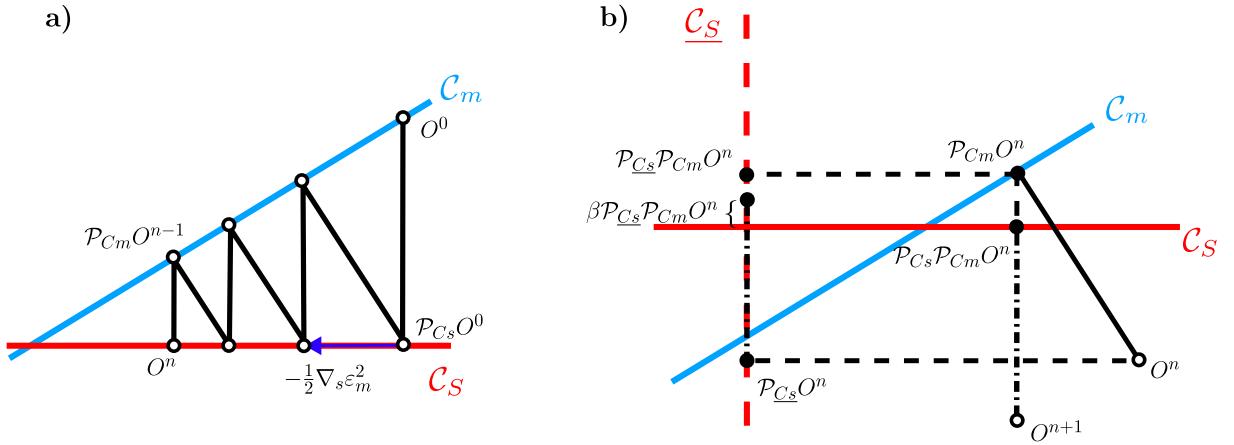


Figure 2.2: **a** Geometrical representation of the ER algorithm. Red and blue lines represent the support and Fourier modulus constraint sets respectively. The alternated projections on the two constraint sets drive eventually the estimate to the intersection. This is however only valid in a local subset of the solution space, where the - generally non-convex - modulus constraint set is convex and . **b** Geometrical representation of the HIO algorithm . Inspired by [79]

2.2.3 Relaxed Averaged Alternating Reflections (RAAR)

Another AP algorithm typically used in CDI is the Relaxed Averaged Alternating Reflections (RAAR) developed by Luke in 2004 [81]. According to this algorithm the object at the k -th iteration is:

$$O^{k+1}(\mathbf{r}) = \beta \frac{1}{2} (\mathcal{R}_{Cs} \mathcal{R}_{Cm} + \mathcal{I})(O^k(\mathbf{r})) + (1 - \beta) \mathcal{P}_{Cm}(O^k(\mathbf{r})) \quad (2.13)$$

where $\beta \in [0, 1]$. Eq.2.13 can be split into two terms. The first term, known as Average Alternating Reflection (ARR) [82] acts like an average of the current estimate and its reflection across both constraints sets. The alternated reflections search for the intersection of the two sets, which is a fixed point of the problem, while exploring more broadly the solution space as they do not project directly on the constraint sets. The second term is a relaxation term that projects the current estimate on the modulus set, like in the HIO and ER.

In short, one could see the RAAR as a controlled (through β) balance of exploration and stability.

2.2.4 Support update

At this point of the discussion we should ask ourselves how do we know the *support* function \mathcal{S} required in all PR algorithms? We have said that in the BCDI case we typically do not know the shape of the particle a priori, and should therefore come as product of the PR. It is common practice in iterative phase retrieval to estimate the initial support from the object's autocorrelation,

$$A(\mathbf{r}) = \mathcal{F}^{-1}\{I(\mathbf{q})\} \quad (2.14)$$

However, the autocorrelation extends over a region roughly twice the linear size of the true object (see Fig.).

An important step forward was introduced by Marchesini in 2003 [83]: the *shrinkwrap algorithm*, which adaptively refines the support during reconstruction. After a given number of phase retrieval iterations, the modulus of the current object estimate is convolved with a Gaussian kernel and subsequently thresholded (typically at $\sim 20\%$ of the maximum value), yielding a binary mask that defines the updated support.

This adaptive support refinement was shown to significantly improve convergence and reconstruction quality in many experimental cases (depending on the signal-to-noise ratio and choice of threshold), and it is now a standard component of BCDI phase retrieval pipelines.

Later in the text we will refer to “standard iterative algorithms” or “conventional PR” as the combination of AP algorithms and support updates like Shrinkwrap.

KEYNOTES ON CONVOLUTIONAL NEURAL NETWORKS

In this chapter a short overview on the basic concepts of Convolutional Neural Network will be given.

3.1 Context: CNN for Inverse Problems

3.2 Principles and building blocks

3.3 U-Net and MSD-Net

3.4 Training

CHAPTER 4

DEEP LEARNING FOR DETECTOR GAPS INPAINTING

In this chapter, the “detector - gaps problem” in Bragg Coherent Diffraction Imaging and the approach to solve it using Deep Learning (DL) are discussed. The main state-of-the-art methods are presented briefly and the topic of image inpainting with DL is introduced. The focus will then shift to the works that led eventually to the optimal “Patching-based” approach that can also be found in the published paper entitled “*Patching-based deep learning model for the Inpainting of Bragg Coherent Diffraction patterns affected by detectors’ gaps*” (<https://doi.org/10.1107/S1600576724004163>). The chapter is closed with some analyses of the performance of the DL models in a variety of simulated and experimental cases.

4.1 The “Gap Problem”

At time of writing, standard BCDI experiments employ pixelated photon counting detectors to record the diffraction patterns. These detectors can guarantee high spatial resolution, low-noise counting and fast read-out times. Two examples of these devices, currently used at the ID01 beamline are the MAXIPIX and EIGER detectors [44, 84]. These detectors are often built by tiling together several sensing chips in order to cover a larger area, and are typically bonded to an Application-Specific Integrated Circuit (ASIC) using bump bonding. This implies always some physical connection which is not a pixel on one side of a chip. Additionally, intra-chip edges also constrain larger pixels which disrupt the oversampling condition, hence cannot always be used. These issues result in the presence, in the overall sensing region, of vertical and/or horizontal stripes that are not filled with useful signal. The width of these lines varies depending on the device but normally does not exceed the equivalent of some tens of pixels. Specifically, the MAXIPIX detector, with sensing area of 516×516 pixels of $55\mu m \times 55\mu m$, is composed of four modules separated by $220\mu m$ wide gaps (equivalent of 4 pixels) while the edge pixels measure $165\mu m$ (i.e. 3 pixels), thus making a gap, when adjacent, of 6 pixels width.

The EIGER detector instead has two types of larger gaps of 12 pixels and 38 pixels width. The detector gaps problem does not affect BCDI only, but it is shared among other x-ray techniques that deal with single photon-counting pixelated detectors and/or beamstops. We have seen in chapter 0.1 that during a BCDI scan the 2D images acquired by the detector are

stacked to form a 3D array. This leads these lines to become planes of missing signal in the dataset. The problems arise when reconstructing the data with these gaps. In fact, these regions of non-physical zero intensity deceive the Phase Retrieval algorithms inducing the presence of artifacts in the reconstructions [85] (see Fig.4.1).

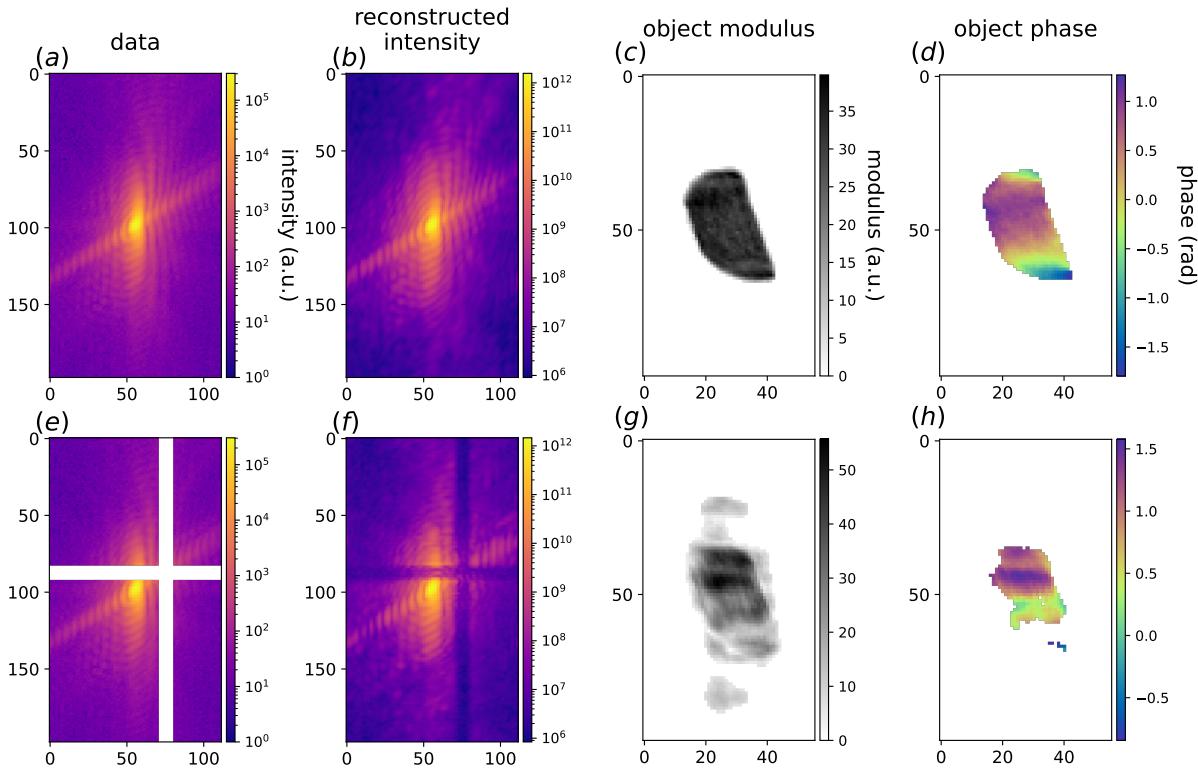


Figure 4.1: Effect of detector gaps in BCDI reconstructions (a) The central slice of an experimental diffraction pattern. (b) The same slice of the diffracted intensity calculated from the retrieved object. (c - d) slice of the modulus and phase respectively of the particle obtained from the phasing of the gap-less dataset. (e) Same slice as in (a) with an artificially added 9 pixel-wide, cross-shaped gaps to mimic the detector's ones. (f) The same slice of the diffracted intensity calculated from the retrieved object when not masking the gap regions. (h - g) slice of the modulus and phase respectively of the particle obtained from the phasing of the gap-affected dataset. The distortions caused by the gaps are evident. Note that for this figure an extreme but explanatory case, (large gaps near central peak of a low-oversampled data) was illustrated.

It follows that the reliability of the reconstructions in this case is compromised as the strain distribution can be deeply affected by the artifacts. A good practice during standard BCDI experiments is to avoid the gaps by moving the detector if possible. However, this tends to be problematic for the case of high-resolution BCDI, i.e. when the diffraction pattern measurement extends to higher q -values, thus covering more than one sensing chip and necessarily crossing a gap region. Under these circumstances it becomes important to reduce the amount of artifacts deriving from the gaps.

4.2 State of the art

Here we will discuss the current strategies employed to treat the detector gaps. As someone could argue, the simplest yet not practical, solution would be to slightly move the detector sideways and acquire a second full scan with the gap hiding a different region of the same Bragg peak, and then merge the two measurements into a single gap-less one. This would at least double the acquisition time making it, de facto, not an option during standard experiments.

The PyNX software, routinely used for the BCDI phase retrieval at ID01, allows the user to define a mask of the gap regions and ignore those pixels during the execution. This improves the quality of the reconstruction, but one can still notice the presence of oscillations appearing in both the object's modulus and phase. The origin of these artifacts can be found in the diffracted intensity calculated from the reconstructed particle as one can clearly see that the gap-regions is filled with non-physical high intensity (see Fig. 4.2(f))

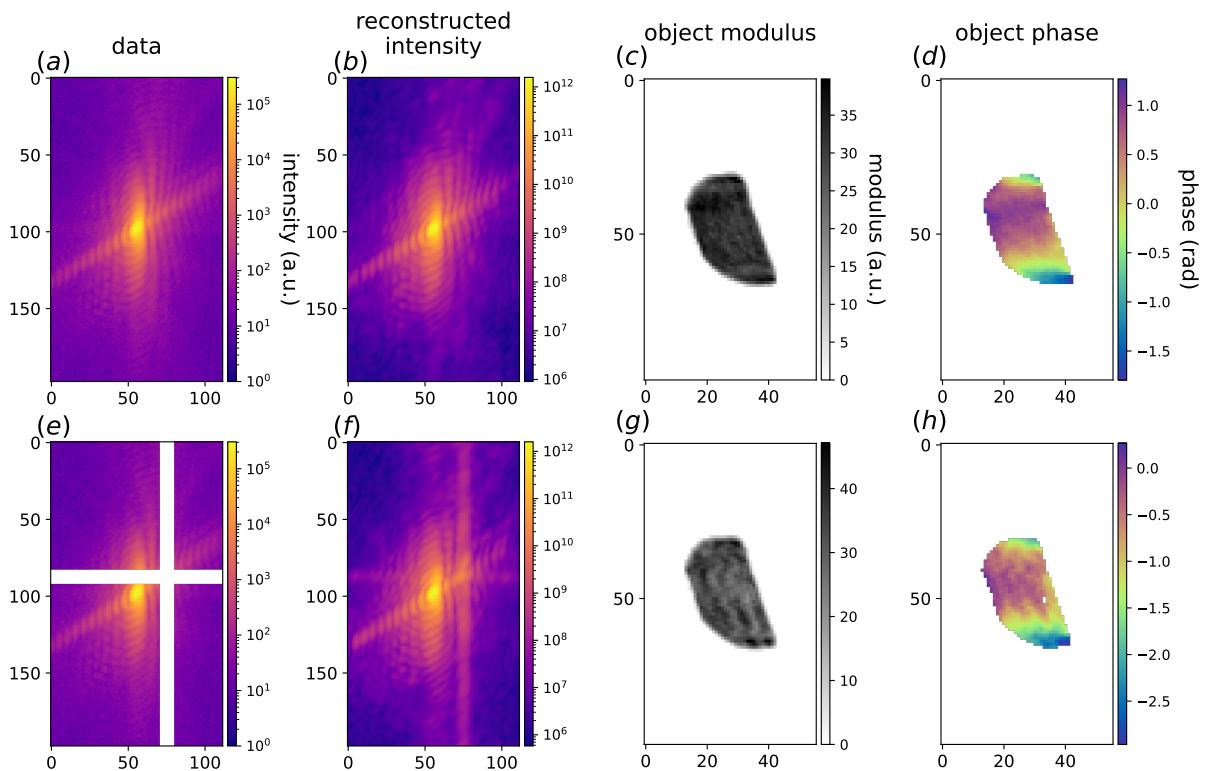


Figure 4.2: Masking the gap region during phasing (a) The central xz slice of an experimental diffraction pattern. (b) The same slice of the diffracted intensity calculated from the retrieved object. Comparing this figure with 4.1(b) one can see that when excluding the gap region from the phasing with a mask, the calculated intensity shows bright non-physical streaks instead of the gaps. (c - d) xz slice of the modulus and phase respectively of the particle obtained from the phasing of the gap affected data with a mask of the gap regions. Despite the much higher quality of the reconstruction, one can notice some oscillatory artifacts appearing in both the modulus and the phase of the retrieved object.

Methods relying on compressive sensing and TV regularization were proposed by He in [86] and Malm [87] respectively. Another, more invasive, option is to *fill* these gaps with an estimate of the intensity distribution that would be there, before the phase retrieval. The task of filling gap in images is usually referred to as “inpainting”.

4.2.1 Background on Image Inpainting Research

Computational image inpainting has been widely studied in the field of photography and imaging for many years [88, 89]. The inpainting problem can be defined as the task of using known information extractable from the image, to repair the parts where this information is missing, where for known information the colors, the textures and the semantic features are intended. In the history of image inpainting a clear-cut can be observed when deep learning methods have started to be employed. For traditional inpainting, different techniques have been explored, from the texture synthesis methods pioneered by Efros and Leung [90] to the use of PDEs as Navier-Stokes equations proposed by Bertalmio *et al.* [91] and then again from sparse representations [92] to hybrid methods combining variational and statistical methods [93].

More recently, Deep Learning models, headed by Convolutional Neural Networks (CNN), have taken the place of more traditional methods as they can attain higher accuracy for more complex inpainting tasks. By undergoing a training process, CNNs can “learn” to recognize and reproduce the semantic features of the training dataset, and thus leverage them during inference as additional information beside the colors and textures of the specific image to restore. As we have seen in 3, the typical CNN architecture for image generation consists of an encoder, which retains the features of the input image and compresses them into a lower dimensional latent space, and a decoder, which is responsible for the generation of the output image starting from the latent space. The model are then trained according to a loss function that pushes the model’s predictions to be close to a given ground truth reference. In some cases, the loss function can be replaced by another CNN that is trained to discriminate true images from the ones predicted by the model. These complementary networks are known as Generative Adversarial Networks (GAN), firstly proposed by Goodfellow *et al.* [94], and have also been used for image inpainting (e.g. [95]).

Since reviewing the vast amount of works about CNN for image inpainting is beyond the scope of this thesis and for more information, we redirect the reader to the reviews written by Elharrouss *et al.*, Xianget al. and Xu *et al.* [89, 96, 97]. For what concerns the application of DL based inpainting for scientific imaging, early works date back to 2018 as in the case of Sogancioglu *et al.* for x-ray human chest 2D radiographic images [98] and to 2020 for 2D microscopic images [99]. A couple of years later Tanny Chavez and coauthors published a paper comparing the performances of different CNN models for the inpainting of 2D x-ray diffraction images [100]. The work is precisely addressing the gap problem for x-ray detectors used for powder diffraction measurements and is awarding UNet and Mixed Scale Dense (MSD) models for the best performance on experimental data. The DL models outperform interpolations obtained with biharmonic functions across 7 and 17 pixel-wide gaps. This work has been of inspiration for the design of our DL model for BCDI gaps inpainting. In the same year, another work on DL based inpainting for x-ray detector gaps was published by Alfredo Bellisario and coauthors [101]. The authors tested a UNet-like model on the inpainting of 2D simulated, noiseless coherent diffraction patterns against gaps of different sizes (2 to 20 pixels) along the central row. The gaps were placed such that the center of the peak was covered, a choice that, as we will see later, yields better results than predictions on peripheral areas. To our knowledge, at the time of writing, no other works about deep learning based inpainting for X-ray detector gaps are present in the literature.

4.3 Model design: 2D case

For simplicity, I have begun with the 2D case, using simulated diffraction patterns and inpainting randomly placed vertical gaps of different width. First, a training set of simulated data was created, composed of pairs of gap-affected images and corresponding gap-free ground truths, then a U-Net-like model was built and trained in a supervised fashion.

4.3.1 Dataset creation

The creation of training datasets of simulated 2D BCDI patterns for both the gap-inpainting and phase retrieval tasks has followed the procedure described in this paragraph.

In first place, once chosen the size of the array, a randomly shaped polygon was created in the center using `scipy.spatial.ConvexHull` function. This guarantees the object to have a compact support with homogeneous electron density as assumed for BCDI. Subsequently, a random phase field of the same size with variable phase range and correlation length was generated thus the complete complex object was formed.

In order to make the object more realistic a Gaussian filter and Gaussian - correlated noise [102] have been applied to the object's modulus, so to smoothen the edges and simulate real cases respectively. At this point the object was resized to the shape required to match the chosen oversampling ratio and the 2D Discrete Fourier Transform was computed. As last stage, Poisson noise was added to the diffraction patterns with different magnitudes to simulate various X-ray flux conditions (see Fig.4.3).

Datasets contain a number of diffraction patterns in the order of thousands and for each of them the random variables have been varied as well as the oversampling ratios. In the datasets for the training of phase retrieval models (presented later in Chapter 5), the reciprocal space phase corresponding to each diffraction pattern was saved as well and used as ground truth label. For inpainting tasks a randomly located vertical gap mask was created and applied to the intensity data. In some cases cross-shaped gaps were added instead to simulate the experimental condition of the Bragg peak in the vicinity of the corner of the sensing area. The size of the gaps was chosen to be consistent across the dataset and four different cases were studied (3px, 6px, 9px, 12px). Fig.4.3.

4.3.2 2D Model design

The 2D model that I have implemented is a U-Net that takes in input batches of 32 simulated BCDI patterns affected by both vertical and cross-shaped gaps. Each diffraction pattern was transformed into logarithmic scale to enhance the spatial features and then normalized between 0 and 1. This normalization was proven to be convenient to any DL model [103]. Regarding the logarithmic transformation, it is important to notice that in order to avoid problems for zero intensity values, the $\log(I + 1)$ was taken. The shape of each image was chosen to be of 128×128 pixels. The encoder of part of the models was built as follows. The inputs go through five convolutional blocks inside each of which a convolutional layer, a Leaky ReLU activation function and a MaxPooling operation are applied. The feature map's size is therefore reduced down to 2×2 while the channel dimension is brought up to 768 filters while the

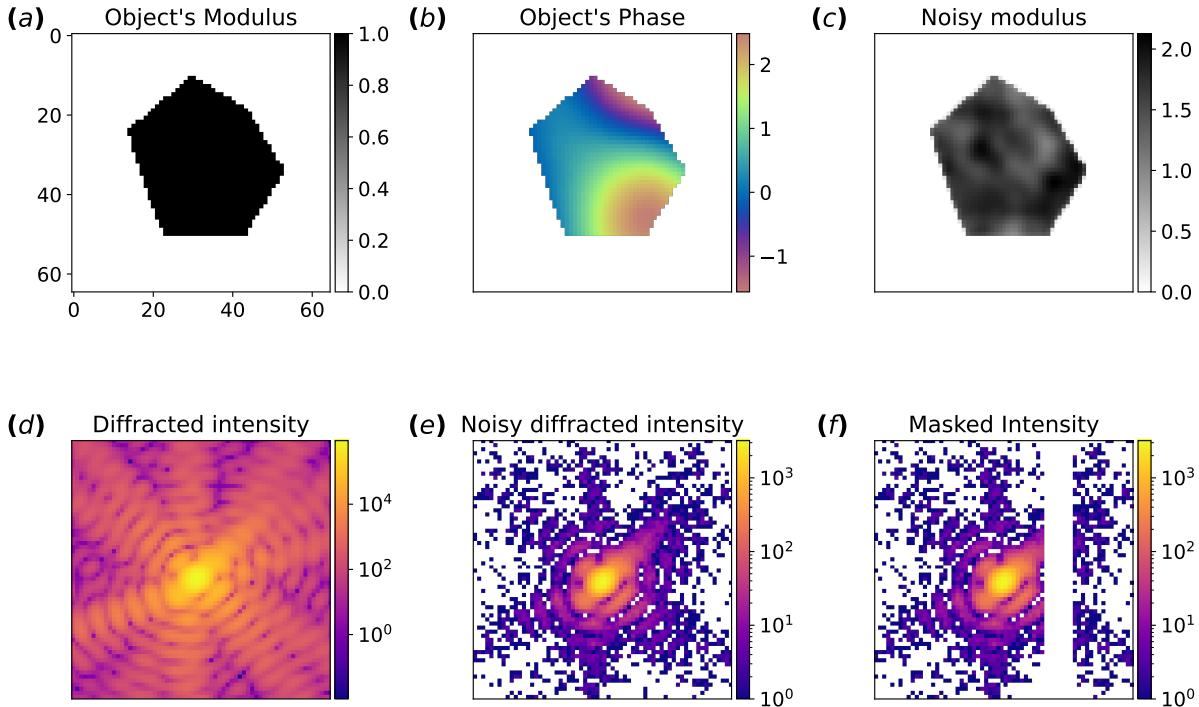


Figure 4.3: Steps for the simulation of a single 2D diffraction pattern (a) Simulated modulus of a 2D object with random shape and compact support. The object is padded with zeros to match the chosen oversampling ratio. (b) Simulated object’s phase (c) Object’s modulus after smoothening the edges and adding random Gaussian noise. (d) Squared modulus of the Fourier Transform of the complex object (in log scale). (e) Poisson noise is added to the simulated diffracted intensity. (f) A 6 pixel-wide vertical gap is added to the diffracted intensity at a random position.

kernel size is kept at 3×3 . At the end of the encoder the feature map passed to the decoder is a $(32, 2, 2, 768)$ tensor. The decoder, mirroring the encoder, is composed of five blocks inside each of which there is a transposed convolutional layer that upsamples the feature maps (stride = 2) and a Leaky ReLU activation function. Skip connections connecting each encoder block to its corresponding shape-like decoder block have been implemented as well. This measure has proven to be beneficial for the information flow between encoder-decoder [104]. The last activation layer of the model is a sigmoid function that guarantees an output bounded between 0 and 1.

A simple Mean Squared Error (MSE) was used as cost function inside the gap region only, training the model on 12'000 diffraction patterns over 10 epochs, with ADAM optimizer and a learning rate of 10^{-4} . We have tested the Mean Absolute Error (MAE) and the Structural Similarity Index Measure (SSIM) [105] as well afterwards and compared the results after the same training. Here in Fig. 4.4 we report the comparisons for the 9 pixel-wide gap on a test simulated diffraction pattern. The accuracy scores were calculated using the Pearson Correlation Coefficient (PCC).

$$PCC = \frac{\sum_{i \in \text{gap}} (\mathbf{I}_i^{\text{true}} - \langle \mathbf{I}^{\text{true}} \rangle)(\mathbf{I}_i^{\text{pred}} - \langle \mathbf{I}^{\text{pred}} \rangle)}{\sqrt{\sum_{i \in \text{gap}} (\mathbf{I}_i^{\text{true}} - \langle \mathbf{I}^{\text{true}} \rangle)^2} \sqrt{\sum_{i \in \text{gap}} (\mathbf{I}_i^{\text{pred}} - \langle \mathbf{I}^{\text{pred}} \rangle)^2}}, \quad (4.1)$$

Where \mathbf{I} is the intensity inside the gap. The PCC measures the linear correlation between two sets of data, and it is naturally normalized between -1 and 1 for which the two datasets are said to be linearly correlated. On the contrary the 0 value implies uncorrelated sets.

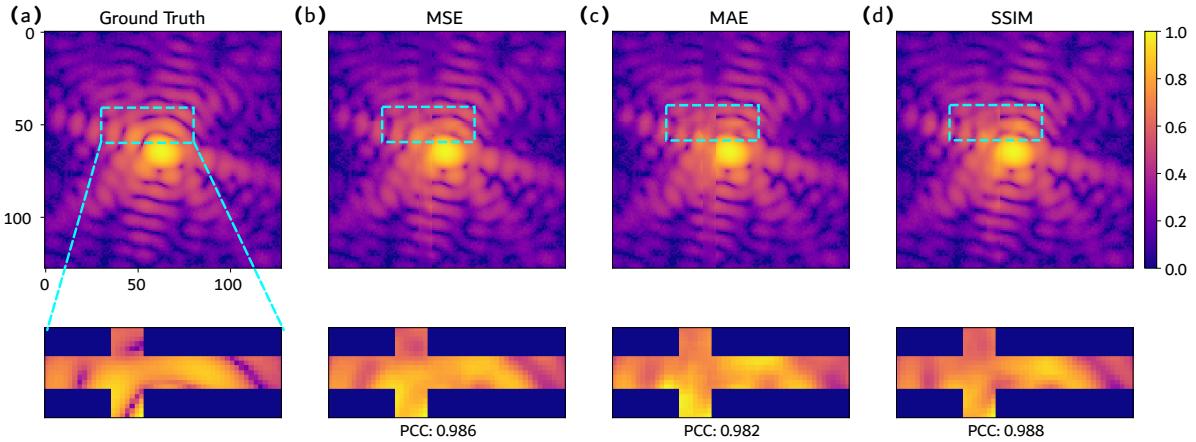


Figure 4.4: Comparison of different losses Results on a test simulated diffraction pattern for the inpainting of a 9 pixel-wide cross-shaped gap produced by the same UNet model trained for 10 epochs with different loss functions. (a) Shows the ground truth. (b) The prediction of the model trained with the MSE, (c) with the MAE, (d) with the SSIM. Corresponding accuracy scores calculated with the Pearson Correlation Coefficient (PCC) are shown as well. While MAE fails to recover the oscillations, SSIM yields better results.

In the light of these results, the MAE metric was discarded and the sum of MSE and SSIM adopted instead. Finally, another term that computes the MSE between the *gradients* of the ground truth and predicted intensity inside the gap region was added in the definitive loss function.

Once established what was considered the best loss function, different models have been explored . Following the work of Chavez *et al.* mentioned above ([100]), a Mixed-Scale Dense Network (MSD-Net) was used. The advantage of this type of networks is the significant reduction of trainable parameters, and the use of *dilated* convolutions with respect to U-Net ones. While the former property guarantees faster training and lower chances of overfitting, the latter enhances the capture of long-range correlations. Moreover, in an MSD-Net, the image's spatial dimensions are kept constant throughout the whole network as no downsampling nor upsampling is made. The MSD-Net consists of sequential blocks in each of which the input is transformed by two different convolutional layers with growing dilation rates. Each output of the convolutional layers is concatenated to the input feature map and the result is passed to the following block. While the kernel size is kept constant to $3 \times 3 \times 3$ pixels the dilation rate increases linearly from 1 to 30. The last layer is a sigmoid function as well as for the U-Net. The total number of trainable parameters is of the order of 320'000, two orders of magnitude lower than the U-Net.

In order to combine the hierarchical dimensionality reduction of the U-Net with the fine-features capturing of the MSD-Net we have implemented a modified U-Net that adopts dilated convolutions inside the first three encoder blocks. In particular, they return the input tensor concatenated with the outputs of four dilated convolutional layers computed from the input. Dilation rates of (16,8,4,2), (10,5,3,1) and (5,3,2,1) were chosen respectively. As the MaxPooling

operation down-samples the feature maps into smaller sizes, we limited the dilated convolutions to the first three blocks. Moreover, they were used in the encoder layers only as they are mostly used for feature extraction [106]. The characteristics of each model are summarized in form of pseudo - code in Table 4.1.

The three different models have been trained with a combined loss function (MSE + SSIM + MSE on the gradients) on the same training dataset for 10 epochs each. The results showed poor performances of the MSD-Net with respect to the two U-Nets. Slightly higher accuracy was achieved by the modified U-Net (see Fig. 4.5).

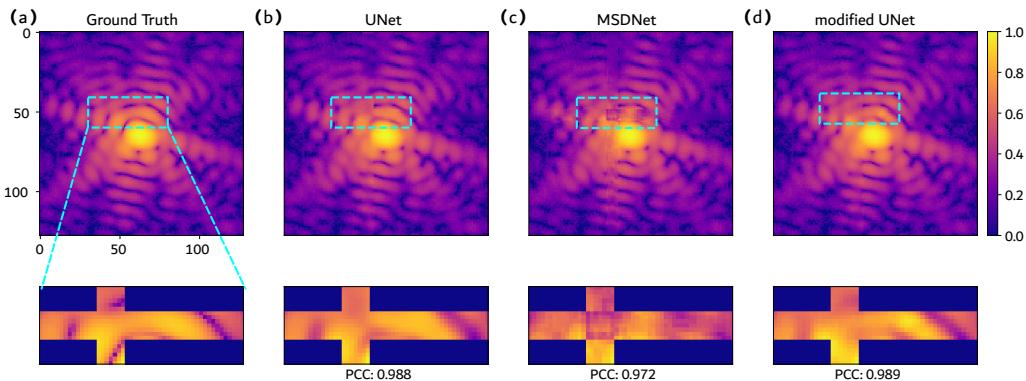


Figure 4.5: Comparison of different models Results on a test simulated diffraction pattern for the inpainting of a 9 pixel-wide cross-shaped gap using three different models trained with the same loss function. (a) Shows the ground truth. (b) The prediction of the U-Net, (c) of the MSD-Net, (d) of the modified U-Net. Corresponding accuracy scores calculated with the Pearson Correlation Coefficient (PCC) are shown.

We conclude here the introductory studies on 2D simulated data. These preliminary tests allowed to explore different DL architectures and loss functions and select the optimal choices for the inpainting of BCDI detector gaps.

4.3.3 Accuracy VS Gap position

Before moving to the 3D case, it is worth spending a few words on the assessment of the DL model upon different conditions. Namely, the accuracy as function of the position of the gap inside the diffraction pattern and as a function of the oversampling ratio. For the first test 150 2D diffraction patterns were simulated starting from random particles shapes, random oversampling ratios and Poisson noise intensity. For each diffraction pattern a vertical 9 pixel-wide gap was placed at all positions from left to right, then the DL prediction and corresponding accuracy score when compared to the ground truth were calculated. The accuracy was again calculated with the Pearson Correlation Coefficient. This score was then averaged for each gap

	U-Net	MSD-Net	Unet_mod
block1	<pre> def encoder_block(x_input, num_filters, ker): s = Conv2D(num_filters, ker, 'leaky_relu')(x_input) x = MaxPool2D(2)(s) return x, s </pre>	<pre> def MSD_block(x, in_channels, dilations,kernel_size=3): if isinstance(dilations, int): : dilations = [(j % 10) + 1 for j in range(dilations)] out_channels = in_channels + len(dilations) for d in dilations: x1 = Conv2D(out_channels //2,kernel_size,1, dilation_rate= dilation, 'same', 'leaky_relu')(x) x = tf.concat([x1,x] ,axis = -1) return x, out_channels </pre>	<pre> def encoder_block_mod(x_input, ker, num_filters, rate): f = num_filters // 4 s = tf.concat([x_input] + [Conv2D(f, ker, dilation_rate=r, 'leaky_relu')(x_input) for r in rate], axis=-1) return MaxPool2D(2)(s), s </pre>
block2	<pre> def decoder_block(x_input, num_filters, ker, skip_input = None): if skip_input is not None: x_input = Concatenate()([x_input, skip_input]) x = Conv2DTranspose(num_filters, ker, strides=2, 'leaky_relu') (x_input) return x </pre>		<pre> def decoder_block(x_input, num_filters, ker, skip_input = None): if skip_input is not None: x_input = Concatenate()([x_input, skip_input]) x = Conv2DTranspose(num_filters, ker, strides=2, 'leaky_relu') (x_input) return x </pre>
body	<pre> x, s1 = encoder_block(inputs, 48,3) x, s2 = encoder_block(x, 96,3) x, s3 = encoder_block(x, 192,3) x, s4 = encoder_block(x, 384,3) x, s5 = encoder_block(x, 768,3) x = Conv2D(1536,3, 'leaky_relu')(x) x = decoder_block(x,768,3) x = decoder_block(x,384,3, s5) x = decoder_block(x, 192,3,s4) x = decoder_block(x, 96,3,s3) x = decoder_block(x, 48,3,s2) x = Conv2D(24,5,'leaky_relu')(x) x = Conv2D(12,5,'leaky_relu')(x) x = Conv2D(6,5,'leaky_relu')(x) out = Conv2D(1,5,'sigmoid')(x) </pre>	<pre> x,out_ch = MSD_block(inputs ,1,[1,2]) x,out_ch = MSD_block(x,out_ch ,[3,4]) ... x,out_ch = MSD_block(x,out_ch ,[31,32]) out = Conv2D(1,3,'sigmoid')(x) </pre>	<pre> x, s1 = encoder_block_mod(inputs,3,48,[16,8,4,2]) x, s2 = encoder_block_mod(x ,3, 96,[10,5,3,1]) x, s3 = encoder_block_mod(x ,3, 192,[5,3,2,1]) x, s4 = encoder_block(x, 384 ,3) x, s5 = encoder_block(x, 768, 3) x = Conv2D(1536,3,'leaky_relu ')(x) x = decoder_block(x,768,3) x = decoder_block(x,384,3,s5) x = decoder_block(x,192,3,s4) x = decoder_block(x,96,3,s3) x = decoder_block(x,48,4,s2) x = Concatenate()([x, s1]) x = Conv2D(24,5,'leaky_relu')(x) x = Conv2D(12,5,'leaky_relu')(x) x = Conv2D(6,5,'leaky_relu')(x) out = Conv2D(1,3,'sigmoid')(x) </pre>
parameters	31,827,673	322,458	32,652,337

Table 4.1: Comparison of Unet, MSDNet, and Unet_mod components.

position, over the 150 diffraction patterns and the result was plotted as a function of the gap position. Fig. 4.6 shows the resulting curve that clearly highlights that the model performs better in regions with high intensity. This can be qualitatively explained: (i) central pixels contain larger spatial features both because of the nature of the Bragg peak, and because of

the lower noise level. This makes it easier for the model as it reduces the complexity of the prediction. (ii) As we move away from the center of the Bragg peak, the Signal to Noise Ratio (SNR) decreases, along with the *density of signal*. High accuracy scores in these regions would require the model to be able to predict noise correctly which is by definition impossible as it is an uncorrelated random process. One could argue that the accuracy curve would follow the statistical distribution of the gap positions inside the DL model training dataset. However, each mask has been applied at a position drawn from a discrete uniform probability function spanning the full data size, so can be excluded.

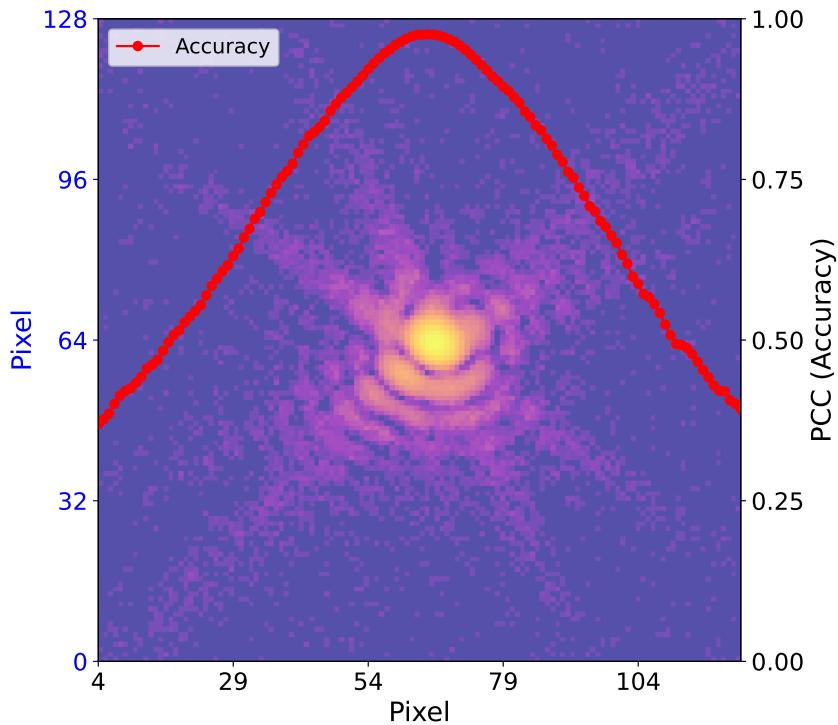


Figure 4.6: (Accuracy VS Gap position) Average Pearson Correlation Coefficient calculated over 150 9 pixel-wide vertical predicted gaps for each position of the gap inside the diffraction patterns. The model shows higher accuracies when the gap masks high intensity regions.

To conduct the second test, 150 diffraction patterns were simulated for the same particle varying gradually the oversampling ratio (see Sec.2.1) between 2 and 6. For each image a 9 pixel-wide vertical gap was then applied at all X positions and the DL prediction was computed. The accuracy scores have been averaged for each prediction in the same image and plotted against the oversampling (Fig. 4.7). As expected from the previous discussions, the model performs better for larger oversampling ratios, because of the bigger size of the features with respect to the gap width and because of the more uniform *density of signal*. It is worth clarifying that, for a given particle, the total amount of intensity in the diffraction patterns is in principle constant regardless of the oversampling ratio as it is fixed by Parseval's theorem¹. However, if the size of the dataset is kept fixed for different oversamplings, the effect is the same of a zoom lens that increases or reduces the field of view. Therefore, while for low oversampling ratios the full peak is recorded, for higher ones the peak is cropped, and less intensity is present in the image.

¹Interpreted also as an energy conservation law, Parseval's theorem states that $\int |f(x)|^2 dx = \int |\mathcal{F}\{f(x)\}|^2 dk$. In other words, the overall amount of signal is constant in direct space as in Fourier space.

This effect, coupled with the typical radial intensity decay of Bragg peaks and of Poisson noise, reduces the *density of signal* in largely oversampled BCDI patterns. Here the *density of signal* refers to the amount of information per pixel, which becomes smaller and more uniform. On the contrary, for low oversampling ratio the *density of signal* is less uniform as it is high inside bright regions (lot of information concentrated in few pixels) and low in noisy regions far from the peak. It follows that in order to properly assess the accuracy against the oversampling ratio one should consider diffraction patterns over the same extent in q -space, thus changing the size of the images. This more accurate evaluation was carried out for the 3D case and can be found in the next section.

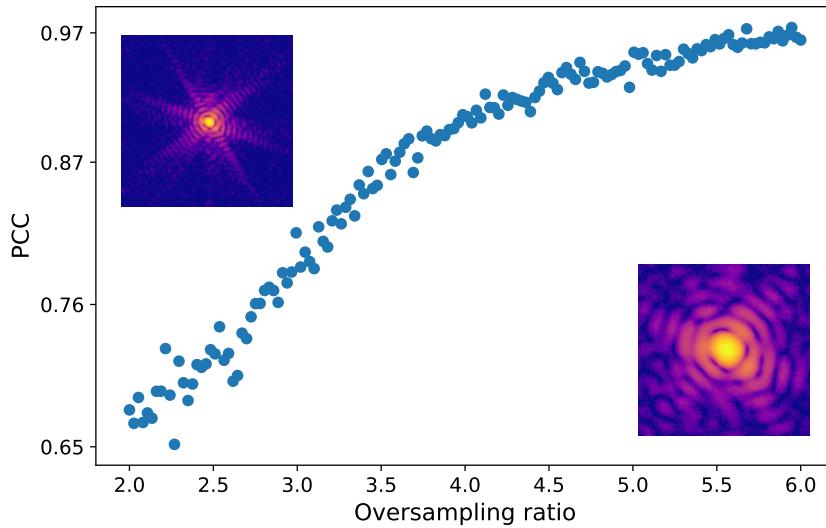


Figure 4.7: (Accuracy VS Oversampling ratio) Average Pearson Correlation Coefficient calculated over 280 9 pixel-wide vertical predicted gaps for each position of the gap inside the diffraction patterns. The model shows higher accuracies for high oversampling ratios.

4.4 3D case - Patching approach

When considering the 3D case, and especially the experimental conditions, there are a few practical issues that need to be overcome. In fact, experimental BCDI datasets that are more often affected by a detector's gaps are necessarily large datasets (e.g. $512\text{px} \times 512\text{px} \times N_{\text{rocking_steps}}$ where $N_{\text{rocking_steps}}$ is of the order of hundreds of pixels in typical BCDI experiments). Training a U-Net like model for 3D images of that size is overly expensive in terms of computing memory and time. Moreover, a common problem with this type of architectures is that the size of the images they can process is fixed by the first initialization. This means that one would need to resize, via binning or interpolation, the experimental datasets to the shape accepted by the DL model, and back to the original shape after the inpainting. Besides the impracticality, these operations are not recommended as they induce further modification and information loss to the original data. For these reasons a patching approach that loosens these constraints while preserving sufficiently high accuracies was considered.

The patching method exploits the regularity of the oscillations within BCDI datasets. The periodicity of the fringes in reciprocal space, peculiar property of this coherent diffraction

technique, follows from the oversampling condition as it is given a product of the Fourier transform of an object less than half of the Fourier window containing the diffraction pattern. It can be observed by eye and in many cases makes the prediction inside a gap region intuitively possible starting from just a few neighboring pixels. In our case we have decided to work with 32 pixel-sided cubic sub-volumes (patches from now on) cropped out of entire diffraction patterns. Among the “GPU-friendly” tensor sizes [107] we opted for 32 as good trade-off between amount of contained information and computing power required for training and inference. Moreover, this size is also greater than twice the size of the usual gap sizes and enough to catch several fringes in typical experimental oversampling conditions (< 10 usually).

4.4.1 Dataset creation

The training dataset consists of 50% patches from experimental data and 50% from simulated data. The experimental measurements were acquired at the ID01 beamline of the ESRF during different beamtimes on different particles. Namely, (i) Pt particles de-wetted on sapphire and YSZ (yttria–stabilized zirconia) with Winterbottom shape, measured under various temperatures and gas conditions, (ii) Pd and PdCe particles on glassy carbon, with Wulff shape, measured in an electrochemical environment following hydrogen loading. (iii) Ni particles on sapphire undergoing changes during CO_2 adsorption and (iv) “cubic $CaCO_3$ ” particles on glassy carbon. The synthetic diffraction patterns were instead simulated in three steps. The first step consisted in the creation of simulated 3D particles of different shapes (Winterbottom, Wulff, Cubic, Octahedral and random) using pre-existing scripts developed by Dr. Dupraz and Dr. Bellec [108]. These codes allow the user to construct a cubic FCC crystal of a given element, taking into account the inter-atomic potential, the atomic mass and the lattice parameter.

The final particle was finally obtained by "cutting" off atomic planes along given (or random) directions, depending on the chosen shape. Only Gold nano-particles were simulated, and this is, in first approximation, equivalent to any generic element as a different lattice parameter would just shift the Bragg peak to a different position in reciprocal space, with no significant alterations of the diffraction pattern itself. Each particle's configuration was then automatically saved in a LAMMPS-readable file. In a second stage, energy relaxation using LAMMPS software for Molecular Dynamics was performed [109]. This step induces small displacements to the perfect lattice, especially near the surface and the substrate. In the last stage, the 3D diffraction pattern of a selected Bragg reflection was computed using PyNX scattering package [49]. This software, optimized for GPU acceleration, produces a 3D representation of a selected Bragg peak using the kinematic sum. It was then possible to adjust the parameters that control the oversampling ratio, the size of the array in which the Bragg peak was centered and the rotation of the q -space. In this case, 128 pixel-size cubic diffraction patterns were simulated and, in order to augment the training dataset, various oversampling ratios (from 2 to 5) and different rotations for each particle were selected.

As we have seen in Chapter 1, in the kinematic scattering approximation the energy of the incident X-ray does not alter the diffraction pattern if not as a “zooming” factor. It follows that simulating diffraction patterns for different X-ray energies for a fixed pixel size and distance results in different oversampling ratios.

Before taking portions of these simulated BCDI patterns, Poisson noise was applied, randomly scaling the λ parameter for each image.

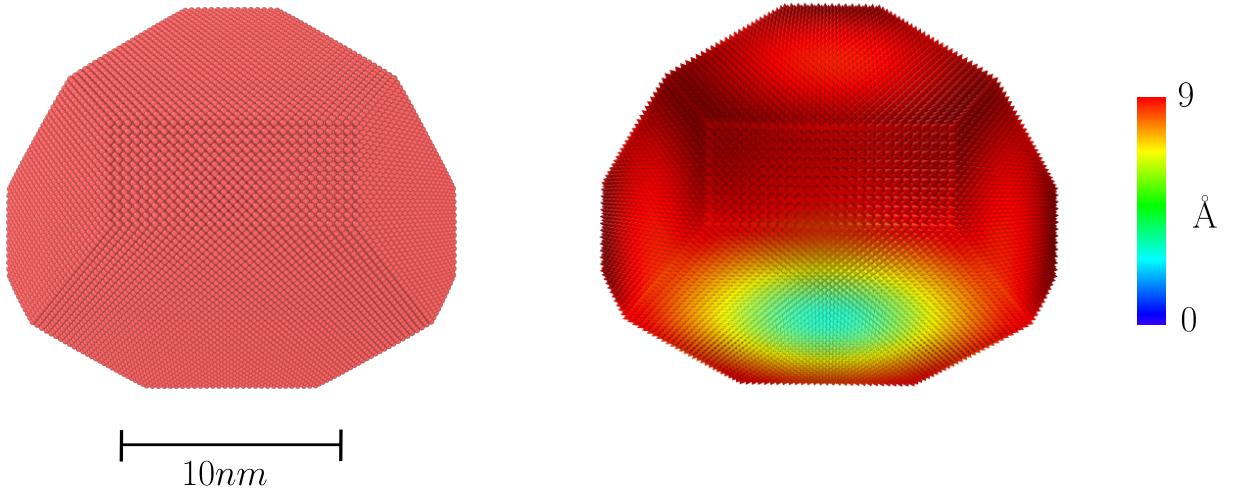


Figure 4.8: **Left:** Simulated Au particle with Winterbottom shape (134114 atoms). **Right:** Atomic displacement field of the same particle after LAMMPS energy relaxation showing the typical distribution at the interface with the substrate.

At this point, I proceeded with the extraction of patches taken at *pseudo-random* locations inside each 3D pattern. The selection in fact was not totally random as the extraction of patches from the outer regions, far from the center of the peak was favored. There are mainly two reasons for this choice, namely (i) compensate the inherent uneven accuracy score against the position of the gap (see Fig.4.6) by increasing the training data far from the center and (ii) emulate as much as possible the experimental conditions, in which unavoidable gaps are typically far from the center of the peak. For each sub-volume a 3D mask of the gap was created for different gap sizes (3,6,9,12 pixel-wide). The gap was placed vertically, in the center, along the third dimension, resulting in a “empty slab”. Notice that, while in practice a gap may obscure the diffraction pattern at any position, it is unnecessary to consider other placements within a patch, since one can always select a patch symmetrically around the gap. Cross-shaped gaps were also included in the training dataset, with a population ratio of 1:5 compared to vertical gaps. They were created by adding a horizontal gap at a random height to an existing vertical gap (Fig. 4.9). The final training dataset consisted of 30'000 $32 \times 32 \times 32$ pairs of patches, with gap and ground truth, created as described above.

4.5 3D model architecture

The DL architecture used for the 3D patching inpainting is illustrated in Fig. 4.10. Given the reduced size of the inputs, the encoder was composed of four blocks only, in each of which there were convolutional layers and max pooling layers. The feature map was thus reduced to a $2 \times 2 \times 2 \times 478$ tensor before being passed to the decoder. Notice that, as introduced above in section Sec.4.3, dilated convolutions were employed in the first two blocks to enhance the extraction long-range correlated features. After four decoder blocks three simple convolutional layers with 24,12 and 6 channels were used respectively in order to restore the possible smoothing effect of the decoder. As in the 2D model, the last activation function is a sigmoid that ensures the output to be in the range (0,1). The model contains 2'770'000 trainable parameters, significantly less than the 2D models working on full size patterns.

The training was performed loading batches of 32 images at the time over 100 epochs using

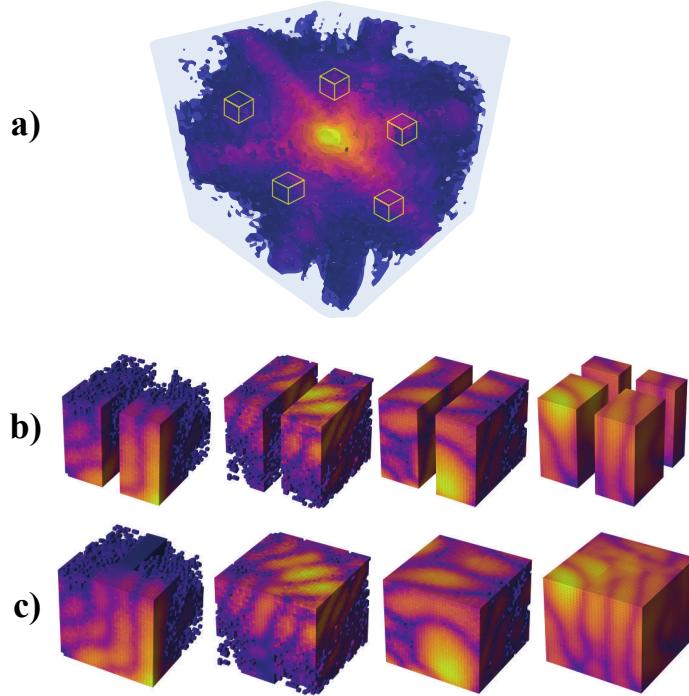


Figure 4.9: Schematic of the patches extraction. a) The 3D BCDI diffraction pattern and the patches. b) $32 \times 32 \times 32$ pixel-size patches with 9 pixel-wide vertical and cross-shaped gaps. c) Same patches with the DL inpainted gaps.

ADAM optimizer [110]. The optimizer was initialized with a learning rate of 10^{-3} and decreased it progressively with the ReduceLROn-Plateau callback feature available in Tensorflow. In order to exploit the maximum of the training dataset only 4% and 2.5% of the whole dataset was left for validation and testing respectively.

4.6 Results in detector space

In this section the results of the DL model on both simulated and experimental diffraction patterns are presented. The following section will focus on the results in real space and the reduction of the artifacts in the reconstructed objects.

Once completed the training, the model was first tested on portions taken from the test dataset. It is possible to qualitatively observe that the model works equally well for both simulated and experimental data (see Figs. 4.11 - 4.12). From a first visual assessment one can also confirm that low noise regions with larger features are better restored than others as previously stated in Sec. 4.7. Another curious effect that can be observed, is the “smoothing” of features around noisy areas (see first column in Fig. 4.11 and last column in Fig. 4.12). In fact, the “grainy” aspect of these regions is caused by Poisson noise which cannot be predicted by the DL model as it is uncorrelated. In those regions the DL performs therefore an averaging that “smoothens” the features and acts like a denoiser. This effect has been already studied in the literature and exploited for denoising applications like the Noise2Void model [111].

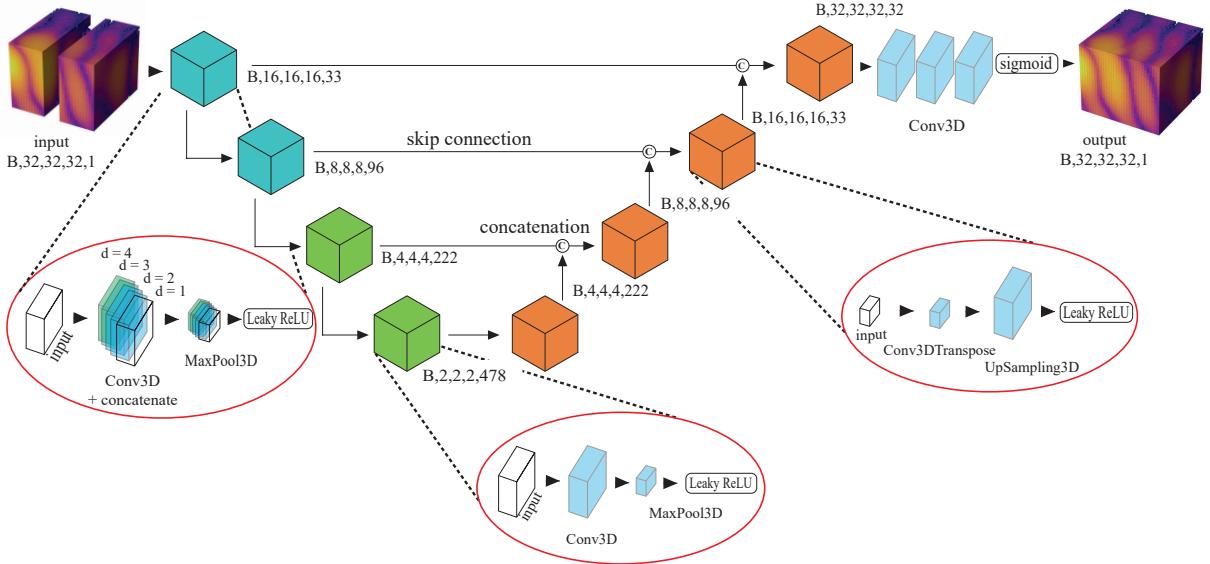


Figure 4.10: Schematic of the 3D model architecture The model uses a modified U-Net structure. In the first two encoder blocks (highlighted by the left red circle), dilated convolutions are applied where the original input is concatenated with its convolutions at various dilation rates ($d = 4, 3, 2, 1$) prior to the MaxPooling operation. The input consists of small gap-affected portions, grouped into batches of 32 (B). These portions (top left) are progressively processed by the encoder until they are reduced to a $2 \times 2 \times 2$ pixel-size feature map. In the decoder, each building block (represented as orange cubes) receives as input the concatenation of the output from the previous block and the matching output from the encoder block of the same size. The final result (top right) is a batch of inpainted versions of the input portions.

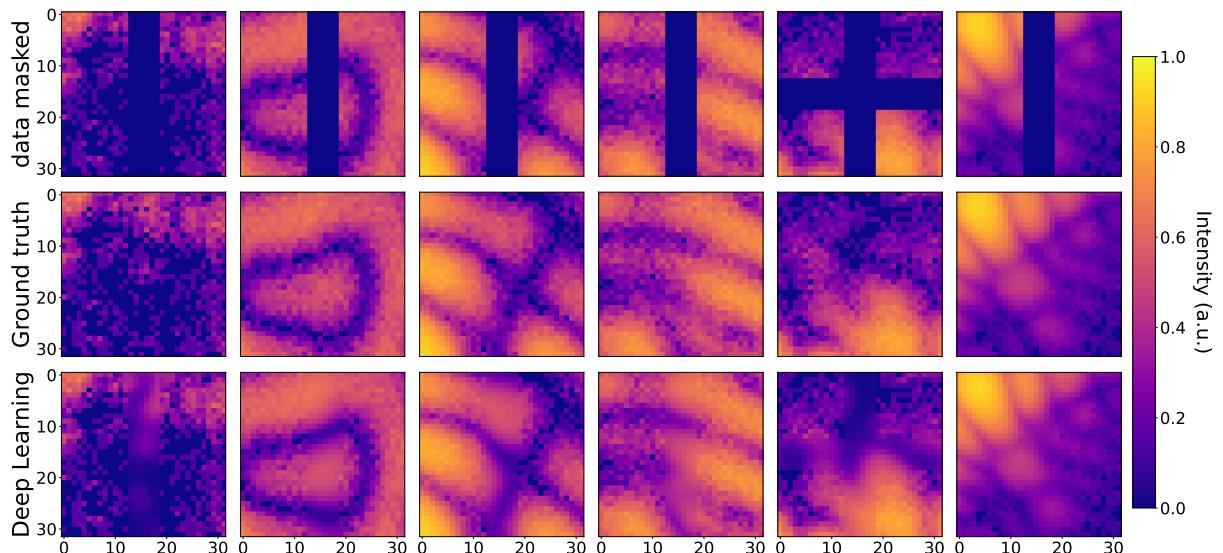


Figure 4.11: Results on portions of test simulated data. Central slices of portions taken from the simulated test dataset. Masked input with 6 pixel-wide gap in the first row, corresponding ground truth and DL inpainted in second and third row respectively.

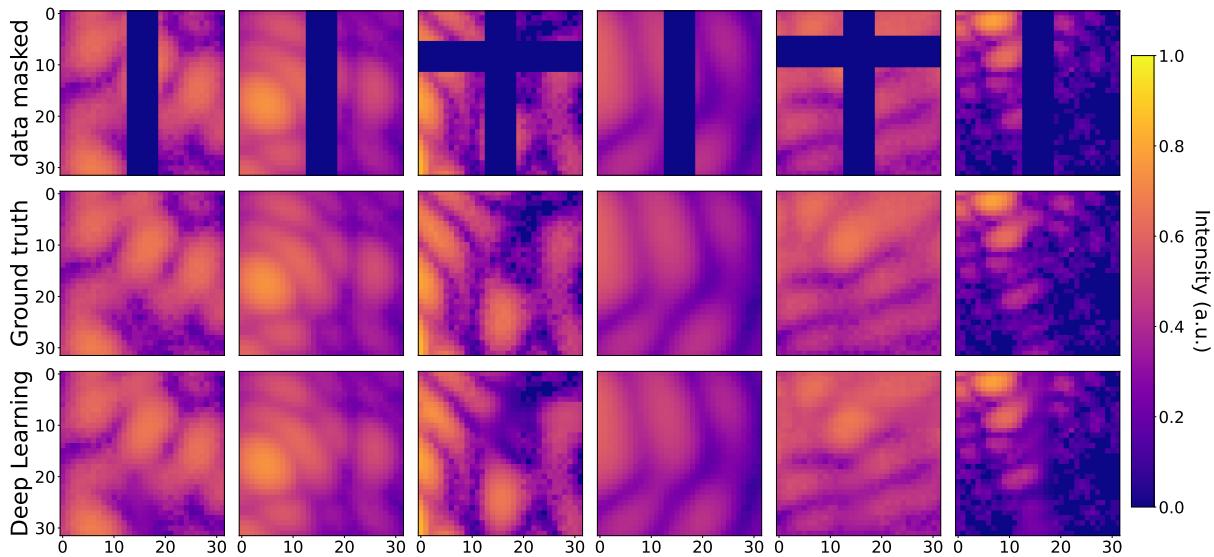


Figure 4.12: Results on portions of test experimental data. Central slices of portions taken from the experimental test dataset. Masked input with 6 pixel-wide gap in the first row, corresponding ground truth and DL inpainted in second and third row respectively.

4.6.1 Full gap inpainting

For the inpainting of a gap inside a full 3D BCDI pattern it is sufficient to apply repeatedly the DL model on sub-volumes cropped such that the gap plane lies vertical in the center of the array perpendicularly to the third dimension. Each sub-volume needs to be preprocessed exactly in the same way described above, i.e. transformed into logarithmic scale and normalized between 0 and 1. Moreover, it is advised to apply a mask on the gap, to match exactly the gap width the model has been trained with. One can then proceed along the gap moving forward one pixel at the time, compute the inpainted gap and average the prediction over the overlapping pixels with the previous predictions. By doing this, potential errors are averaged out and the accuracy of the prediction is maximized. However, for large datasets this can be time-consuming. For example, for a $128 \times 128 \times 128$ pixel-size diffraction pattern with a cross-shaped gap the time needed to compute the full inpainting amounts to 11 minutes (using a NVIDIA Tesla V100-SXM2 GPU with 32GB RAM). However, it is possible to increase the step size to significantly reduce the computing time without affecting excessively the accuracy (see Fig. 4.14). It was proven that the amount of time for the full inpainting follows a power law (see Fig. 4.13a) and the accuracy starts dropping significantly for more than 5 pixels skipped at the time (see Fig. 4.13b).

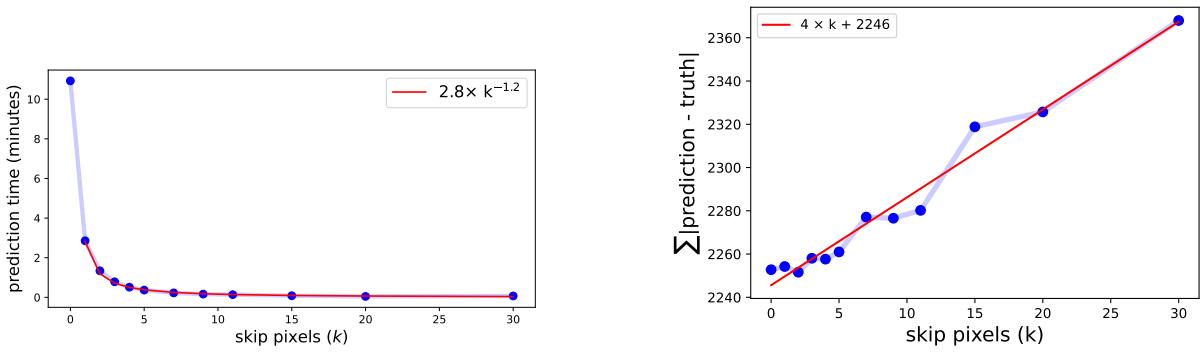


Figure 4.13: Left Full inpainting time for a 6 pixel-wide cross-shaped gap on a $128 \times 128 \times 128$ pixel-size diffraction pattern as function of the step size, i.e. the amount of pixels skipped between patch DL predictions along the gap. Right Sum of the absolute errors as function of the skipped pixels.

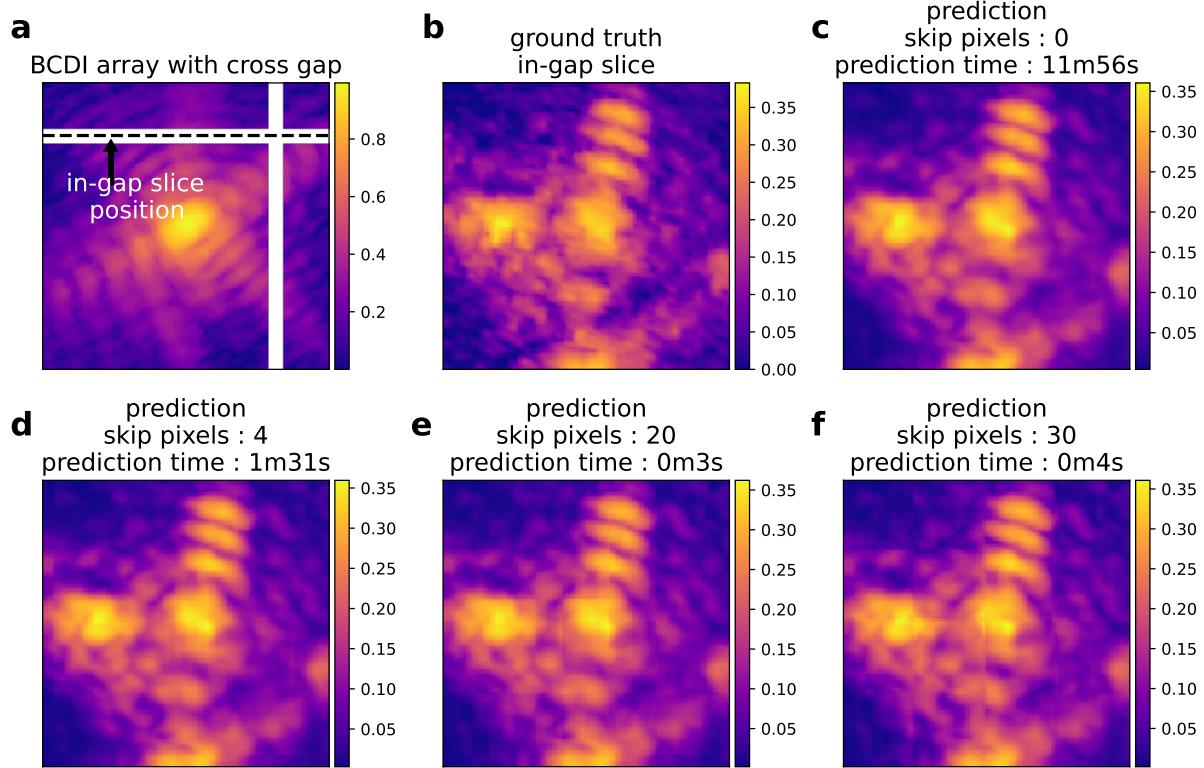


Figure 4.14: Full inpainting of an experimental BCDI pattern for different amounts of skipped pixels. a slice of the diffraction pattern perpendicular to the gap plane. b Ground truth intensity inside the gap. c-d-e-f In-gap prediction with 0, 4, 20 and 30 skipped pixels respectively, with corresponding execution time. Skipping 4 pixels is a good trade off between time and accuracy.

4.7 Performance assessment

In order to assess the DL model's performance with respect to other inpainting methods, it was tested against conventional interpolation methods. Specifically, an experimental BCIDI pattern with a 6 pixel-wide cross-shaped gap was considered and the inpainting results of the DL model with (i) linear interpolation (ii) cubic interpolation (iii) nearest-neighbor interpolation were compared. These techniques allow for a quick estimation of the intensity distribution inside the gaps but fail to recover fine features (see Figs. ??). In particular, it can be noticed in the *in-gap slice* (Fig. ??a) that linear interpolation for instance doesn't retrieve correctly the space curvature of the fringes while nearest neighbor and cubic interpolations show artifacts in correspondence of the perpendicular gap. When considering the central slice perpendicular to the gap planes (along the rocking curve dimension) one can notice even more how the DL model outperforms conventional interpolations (Fig. ??b).

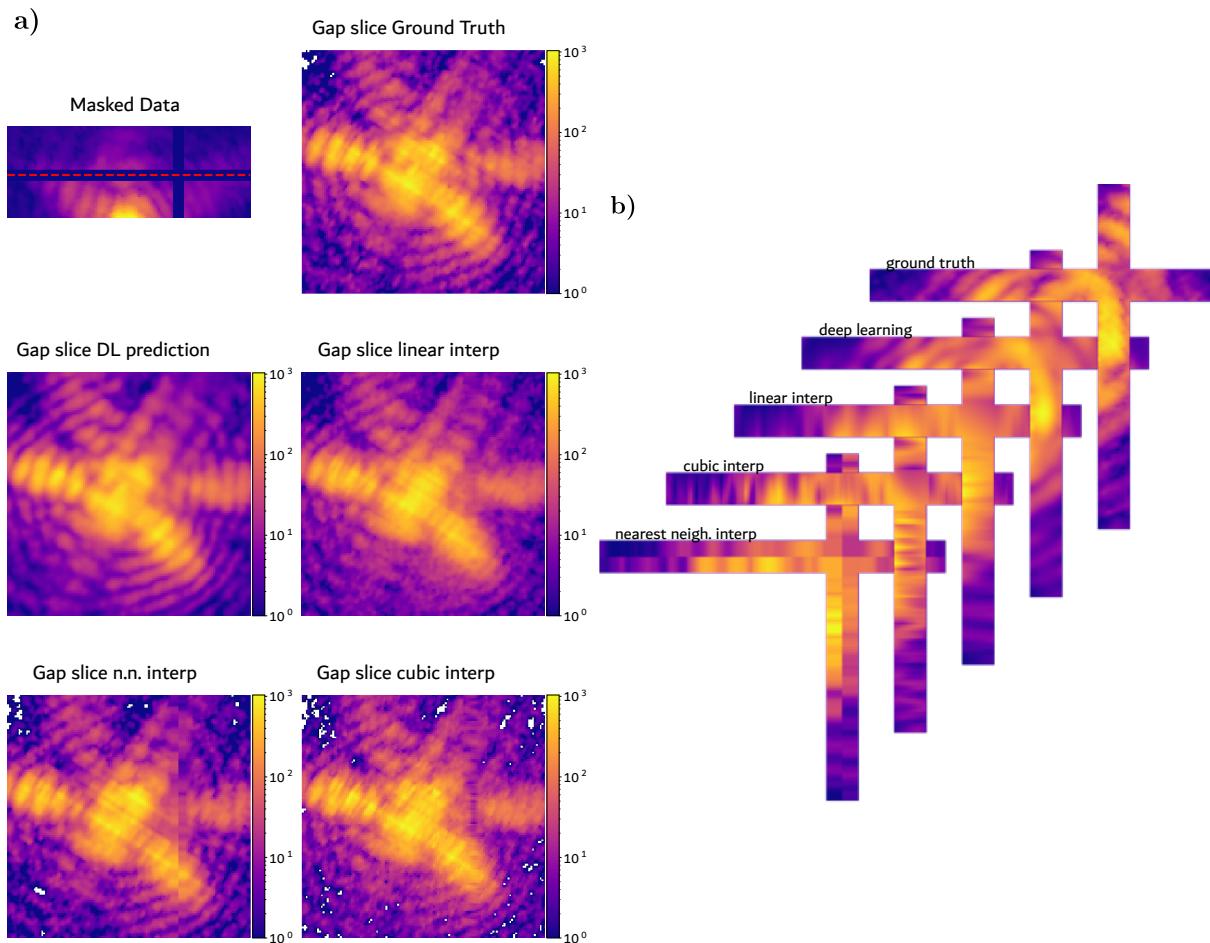


Figure 4.15: Comparison of DL inpainting and standard interpolations on experimental data: a) In-slice cut of the ground truth data and the predictions for each inpainting method. The DL outperforms standard interpolations, correctly recovering the fringes. The “blur” effect on the DL predicted slice is the result of the averaging of features in presence of noise, typical of CNNs [111]. **b)** Cross-shaped gaps for the same dataset (perpendicular view with respect to a)). The DL model outperforms other methods from this perspective as well.

Similarly to the 2D case above, the performance of the model has been evaluated against the amount of intensity inside the sub-volume and against the oversampling ratio. The test was repeated for different gap widths, namely 3,6,9,12 pixel-wide, using vertical gaps placed in the center of each patch. For the first performance assessment a full simulated $128 \times 128 \times 128$ pixel-size BCDI pattern was considered, and 1000 patches were randomly cropped out of it. A vertical gap was then applied in the center of each patch for different gap sizes and then computed the prediction with the corresponding DL model. The intensity (in photon counts) inside each patch was then summed and the obtained values for the 1000 samples were binned into 20 classes for better visualization. The accuracy scores, calculated with the PCC, were then averaged inside each bin class. The results are displayed in Fig. 4.16. As expected from the 2D case, better accuracy scores are obtained for portions containing larger amount of signals, where noise levels are lower and the features of the diffraction pattern are more visible. Moreover, the plot logically shows that smaller gaps are generally better recovered, but it is worth noticing that the accuracy spread across different gap sizes widens for noisy portions and narrows down as the amount of signal increases. These trends suggest that DL models are overall robust to different gap sizes especially for high intensity regions, which are the most important ones as they contribute the most during the reconstruction.

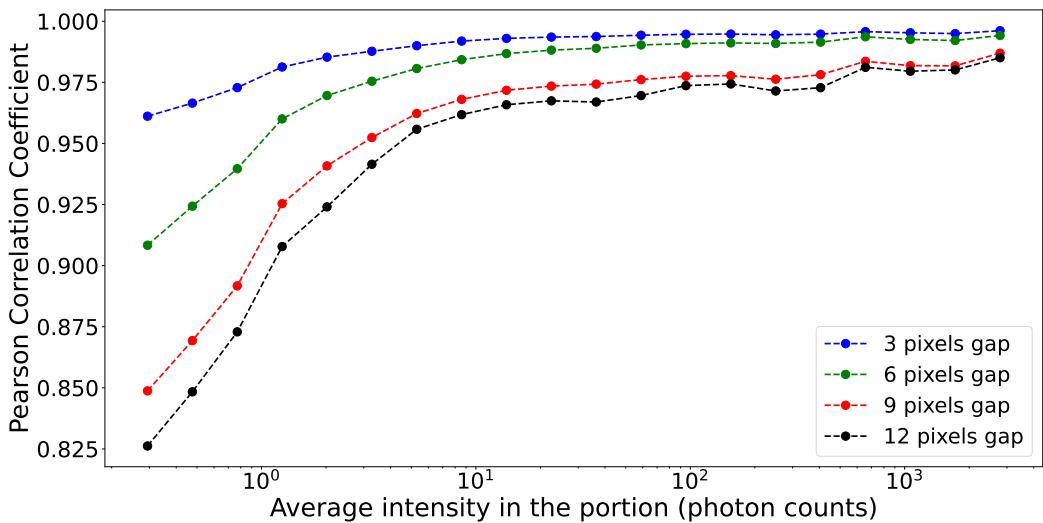


Figure 4.16: Accuracy scores (PCC) of the DL patching model versus the amount of signal of the patch for the four different gap sizes. As expected, the model perform better on smaller gaps and high SNR. Surprisingly, for high-signal regions the accuracy scores for large gaps approach the ones for smaller gaps.

The last test concerns the study of the accuracy for different oversampling ratios. As anticipated above for the 2D case, to carry out properly this evaluation, one should consider the same diffraction pattern extending to the same equivalent q -space value for each oversampling ratio. This in practice is done increasing the dq per pixel as decreasing the oversampling ratio, results in a smaller size of the overall BCDI pattern. In this case the same BCDI pattern was simulated for oversampling ratios spanning from 2 to 7. For each oversampling ratio, a vertical gap mask was applied to the whole BCDI array and the DL prediction was calculated with no-skip pixel (see Sec. 4.6.1). The gap was then shifted laterally, and this procedure was

repeated until the whole BCDI array was predicted using our model, thus leading to a full BCDI predicted image. The PCC was then calculated using the whole BCDI array for different oversampling ratios and model gap sizes. The results are displayed in Fig. 4.17. As expected, the predictions are more accurate for large oversampling ratios and small gap sizes (i.e., large oscillation periods relative to the gap width).

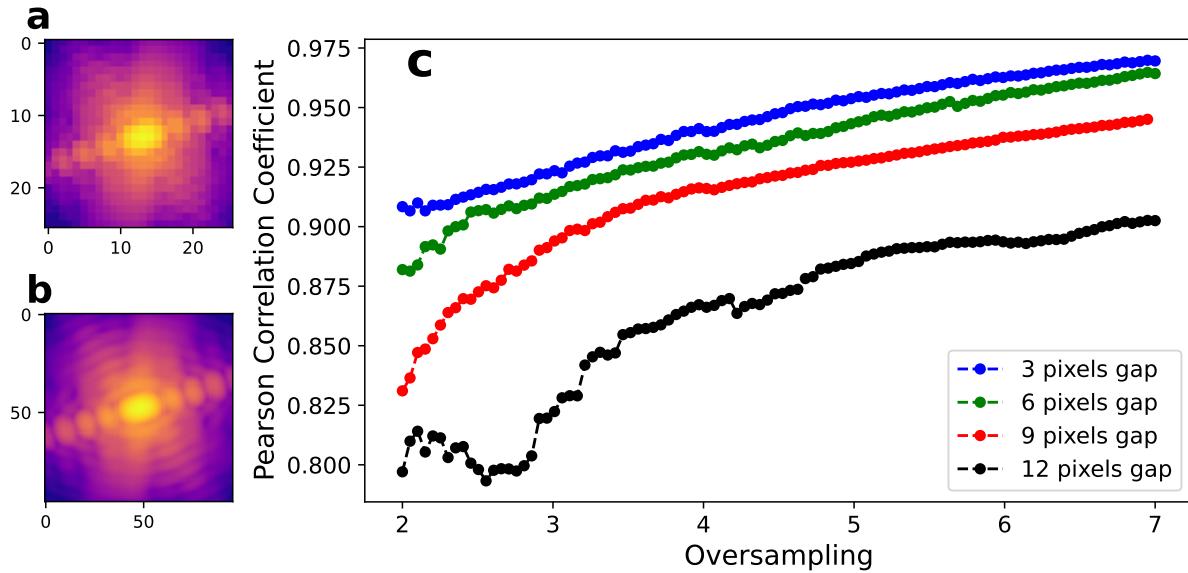


Figure 4.17: Accuracy scores (PCC) of the DL patching model against the oversampling ratio. **a)** Simulated diffraction pattern with low oversampling ratio. **b)** The same pattern simulated on a larger grid for an increased oversampling ratio. **c)** Pearson Correlation Coefficient (PCC) of the full inpainted diffraction pattern for the four different gap sizes considered. The trend overall confirms the expectations.

4.8 Results in real space

In this section the effects of DL inpainting on the reconstructed objects will be discussed for both simulated and experimental data. In particular, it will be presented the assessment, both qualitatively and quantitatively, of the gap induced artifacts in the modulus, phase and strain fields of the reconstructions and their reduction thanks to the DL inpainting. To carry out these analyses an experimental BCDI dataset acquired at ID01 was considered. The same dataset had already been exploited by Carnis *et al.* in 2019 for similar studies on gap-induced artifacts [85]. The dataset corresponds to the BCDI pattern around the (111) peak of a Pt particle (400 nm in size). Similarly to what the authors did, I have kept the modulus of the reconstructed object and set the real space phase to zero, making it our reference ground-truth object \mathbf{O} . This measure helps to highlight the gap induced artifacts of the phase and strain fields as there is zero-phase reference to compare the results with. The corresponding diffraction pattern was then calculated with the Discrete Fourier transform (DFT) obtaining a complex diffracted amplitude $\mathbf{A} = \mathcal{F}(\mathbf{O})$. A cross-shaped gap was subsequently applied to \mathbf{A} and the corresponding object \mathbf{O}_{gap} was calculated with the inverse DFT. From the same gapped \mathbf{A} , the intensity $\mathbf{I} = |\mathbf{A}|^2$ was also “inpainted” using our DL model and corresponding object \mathbf{O}_{DL} was calculated with the inverse DFT as well, using the ground truth reciprocal space phase.

The procedure was repeated for four different gap sizes (3, 6, 9, 12 px-wide) matching exactly the cases mentioned in the work of Carnis and coauthors. Figure 4.18 shows the projection along the rocking curve axis (XY slice in this case) of the ground truth, the gapped and the DL inpainted diffracted intensity for the 9 pixel-wide gap case.

Notice that in all cases of Fig 4.18, there are vertical and horizontal lines of higher intensity not attributable to the typical streaks from the truncation rods. These arise from the detector gaps in the original dataset, which are in fact responsible for the oscillatory stripe artifacts visible on the object's modulus for all three cases in Fig. 4.19, ground truth included. The DL inpainting of these original gaps is presented later in Sec. 4.8.1.

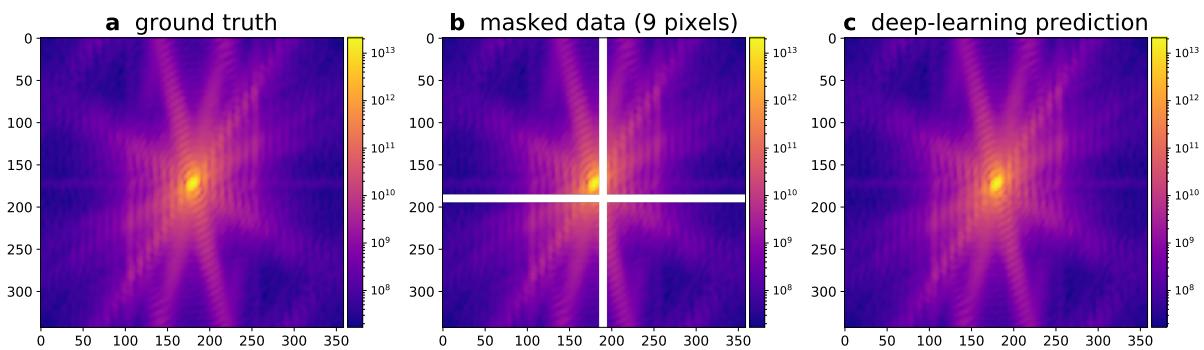


Figure 4.18: Projections along the rocking curve axis of the studied diffraction pattern in log scale. **a** Ground truth pattern obtained from the $|\mathcal{F}(\mathbf{O})|^2$. **b** Pattern with a 9 pixel-wide cross shaped gap. The position close to the center of the peak is experimentally unlikely but here it allows us to enhance the artifacts in the reconstructions. **c** Corresponding DL inpainted BCDI pattern. The presence of aliasing can be seen due to the DFT calculation as compared to the correct kinematic sum. This effect is however not relevant for the scope of these analyses.

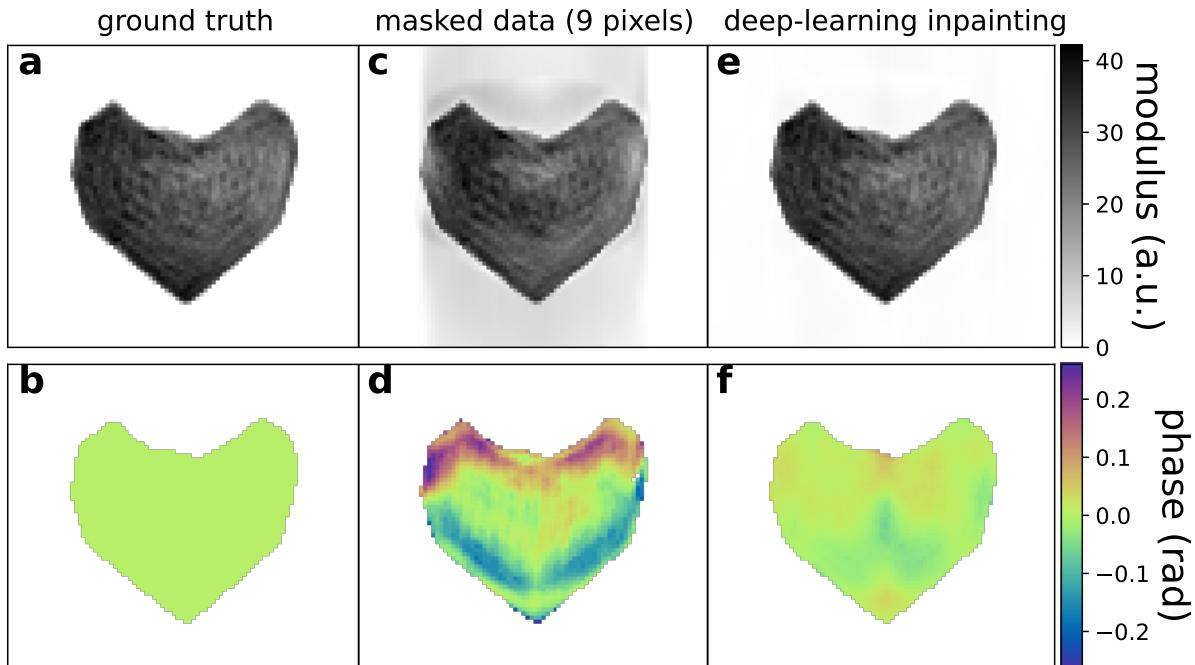


Figure 4.19: Reconstructed objects (central slice). a-b XY central slice of ground truth modulus and phase obtained from the reconstruction of an experimental gap-less dataset. The object's phase has been set to zero artificially after the reconstruction for better assessment of the gap-induced artifacts. c-d Modulus and phase of \mathbf{O}_{gap} , the object obtained from the inverse DFT of the diffraction pattern (with reconstructed reciprocal space phase) with a 9 px-wide cross-shaped gap placed as shown in Fig.4.18. e-f Modulus and phase of \mathbf{O}_{DL} , the object reconstructed from the DL-inpainted diffraction pattern. The phase of this last object shows reduced artifacts with respect to d.

Figure 4.19 illustrates instead the central slice (XY) of the reconstructed objects for the three cases. It is evident that while \mathbf{O}_{gap} shows significant abnormalities in both modulus and phase, \mathbf{O}_{DL} is much closer to the ground truth. In particular one can notice that the gap plane, horizontal in the XY plane, induces artifacts along its perpendicular direction. The result is indeed a stripe of non-zero modulus outside the support and, most importantly, an overall phase variation of 0.4 radians along the vertical direction. This phase variation results in an error of ± 7 pm in the lattice displacement field for the 111 Pt reflection, with more intensity around the surface. These artifacts are particularly problematic in the cases of (electro-)catalytic experiments [21] or in situ gas experiments [112–116], where the particle's surface is primarily involved in the reaction and one could follow the process by monitoring the evolution of the strain in that region. As one could expect the artifacts become more severe as the gap size increases. Fig.4.20 depicts the phases of \mathbf{O}_{gap} and \mathbf{O}_{DL} for the different gap sizes considered in this study while Fig.4.21 the strain distribution in the XY plane.

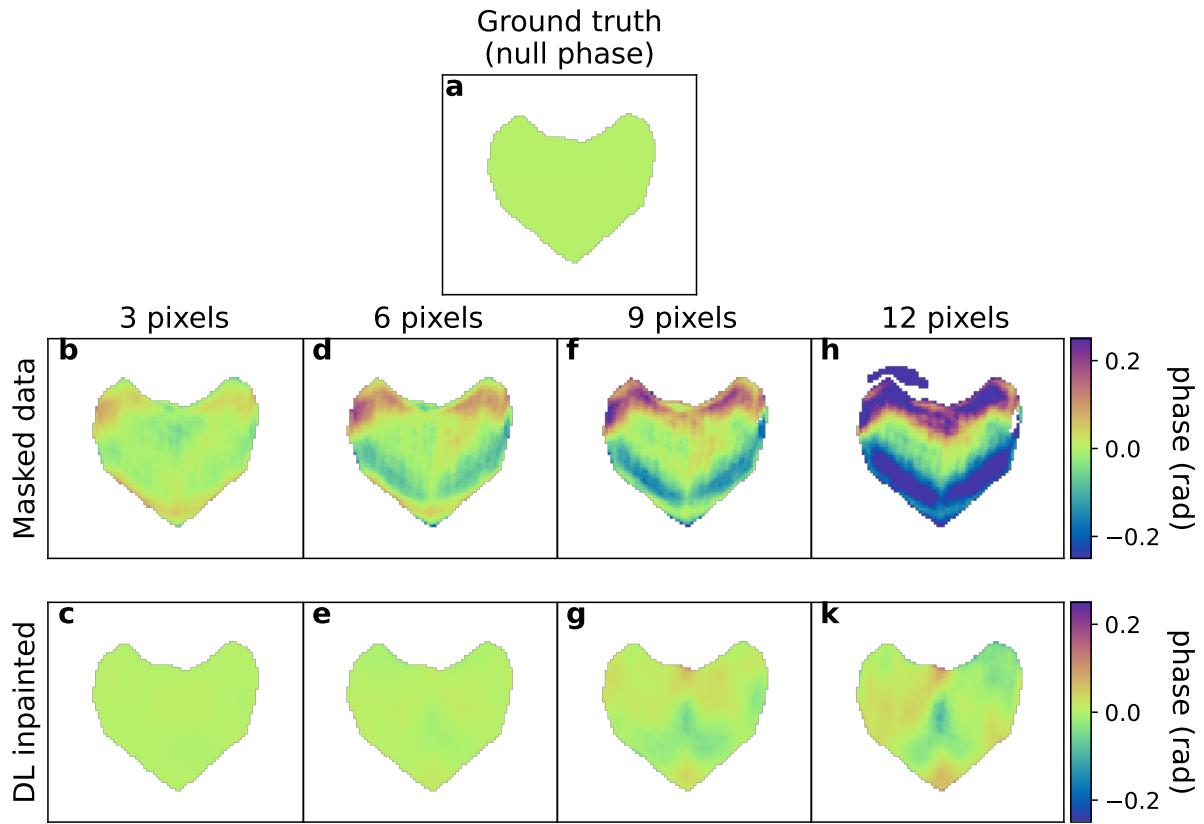


Figure 4.20: Comparison of the phase artifacts vs gap size. XY central slice of the phase of \mathbf{O}_{gap} for different gap sizes (b-d-f-h), and phase of the corresponding \mathbf{O}_{DL} (c-e-g-k). The comparison with the ground truth null phase (a) shows a significant increment of the artifacts in \mathbf{O}_{gap} with the gap size. On the contrary, in \mathbf{O}_{DL} artifacts are severely reduced for all gap sizes.

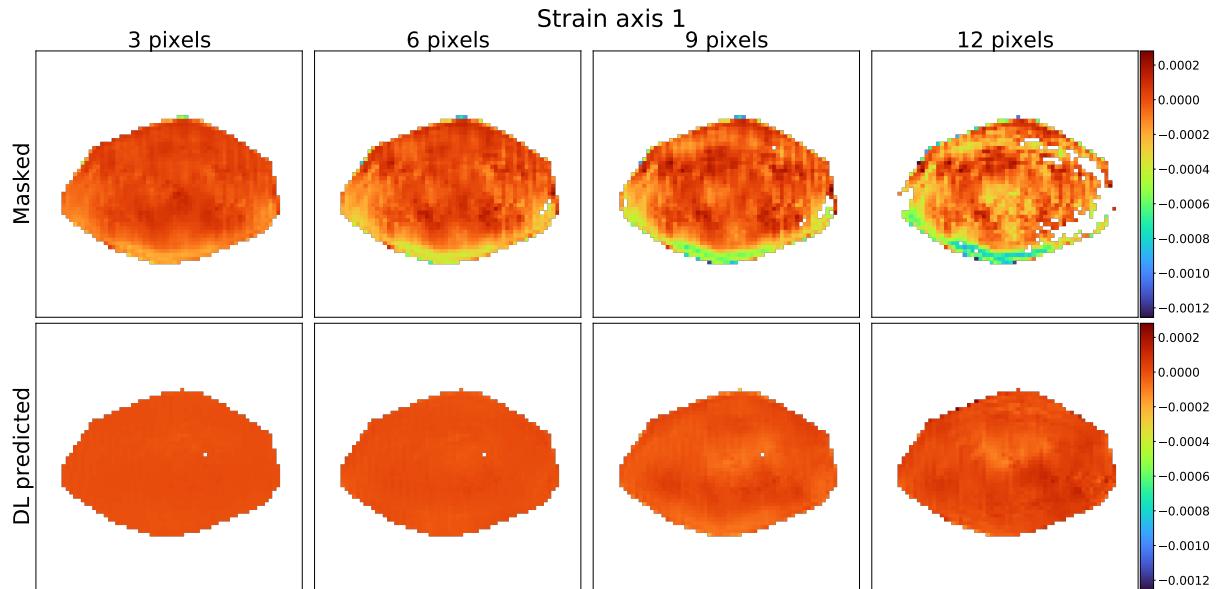


Figure 4.21: Comparison of the strain vs gap size. XZ central slice of the strain distribution for of \mathbf{O}_{gap} (first row) and \mathbf{O}_{DL} (second row) for different gap sizes.

The deviation from the ground truth zero value of the retrieved strain for both \mathbf{O}_{gap} and \mathbf{O}_{DL} can be measured with the root mean squared error (RMSE) across all the different gap sizes. This calculation was already proposed in the aforementioned work of Carnis and coauthors in which the results were plotted in Fig.4. We have reproduced a similar figure adding the results of our DL model (Fig.4.22). The trend of the strain RMSE resembles the curve shown in [85], increasing significantly with the gap size while the DL equivalent curve lies below, dampening the error by approximately a factor 5. Moreover, the strain artifacts induced by the gaps shift the average strain from the zero value as shown in Fig.?? whereas our DL model maintains the average strain around zero.

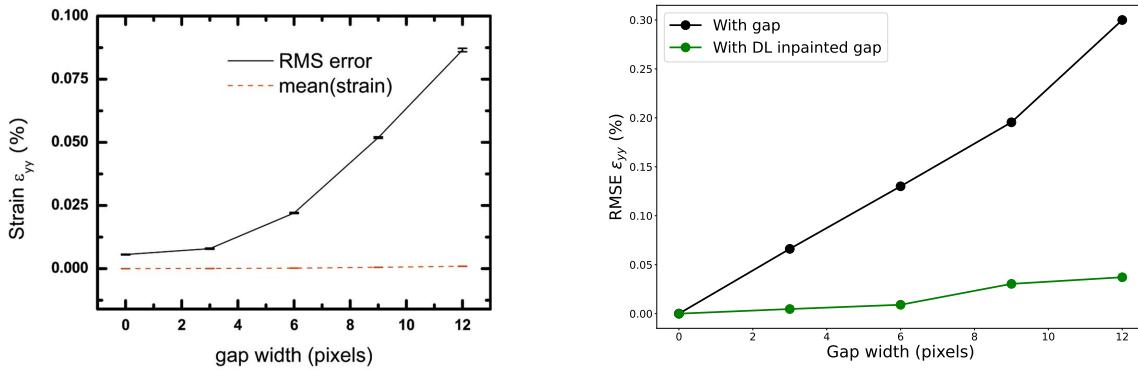


Figure 4.22: RMSE of the strain field versus gap size(Left) The plot realized by Carnis *et al.* in [85] depicts the RMSE of the ε_{yy} strain component for the objects reconstructed from gapped diffraction patterns with different gap-size. The mean strain around zero corresponds to the null object's phase set artificially. **(Right)** Plot of the RMSE of the ε_{yy} strain component versus same gap-size as **(Left)** for gapped (blue line) and DL inpainted (orange line) diffraction patterns.

4.8.1 DL inpainting for high resolution BCDI

As anticipated in the introduction to the chapter, the most prominent case in which a BCDI datasets is affected by a detector gap is a “high-resolution” dataset. The acquired data here extend for large q ranges in all directions resulting in parts of the diffracted signal that cross a region on the detector with a vertical or horizontal gap, hence the need for gap-inpainting. All the more so, it is convenient to use a patching approach since treating the full volume would be computationally too expensive. Moreover, any binning or interpolation to smaller sizes will induce information loss, and is not advised.

An example of high-resolution BCDI dataset of this type is the one we have used so far from the work of Carnis and coauthors. The original dataset is indeed a large ($256 \times 300 \times 300$ pixels) array that contains a cross-shaped, 6 pixel-wide gap. Here we show how the artifacts can change depending on the type of masking of the gaps is chosen during the phasing and how our DL model can outperform these methods. A common approach, when using PyNX software, is to mask the gap such that those pixels don't contribute during the phasing and are left free to evolve (a). Moreover, one could mask only near the intensity streaks affected by the gap (b) or simply leave the gap with zeros and remove the contribution of the gap voxels during the whole phasing (c). These strategies have been used during the phasing of the

cited Pt diffraction pattern and the results in object space compared with that obtained from the DL inpainted pattern. The results, illustrated in Figs. 4.23 - 4.24, show that the amount of oscillatory artifacts progressively decreases as we go from method **a** to **c**, proving the DL inpainting to be the optimal method among them.

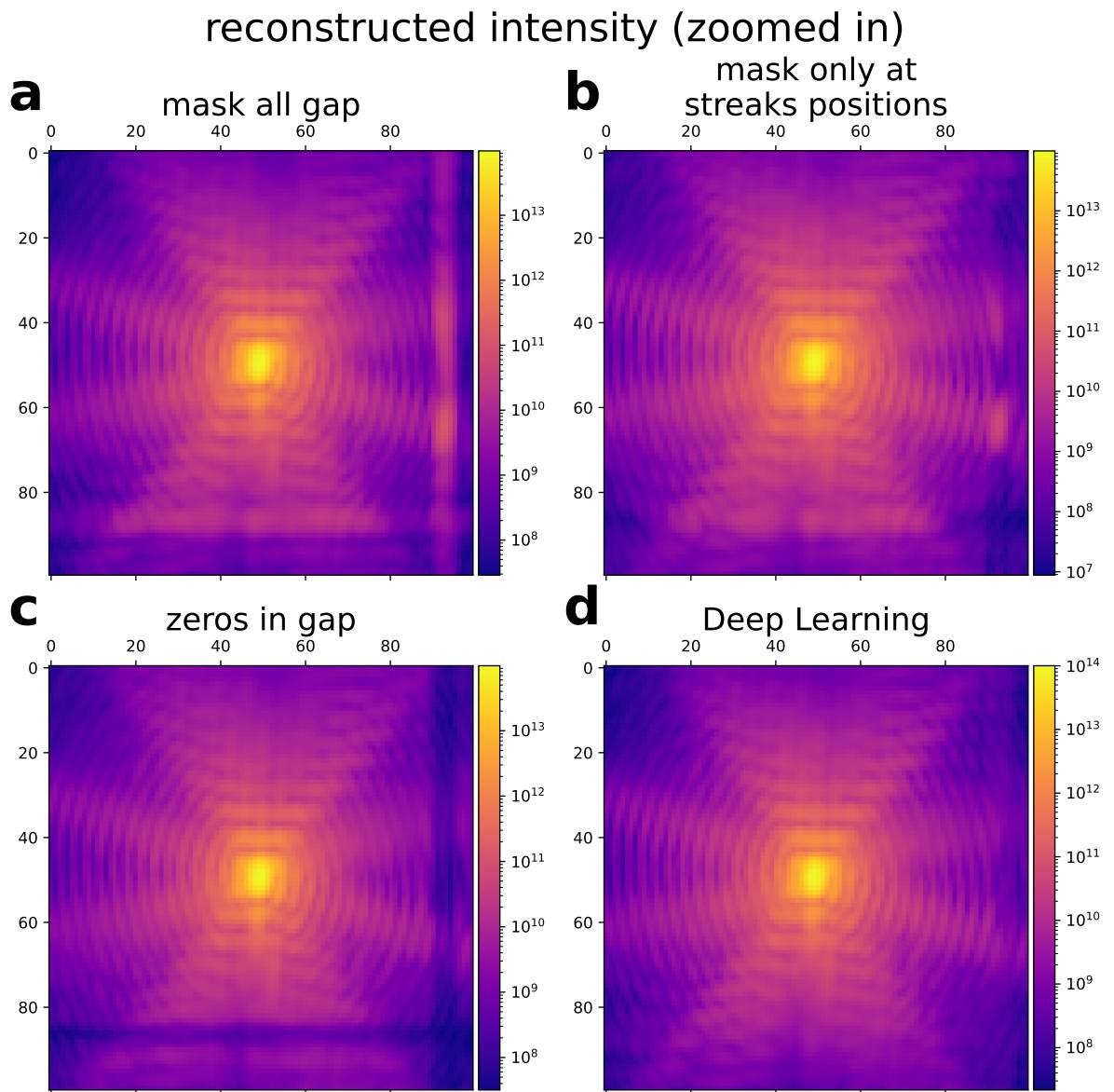


Figure 4.23: Zoom on the projection along the rocking curve axis of the Pt BCDI pattern in Fig.4.18 calculated from the reconstructed object obtained with PyNX software using **(a)** a mask on the gaps, **(b)** a mask on the streaks only, leaving zeros inside the gaps **(c)** and inpainting the gaps with the DL model **(d)**. Inside the regions where the mask was applied the intensity is free to vary throughout the PR iterations.

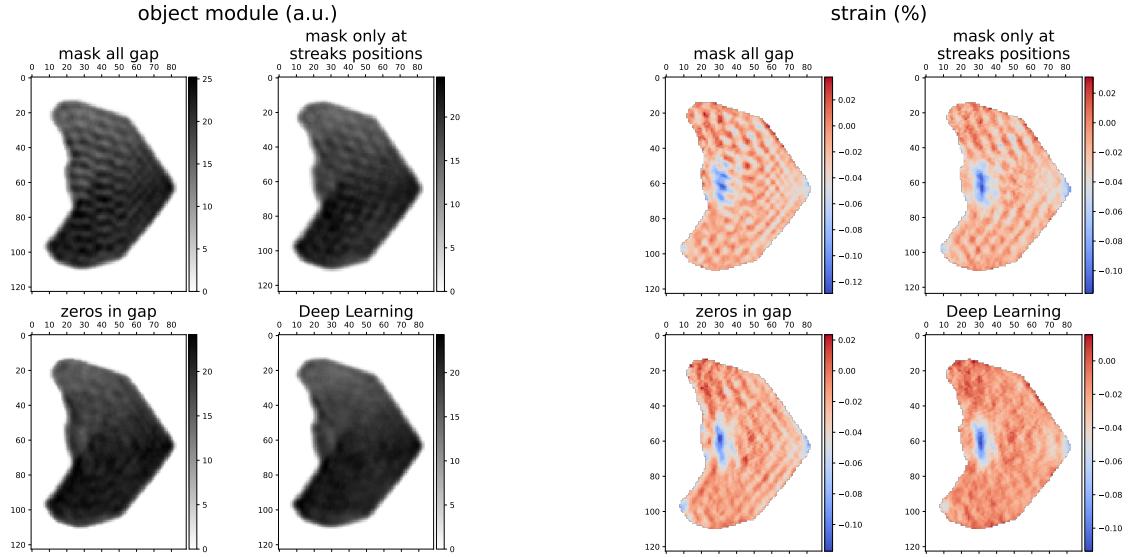


Figure 4.24: Modulus (Left) and strain (Right) of the object’s reconstructions for each case mentioned above in Fig. 4.23. The oscillatory artifacts are smallest for the object obtained after DL inpainting (bottom right corner on both left and right images).

4.9 Fine-tuning

For those cases in which the DL model does not yield satisfactory results when inpainting a new experimental BCDI pattern a fine-tuning of the model was conceived in order to improve the accuracy of the prediction. This fine-tuning is enabled by the patching approach as it consists of a secondary short training of the general model on a small dataset made of portions extracted from the new BCDI pattern to be inpainted. In particular, after loading the gap affected BCDI pattern, 2000 portions were randomly cropped out of it, paying attention not to include the gap region. The model has been trained for the corresponding gap width for 2 epochs with a learning rate of 10^{-5} and batches of 32 images (~ 2 mins on a NVIDIA Tesla V100-SXM2 GPU with 32GB RAM). Biasing the model to the fit the features of that specific diffraction pattern (oversampling ratio, particle shape, noise level, fringes shape) better results can be obtained on the real gap. An example is shown in Fig. 4.9. There, the general DL model was not able to predict the fringes with the correct periodicity inside the gap. After the fine-tuning instead, the model properly recovers the fringes improving the accuracy. This fine-tuning technique is a further example of the advantage of using a patching approach and its usage depends on the user judgement on the quality of the general model inpainting.

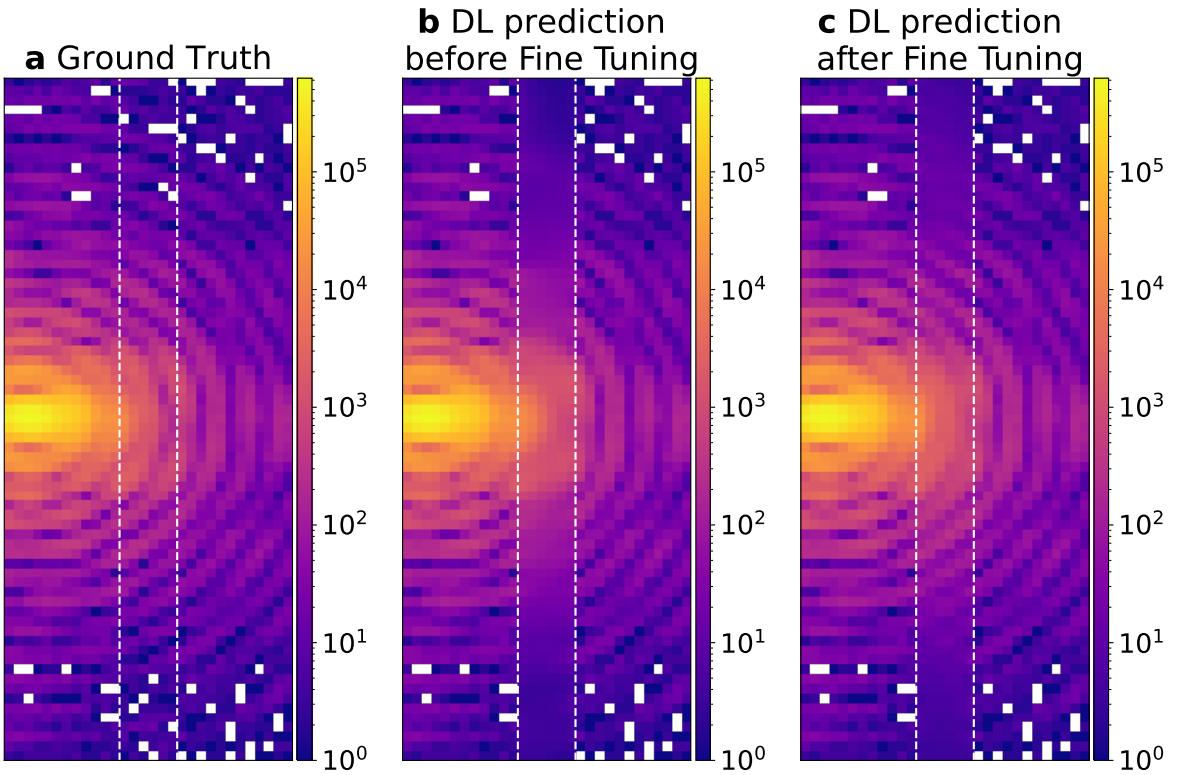


Figure 4.25: Example of improved accuracy after fine-tuning of the DL model on experimental data. **a)** The ground truth portion of the central slice of the BCDI experimental diffraction pattern. The dashed line shows the position of the artificial 9 px-wide vertical gap applied for this test. **b)** Same slice for the DL inpainted pattern *before* fine-tuning. The fringes' periodicity is not recovered correctly. **c)** Results *after* fine-tuning. The fringes are now better recovered after 2 epochs of fine-tuning on a dataset of 6400 patches cropped out of the same diffraction pattern (except the masked region). The fine-tuning training took 110 seconds.

The effect of the fine-tuning can also be assessed on the reconstructed object. Here, the diffraction patterns showed in Fig.4.25 has been phased with PyNX and the results are compared in Fig.4.26. For the phase retrieval, in all the three cases a single run of 400 HI0 + 1000 RAAR + 300 ER iterations has been performed starting from the autocorrelation of the object and with the shrinkwrap threshold of 0.2. As expected, already from visual inspection, the modulus and phase of the object reconstructed from BCDI pattern inpainted after the fine-tuning are more similar to the ground truth ones. On the contrary, the one obtained before the fine-tuning shows some oscillatory artifacts caused by the not correctly restored fringes.

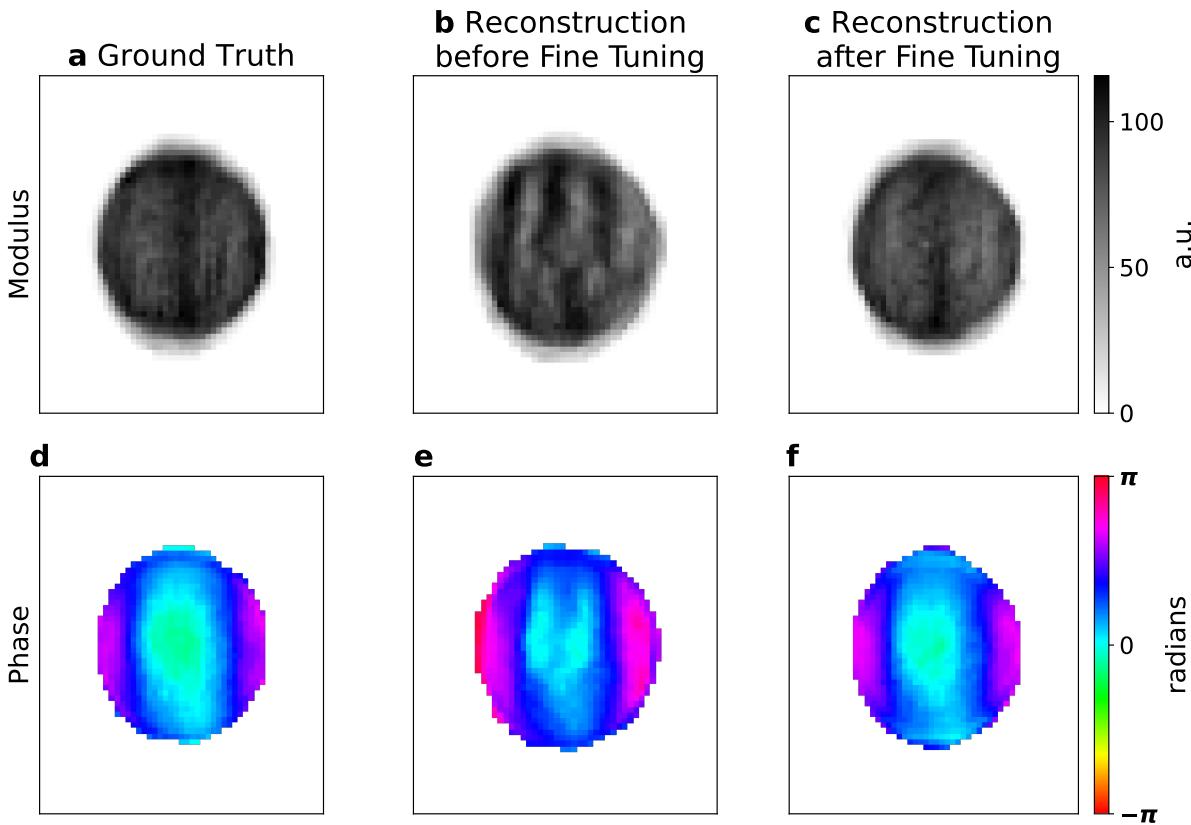


Figure 4.26: Reconstructed object before and after fine-tuning of the DL model From the diffraction pattern in Fig. 4.25, **a - d** Modulus and phase of the ground truth object. **b - e** and **c - f** modulus and phase of the object reconstructed from the DL inpainted diffraction pattern *before* and *after* fine-tuning respectively. The not well recovered fringes of Fig. 4.25**b** produce artifacts in the reconstructed object that are eliminated with the inpainting of the fine-tuned DL model.

4.10 Conclusions

A novel DL model for the inpainting of BCDI patterns with detectors' gaps has been presented and tested for different experimental circumstances. The “patching approach” has proven to be adequate for BCDI for different reasons here summarized.

- Due to the oversampling condition, the BCDI patterns exhibit spatially regular and well-separated oscillations. This local “redundancy” in detector space can be exploited by isolating smaller sub-volumes of the diffraction pattern (*patches*). In the presence of gaps, the missing signal within each patch can then be reconstructed from neighboring voxels, with the reconstruction accuracy depending on the relative size of the gap compared to the patch.
- The patching approach naturally allows for larger training datasets, faster training compared to full-image direct inpainting approaches. Moreover, no lossy resizing is needed to adapt the whole BCDI pattern to the DL model size, as in full-image direct inpainting.

- DL inpainted experimental BCDI patterns have shown reduced artifacts in the reconstructed objects for different gap-sizes, thus confirming improved reliability of the reconstructions.
- Unlike full-image direct inpainting models the patching approach enables a model fine-tuning on each new specific gap-affected dataset to be restored. This additional training can significantly improve the inpainting accuracy in trade of additional computational time of the order of few minutes.

The presented DL model is planned to be implemented in the PyNX package in the next future, such that it can be employed by a broader BCDI user community in standard data analysis pipelines. In these regards, it is worth mentioning that for an unbiased reconstruction, during the last iterations PR iterations, the inpainted pixels would need to be “freed” with a proper mask. In such a way, when the PR algorithm is near convergence, the data-fidelity is preserved.

Future perspective can envision the extension of this patching approach to other CDI techniques. In the X-ray imaging community the detector gaps are in fact a common problem, as well as other limiting elements (beam-stops, defective pixels, overexposure, etc.). Given high-intensity signal covered by beam-stops and the higher accuracy of the DL model on bright patches, one could infer that CDI techniques like small-angle CDI and others in forward geometry could benefit from the presented DL inpainting approach.

CHAPTER 5

DEEP LEARNING FOR PHASE RETRIEVAL

We enter now the core topic of the thesis. Most of the efforts during this PhD have been dedicated to the study of the Phase Problem for Bragg Coherent Diffraction Imaging using DL based approaches. Here I will discuss the main steps of this journey, starting off from the analysis of the most relevant works in literature and concluding with some comments on the final version of a DL model for highly strained particles. The latter has become the subject of an article, currently submitted, entitled “*Phase Retrieval of Highly Strained Bragg Coherent Diffraction Patterns with Supervised Convolutional Neural Network*” [117]. The process that led to the final version of the model will be unraveled, and particular attention will be given to elucidating the key steps and the critical issues encountered along the way. The structure of the chapter can be summarized as follows:

- **State of the art and Reciprocal space phase phasing:** introduces the works on DL for phase retrieval in BCDI and discusses the novel concept of the prediction, using DL, of the reciprocal space phase (RSP) lost during the measurement.
- **2D case and Weighted Coherent Average loss function:** presents the preliminary studies on the 2D case, for low-strain and high-strain simulated particles. Introduces the novel “Weighted Coherent Average” loss function, designed specifically for the prediction of complex phases.
- **3D case - Patching approach:** discusses a patching-method for the low-strain and high-strain cases in 3D.
- **Final 3D model:** presents the final DL model for the prediction of the RSP of highly strained 3D BCDI patterns. Results on simulated and experimental data are shown as well as the combination of DL prediction and iterative algorithms for refinement.
- **Performance assessment:** the final DL model and standard algorithms are tested on simulated data with different strain distributions to assess the benefits of the DL approach.

5.1 State of the art

In this paragraph I will focus on the state of the art for what concerns the Phase Retrieval of BCDI diffraction patterns with deep-learning, tensor-computation and automatic differentiation methods. Conventional phase retrieval iterative algorithms are discussed in the introduction chapter as well as other approaches.

Given the relatively new development of neural networks and more specifically even more recent for BCDI phase retrieval, I will try to give a chronological overview of the main works in the literature pointing out strengths and weaknesses. The use of deep neural networks for inverse problems in imaging was proven to be successful in lensless computational imaging by Sinha in 2017 [118] and holography by Rivenson [119]. However, the first work pioneering the field for BCDI is “Real-time coherent diffraction inversion using deep generative networks” published by Cherukara *et. al* in 2018 [120]. The paper presents two CNNs for the phase retrieval of small (32×32 pixels) 2D simulated BCDI patterns, one predicting the support and the other the phase. A U-Net like architecture with encoder-decoder was implemented, and the model was trained for 10 epochs in a supervised fashion with a cross-entropy loss function (see Appendix). The results showed an excellent agreement between prediction and ground truth also in presence of relatively strong phases. The potential of this new approach for phase retrieval becomes immediately clear when considering the drastic reduction of computational time and resources needed for the model inference. Once the model is trained, the reconstruction can be obtained within few milliseconds on a desktop machine. In 2020 Scheinker and Pokharel proposed another approach [121] that employs a CNN model for 3D diffraction patterns. The fundamental difference is that the object’s support was defined by its surface only, as it is assumed to be *compact* and *homogeneous* inside. Moreover, the surface was parametrized by spherical harmonics and the DL model was trained to predict 28 of the first even coefficients of the spherical harmonics. The model architecture was therefore essentially different since, while the encoder is just transposed to a 3D one, the decoder is replaced by a flattening and dense layer with 28 different classes as output. The model showed good performance on both simulated and experimental data, marking the first DL-based approach capable of real 3D BCDI phase retrieval. In the same year, Wu and coauthors, [122], opted for an architecture made of a single encoder and two identical decoders for the prediction of amplitude and phase of single crystals from the central slice of the BCDI pattern. They conducted the study on simulated data and tested it on one experimental case as well. What is evident from their work is the winning combination of DL prediction and iterative refinement. The speed and generalization capabilities of the CNN allows for fast and good estimations of the object’s support and phase. In addition, the precise and well established iterative methods can bring this initial guess to a more polished and accurate solution in fewer cycles than without DL prediction. This successful combined approach has been later adopted in other works, ours included.

In 2021 two important works were published. First, Chan *et al.* in [123] extended the encoder/2-decoders architecture to the 3D case. In their work they first created a “physics-informed” training set obtained building particles by clipping planes from a cubic FCC structure of atomic positions, relaxing them with LAMMPS software for molecular dynamics and computing the BCDI pattern around the (111) Bragg peak. The procedure is very similar to the one adopted by Lim *et al.* in [108] and described above in Section 4.4.1. Training the CNN on a restricted set of such created BCDI patterns biases the predictions towards physically meaningful particles. Moreover, it is interesting to notice that the training of the model was conducted in a

sort of unsupervised fashion as the loss function calculates the differences between the target diffracted intensity and the intensity obtained by the squared modulus of the discrete Fourier transform of the predicted complex object. Although the authors managed to successfully test their model on an experimental BCDI pattern, the small size ($32 \times 32 \times 32$ pixels) of the images accepted by the CNN was not yet enough for proper experimental use. The work of Wu *et al.* [124] published in the same year, lifted the size to 64 pixel-sided cubes, enabling their model to be tested on several experimental cases. Their CNN model maintained the encoder/2-decoders architecture for a simultaneous prediction of the object's amplitude and phase and explores for the first time the unsupervised training for refinement as well. The authors claimed that this approach is able to achieve better reconstruction quality with respect to current state-of-the-art iterative algorithms in use. The year after, Yao and coauthors published AutoPhaseNN [125], again an encoder/2-decoders architecture that completely trained in an unsupervised manner. This approach is beneficial as it doesn't require datasets labeled with a ground truth, which means that experimental data can be directly used in the training set. Another advantage is that it relaxes the limitation of simulating a sufficiently diverse population of samples, capable of constituting a comprehensive distribution of real cases. AutoPhaseNN was trained to predict an object the diffracted intensity of which matches the observed one according to a normalized Mean Absolute Error metric. The model showed to work on simulated data as well as on experimental data and once more the winning method lies in the combination of DL prediction and iterative refinement. AutoPhaseNN has marked a milestone in the BCDI data analysis, attaining 10X to 100X phase retrieval speed up with reduced efforts for the model training.

Although of different nature, it is worth mentioning the work of Zhuang and coauthors [126] in which two CNNs are used in the “deep image prior” (DIP) framework. DIP [127] typically implies the use of a CNN for an enhanced representation of an image, often to solve inverse problems like super-resolution, de-noising and inpainting. However, it differs from classical deep learning as there is no training dataset but a fit of the target problem exploiting the parameters of the convolutional layers and the efficient gradient descent provided by the automatic differentiation. In their work, Zhuang *et al.* formulated the more general far-field phase retrieval problem as an optimization problem and considered the phase symmetries that affect this class of solutions [128] (see Introduction chapter). Their work employs two DIPs, one for the modulus and one for the phase, and successfully manages to reconstruct simulated objects even in presence of strong phases. The last interesting contribution is the work of Yu and *et al.* [129]. In this paper the authors proposed a DL model that computes complex convolutions, handling real and imaginary parts of the complex tensor in a single passage through the convolutional block. Complex convolutional layers are claimed to be better at preserving the physical connection between real and imaginary parts inside the complex object. Moreover, the authors made use of *skip connections* between encoder and decoder to enhance the training. This is a rather peculiar as this kind of residual links are typically used, in convolutional encoder-decoder networks, for tasks in which the input and output images are visually similar (i.e. segmentation, denoising, inpainting), thus, where it is more evident the information flow from the two blocks of the network. The model was used for the phase retrieval of experimental 2D diffraction patterns, for which an unsupervised refinement was used as well.

Before proceeding with our study, Table 5.1 summarizes the key features of the works from the two leading BCDI research groups at Brookhaven and Argonne National Laboratories, highlighting similarities and differences to guide the development of our model.

First, it is interesting to notice that the architecture's choice, from treating the object's

	Architecture	Last Activation Layer	Loss Function	Refinement
Cherukara - 2018 [120]	Two different UNets	Sigmoids	Cross Entropy	-
Wu - 2020 [122]	Encoder / 2 Decoders	ReLU	MSE on mod and phase + PCC on magnitudes	Iterative
Chan - 2021 [123]	Encoder / 2 Decoders	ReLU	MAE on normalized magnitudes	Automatic Differentiation
Wu - 2021 [124]	Encoder / 2 Decoders	LeakyReLU	MSE on mod and phase + PCC on magnitudes	Transfer learning + unsupervised training
Yao - 2022 [125]	Encoder / 2 Decoders	Sigmoid and Tanh	MAE on normalized magnitudes	Iterative (50 ER)
Yu - 2024 [129]	Complex encoder-decoder + skip connections	ReLU	MAE on real + MAE on imaginary	Transfer learning + unsupervised training

Table 5.1: Comparison of deep learning-based phase retrieval approaches.

modulus and phase separately with two different detached networks, moved over the years to a single “standard” U-Net that accounts for the complex nature of the data. Second, I noticed that the choice of the last activation layers, which are the ones producing the modulus and phase outputs, in their final value range, is not uniform throughout the articles. ReLU and sigmoid activations produce strictly non-negative outputs, making them naturally suited for quantities that are inherently positive, such as the modulus. In contrast, LeakyReLU and Tanh can generate negative values, which makes them valid choices for representing complex phases. However, the practical impact of this choice appears limited: in some cases, models can predict correct moduli using LeakyReLU and correct phases using ReLU or sigmoid. This can be explained by the fact that a global offset in the phase—shifting the entire range to positive values—does not change the physical solution. Consequently, a ReLU can produce a valid phase array, simply offset by a constant, and similarly for the sigmoid, provided the phase range fits within the activation’s output limits.

One of the most important components of the model is the loss function, as its gradients regulate the model’s parameters, hence the prediction. Except the first work that employs a cross entropy loss, normally used for classification tasks, other works opt for Mean Absolute Error (MAE) and Mean Squared Error (MSE), of standard use for regression and Pearson Correlation Coefficient (PCC) as well. Typically, when the loss is calculated between intensities the MAE and the PCC are used as they are more suitable for the high dynamic range of the diffraction patterns. MSE in fact, “would overly de-emphasize errors in mid-intensity regions of the images” [123]. Lastly, I have listed the different ways used to refine the DL predictions. Here we can notice that very soon GPU accelerated gradient descent methods have been used in replacement of conventional iterative algorithms. The unsupervised training allows to easily switch from inference to refinement using the same model in the same GPU optimized computing environment guaranteed by machine learning libraries like PyTorch and Tensorflow.

5.2 Reciprocal space phasing

Taking inspiration from these works, but significantly changing the perspective, it was decided to predict the “reciprocal space” phase (RSP) that is lost during the measurement of the BCDI pattern rather than the complex object in real space. The main, intuitive, reason behind this choice is the visual similarity between the morphology of the diffraction pattern and its corresponding RSP. Furthermore, it is common that many samples studied with BCDI

have facets that happen to be, to some degree, parallel with each other, thus interfering like a single slit, with the typical fringes of intensity that correspond to constructive interferences, interspersed with dark regions arising from destructive interferences. In these specific cases, the RSP shows a regular pattern in which there is always a π shift between two crests of the fringes as shown in Fig.5.1. Once the RSP is retrieved, one can then recompose the full complex diffracted wave-function and obtain the complex object via inverse Fourier transform.

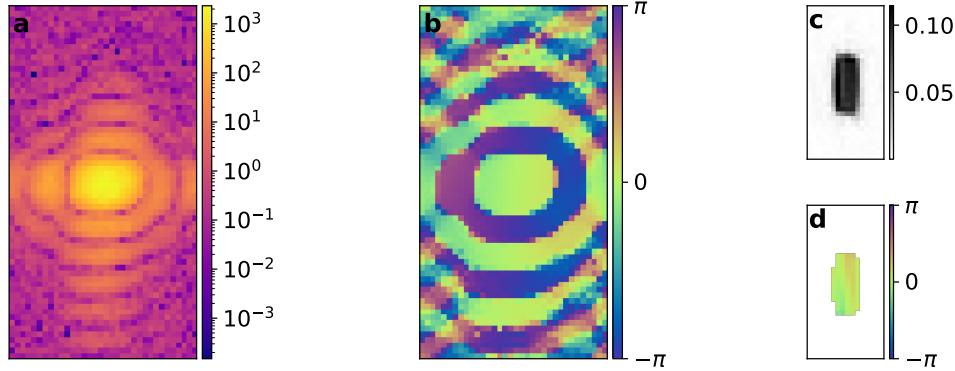


Figure 5.1: Central slice of a typical BCDI pattern (a) with the corresponding RSP (b) obtained after a successful reconstruction of the object (modulus and phase in c - d respectively). It highlights the structural similarity between the diffracted intensity in logarithmic scale and the RSP. Moreover, one can notice that in this case of low strain faceted particle, the RSP varies regularly between 0 and π (or $-\pi$) in correspondence of the intensity fringes.

Moreover, given this “simple” law of constructive-destructive interferences, we hypothesized the possibility to predict patches of this RSP given a portion of diffraction pattern and then, similarly to the inpainting case, stitch them together and obtain the full RSP. This entails a number of complications related to the so-called phase symmetries that I have encountered during the development of the algorithms and that will be discussed in the next sections. Ultimately, the goal of this DL model for phasing is to facilitate the reconstruction of highly strained particles. While other works in literature have mostly leveraged the gain in computing time, here the model aims at tackling those reconstructions for which conventional algorithms struggle to find convergence because of the high strain in the particle. However, in this case, the aforementioned RSP π -shifts in between two fringes is much more complicated since the strong and extended displacement fields inside the crystal alter the Bragg peak, merging and spreading the fringes into an irregularly distributed intensity pattern (see Fig 1.6).

5.3 Dataset creation

I have trained our model in a supervised manner, meaning, in this case, that the training was always conducted on simulated data only, as the RSP is never experimentally detectable. For this reason, I have simulated the training dataset following the same procedure described in Sections 4.3.1 and 4.4.1 for the 2D and 3D cases, respectively. However, in this case, the dataset size was $64 \times 64 \times 64$ pixels and the simulated RSP was used as the ground truth label for training.

I will anticipate here that for the high strain case I created a dedicated training set simulating the strain by applying an artificial “strong” phase to the particles. In order to have a diverse population of strain distributions I have simulated each object’s phase using different functions and parameters, namely: with the sum of two Gaussian functions, with the sum of two cosine functions and using a correlated Gaussian random field [102]. In each case, centers, amplitudes, variances, frequencies, and correlation lengths were randomly chosen to ensure a phase variation within the particle ranging between 2π and 5π . By doing this, strongly distorted BCDI patterns, similar to experimental high-strain ones, were obtained. In particular, the two Gaussian functions phase can closely emulate the effect of the substrate induced strain inside Winterbottom particles.

Similarly to the inpainting case, the BCDI patterns have been transformed into logarithmic scale and normalized between 0 and 1. Batches of 32 images at the time were used.

5.4 2D case low strain

Alike the inpainting case, I have first conducted some preliminary studies in 2D, on noise-less low strain data. Here I will briefly show the model’s architecture, the loss function and the results.

5.4.1 Model structure

The architecture that I used has a U-Net like structure with an encoder and a decoder. The encoder is composed of six convolutional blocks through which the input diffracted intensity is progressively reduced from the 64 pixel-side squares to a 1D flattened vector. Each convolutional block is composed of a convolutional layer, a LeakyReLU activation function and a MaxPooling layer that halves the feature’s map dimensions. (illustrate the parameters later).

At the end of the encoder the so-called bottleneck, composed of a convolutional layer followed by a LeakyReLU activation, processes the feature map before passing it to the decoder which by means of transposed convolutions, LeakyReLU activations and UpSampling layers, brings back the feature map to the input’s size. Skip connections between encoder and decoder blocks are employed as well. The output tensor is the result of a last single-channeled convolutional layer with no activation function. In this way we let the model predict unbounded tensors to account for the phase symmetries.

5.4.2 Loss function

The choice of the loss function was first based on what was used in literature. A sum of the MSE computed on the objects’ amplitudes and one on the phases has thus been used (Eq. 5.2). The ground truth objects were indeed available from the simulated data while the predicted objects have been first calculated with a 2D inverse Fourier transform from the diffracted amplitude and the predicted RSP (Eq. 5.1).

$$\hat{O}(\mathbf{r}) = \mathcal{F}^{-1}\{\sqrt{I(\mathbf{q})} e^{i\varphi_{\text{pred}}(\mathbf{q})}\}(\mathbf{r}) , \quad (5.1)$$

$$\mathcal{L} = \frac{1}{N} \sum_{\mathbf{r}} \left(|\hat{O}(\mathbf{r})| - |O(\mathbf{r})| \right)^2 + \frac{1}{N} \sum_{\mathbf{r}} \left(\phi(\mathbf{r}) - \phi_{\text{gt}}(\mathbf{r}) \right)^2 , \quad (5.2)$$

5.4.3 Results

The training of the model was conducted on 8500 simulated BCDI patterns over 30 epochs with a learning rate of 0.0003 and monitored both training and validation loss. Here, Fig.5.2 shows the model's loss during the 30 epoch long training.

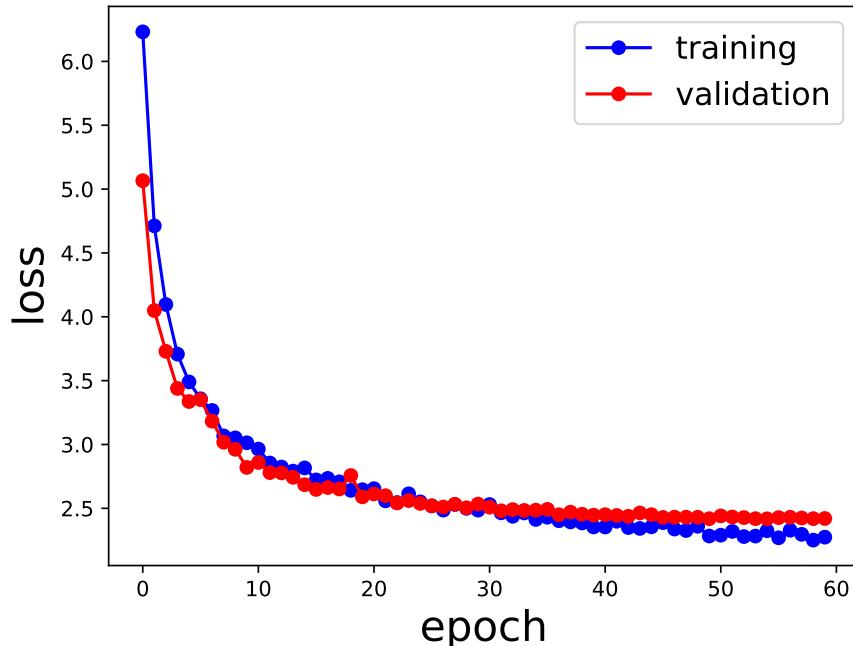


Figure 5.2: Training and validation loss over 30 epochs. The curve suggests a proper learning with no overfitting as both losses are decreasing reaching a plateau and the validation loss follows the same trend of the training loss.

Fig.5.3 illustrates the results of the predicted RSP of some test simulated BCDI patterns. Note that the displayed predicted RSP has been wrapped between 0 and 2π for better comparison with the ground truth but the raw output of the model is in fact an “unwrapped” array. This is expected since no activation layer was applied to the last convolutional layer, meaning that the last operation is the multiplication of the last feature map with the real values inside the convolutional kernel, hence linear.

When comparing the reconstructed objects obtained from the predicted RSP with the ground truth ones (Fig. 5.4) one can draw some interesting conclusions about the model's learning performance. First it can be observed that the model learns the approximate shape and size of the particle, it produces images that resemble reasonable particles, sometimes similar to the ground truth. The amplitude is concentrated inside the support with little noise outside and the phase is overall correct around zero.

However, when looking more carefully, it is clear that the shape is not quite correct, especially for highly non-centrosymmetric objects. For instance, if we consider the object in Fig. 5.4 **c**, we see that the predicted shape seems to be deriving from the incorrect superposition of the correct shape and its twin, as well correct. More in general it seems that the model tends to predict centrosymmetric objects. According to Sicairos *et al.* [130], if we name $\varphi(\vec{q})$ the correct RSP, this phenomenon arises from a predicted RSP phase ϕ composed of $\varphi(\vec{q})$ in some regions of the q -space and $-\varphi(\vec{q})$ elsewhere. In other words, the model is not fully able to break the sign symmetry. This subject was recently studied by Tayal *et al.* [128] and Zhang and coauthors in [131]. In this last study, the authors show that if not broken in the dataset, meaning that during the training the model is exposed to both cases ($\varphi(\vec{q})$ and $-\varphi(\vec{q})$) indistinctly, the model is deceived to produce a mix of the two, since the sign information cannot be recovered from the input intensity. The authors conclude that in order to prevent this detrimental effect, one should break the symmetry in the dataset to bias the model towards one preferred sign.

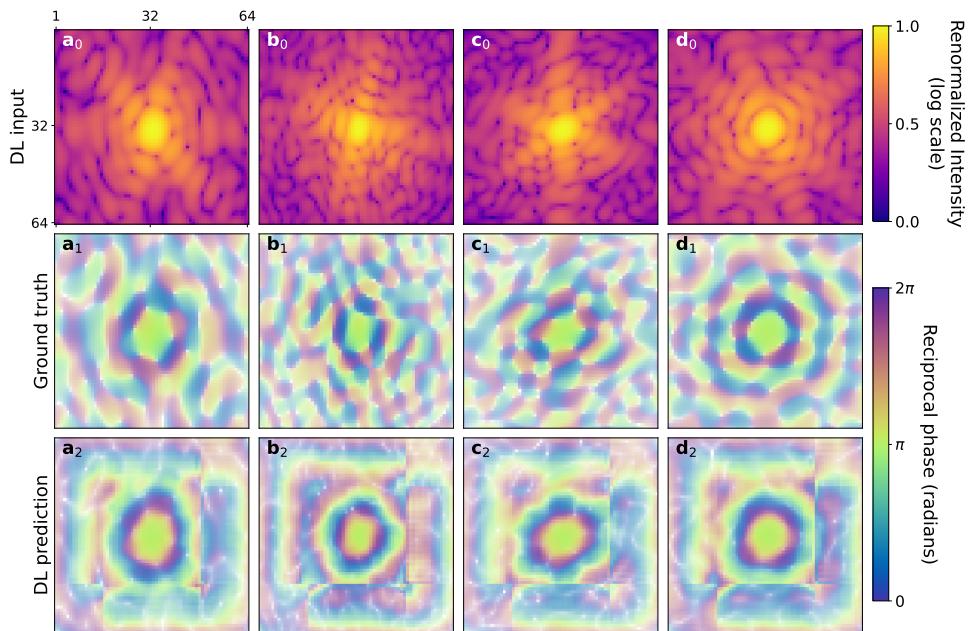


Figure 5.3: Model testing on new 2D data using MSE loss function. First row shows four simulated BCDI patterns, second row the ground truth RSP corresponding to the pattern and last row the DL prediction. The superposition of the two twin solutions resulting in the object space can be inferred here by observing how iso-phase lines in the predicted RSP tend to be more “circular”, as it would be for round direct-space objects. Moreover, some squared artifacts are visible in the predicted RSP. Being the side of the square half the size of the image, one can hypothesize that is originated by an incorrect up-sampling in the last convolutional layer.

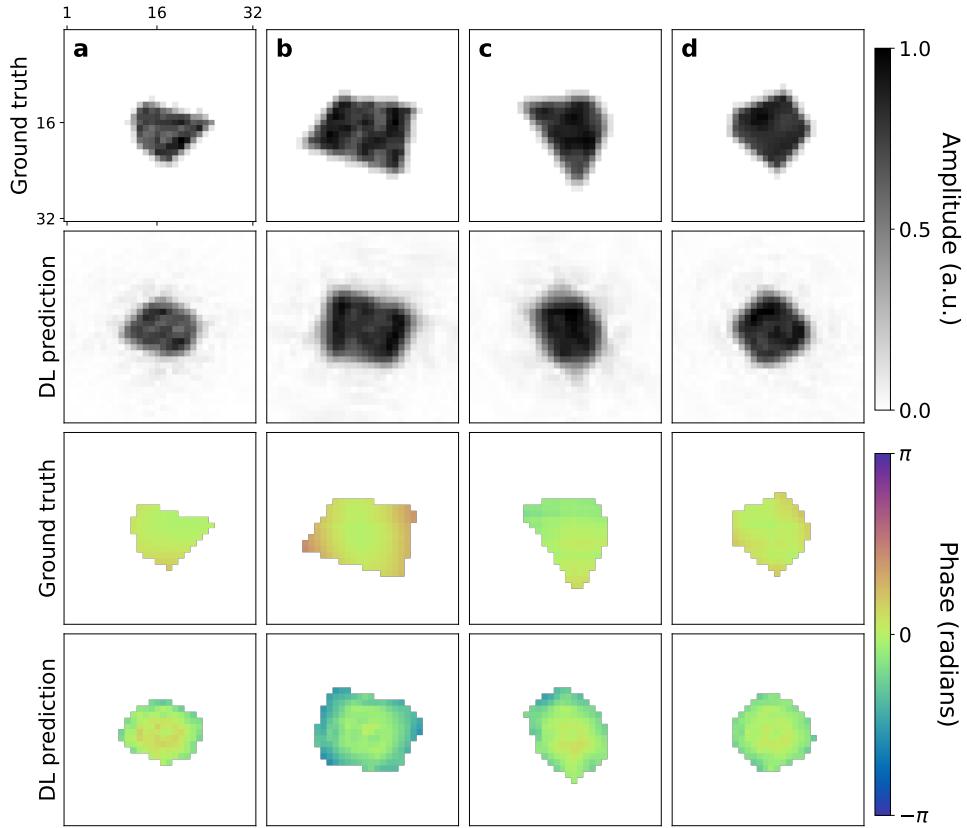


Figure 5.4: Corresponding reconstructed objects. Ground truth and predicted objects' amplitudes (first two rows respectively) and ground truth and predicted objects' phases (first two rows respectively). Note that the predicted objects resemble the superposition of the two twin true solutions.

The procedure presented in the article for the removal of the phase symmetries consists in: (i) the centering of all the objects in real space (phase ramp removal), (ii) the shift of the RSP such that the zero value in the same array position across the dataset (phase offset removal), and lastly, (iii) they flip the sign of the RSP when its value in corresponding to a fixed position across the dataset is negative. In our case the phase ramp symmetry was already broken by simulating particles with the center of mass in the center of the array. In this way the model is already biased towards the prediction of RSPs that yield centered objects. For the offset and the sign, the method proposed by Zhang *et al.* has been implemented in the model and the results are shown in Fig. 5.5 for the RSP and Fig. 5.6 for the reconstructed objects.

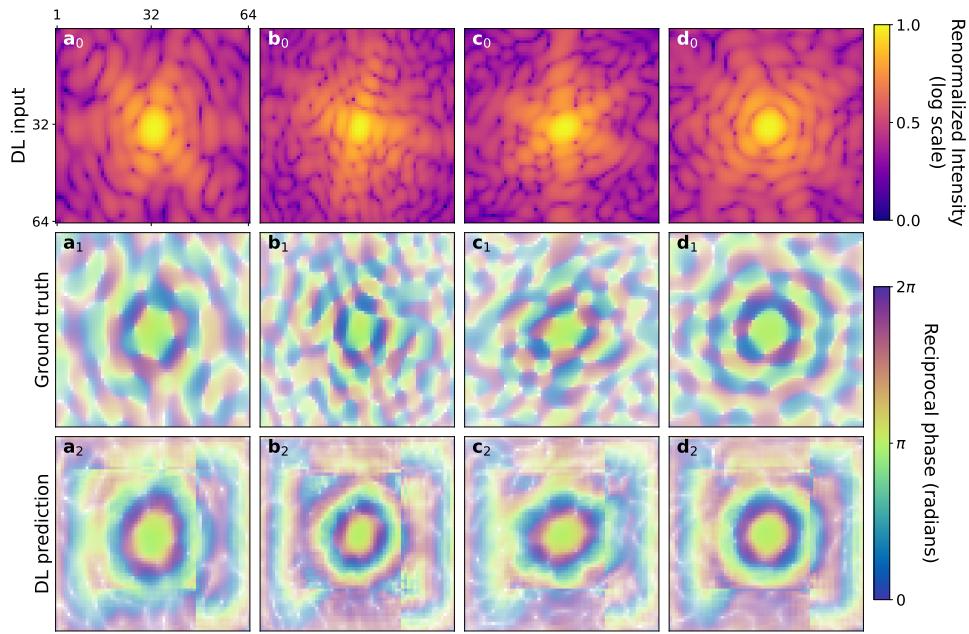


Figure 5.5: Model testing using MSE loss function and biased dataset. First row shows four simulated BCDI patterns, second row the ground truth RSP corresponding to the pattern and last row the DL prediction . The results do not seem to improve since the predictions look identical to the ones in Fig. 5.3.

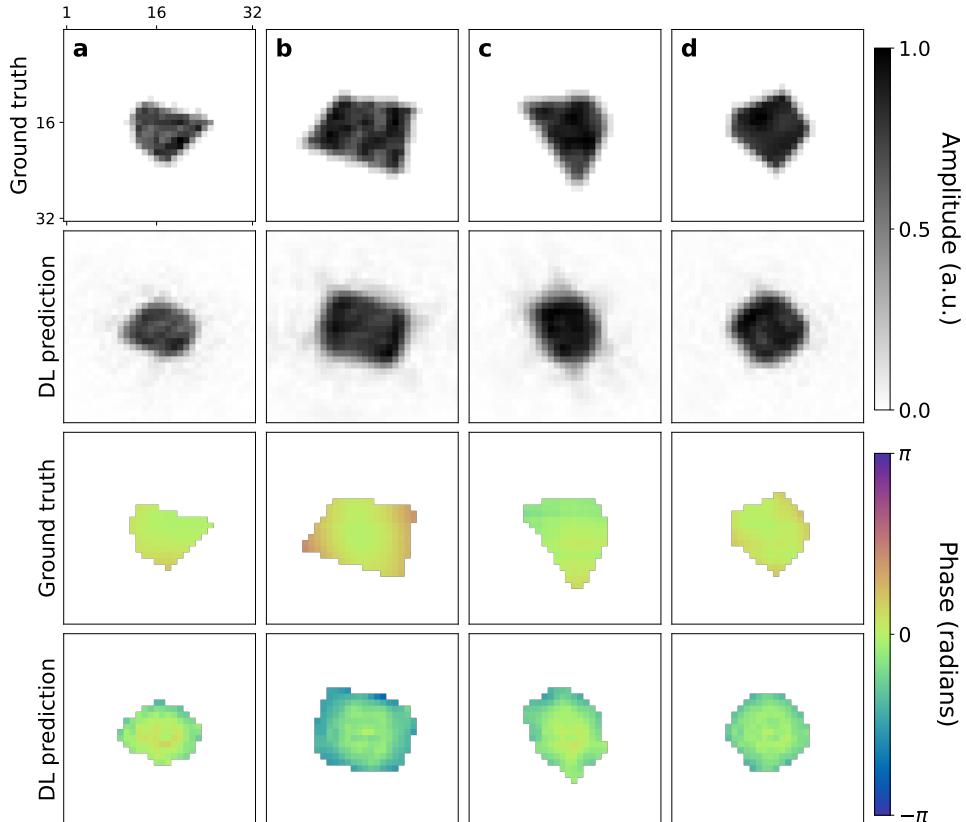


Figure 5.6: Corresponding reconstructed objects. Ground truth and predicted objects' amplitudes (first two rows respectively) and ground truth and predicted objects' phases (first two rows respectively). No significant improvement can be observed after the adopted sign-symmetry breaking procedure.

Unfortunately, the proposed method did not seem to solve the sign ambiguity of the RSP. The model is still unable to discriminate between the plus/minus sign of the RSP and the result is the incorrect overlap of the object with its twin obtained by the inversion symmetry. The phase, though small for this case, is also showing a kind of centro-symmetry as its variations tend to spread radially from the center of the array.

5.4.4 The Weighted Coherent Average loss function

At this point in the study, and in anticipation of applying the model to portions of the RSP, it became necessary to consider a loss function that would operate directly on the phase, without requiring transformations into direct-space. However, the main challenges were posed by the symmetries inherent to the phase. Upon further reflection, it was concluded that a Mean Squared Error (MSE) is not an appropriate metric for comparing the phases of complex functions. Indeed, MSE fails to account for the 2π periodicity and the possibility of a global phase offset. One could argue that 2π wraps can be fixed with a modulo 2π operation and the offset can be removed by shifting the tensor by a constant. However, the modulo wrapping function jumps abruptly by 2π every time phase crosses an integer multiple of 2π , meaning that the gradients are infinite thus not advised for gradient-based optimizations. Moreover, the MSE (or MAE and other *divergent* metrics) will have problems at the 0 - 2π boundary. In fact, when considering the phase mapped in the 0 - 2π range, if we suppose a $\varphi_{pred}^0 = -0.1$ where $\varphi_{G.T.}^0 = 0$, the wrap will move the φ_{pred}^0 to the value $2\pi - 0.1 = 6.183$ amplifying the error ($\Delta\varphi$) from 0.1^2 to 6.183^2 improperly.

In order to bypass these shortcomings a new loss function was designed. Here it follows the reasoning process that leads to the mathematical expression of the loss.

The best way to account for the periodicity and the wrap, without discontinuities and error unbalances, is to evaluate the ground truth - predicted phase differences ($\Delta\varphi_k$) on the unit circle. To do such, it's necessary to express $\Delta\varphi_k$ as angles of a complex exponential. This means that if φ_{pred} is an array of random values, each complex number $z_k = e^{i\Delta\varphi_k}$, when represented on the Argand plane, can be seen as a vector pointing at a random coordinate on the unit circle (Fig. 5.7c). Now, the goal of the optimization is not to minimize $\Delta\varphi_k$ for all k but to have the same $\Delta\varphi_k$ throughout k . In fact, for $\varphi_{pred} \Leftrightarrow \varphi_{G.T.}$ each vector z_k points in the same direction, but it does not necessarily lie on the x-axis ($\Delta\varphi_k = 0$ condition). Therefore, the loss function should ultimately drive all the z_k from randomly distributed to coherently aligned along a common direction.

A helpful quantity in this case can be the complex average vector $\langle z \rangle = \sum_{k=1}^N z_k = \sum_{k=1}^N e^{i\Delta\varphi_k}$ where k runs over all the N pixels. In particular the length of $\langle z \rangle$, represented by the modulus $|\langle z \rangle|$, is an efficient metric for the measurement of the *degree of alignment* of all the z_k between each other. In other words it measures the “coherence” between φ_{GT} and φ_{pred} .

In fact, $|\langle z \rangle|$ scores 0 for randomly oriented z_k , as opposite contributions cancel out each other because incoherent, while it scores 1 for perfectly aligned ones. It follows that one wants to maximize $|\langle z \rangle|$ during the optimization. Moreover, given the natural normalization between 0 and 1 of this metric, it follows naturally that the loss function can be expressed as $L = 1 - |\langle z \rangle|$.

Additionally, an importance mask can be applied during the averaging process. In particular, we know that the brightest pixels of the BCDI pattern are the ones contributing the most to the object's reconstruction. For this reason one could weight the complex average by multiplying by the input magnitudes. The effect of this operation is to "give a direction" to the optimization, meaning that the $\langle \Delta\varphi \rangle$ the model will tend to converge to, will be mostly steered close to the $\Delta\varphi_k$ of the brightest k pixels. The loss can now be expressed as:

$$L = 1 - \left| \frac{1}{N} \sum_{k=1}^N \sqrt{I_k} \exp(i(\varphi_{\text{GT},k} - \varphi_{\text{pred},k})) \right| \quad (5.3)$$

Where N is the total number of pixels in each RSP array and k is the pixel index. \sqrt{I} is the magnitude of the BCDI pattern normalized between 0 and 1 with respect to the sum, and φ_{GT} and φ_{pred} the ground truth and predicted RSP.

The last missing piece is the removal of sign symmetry. Rather than biasing the dataset preferring one sign over the opposite, the function L is computed for both φ_{GT} and $-\varphi_{\text{GT}}$ and in a second passage, the minimum of the two along the batch dimension is kept for backpropagation. The final form of the Weighted Coherent Average (WCA) loss is then given by:

$$L_{\text{WCA}} = \min(L_+, L_-) \quad (5.4)$$

To better visualize the functioning of the WCA loss function, a simple model has been trained to fit the ground truth phase of a single 2D BCDI pattern using the WCA. The complex phase differences vectors were extracted at each step of the optimization together with the updates obtained from the gradients of the WCA with respect to the trainable parameters. Fig. 5.7 shows the evolution of the predicted RSP as well as the progressive alignment of the z_k .

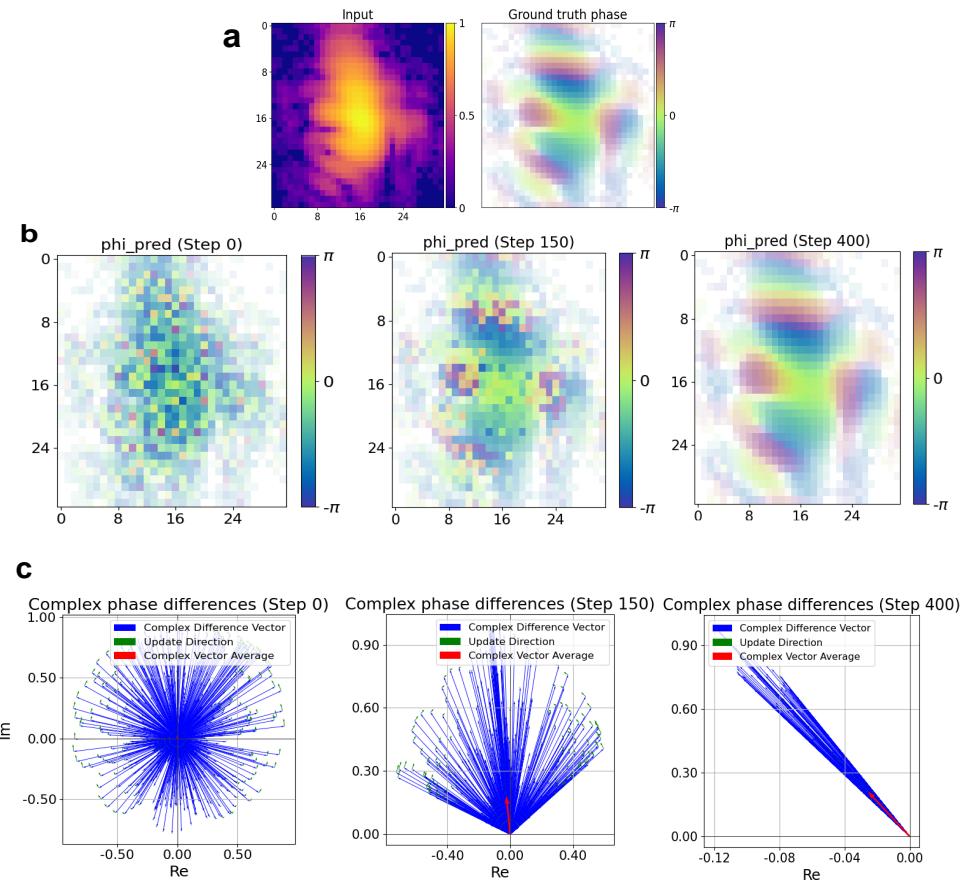


Figure 5.7: Illustration of the WCA loss function. **a** Input intensity (log-scale normalized) and ground truth RSP. **b** Predicted RSP in steps 0 - 150 - 400 of the optimization. **c** Corresponding complex phase-differences vectors z_k on the Argand plane (blue arrows), together with the updates (green arrows) obtained from the gradients of the WCA, and the resultant complex average $\langle z \rangle$ (red arrow). It is visible that during the fit, as the z_k align around a common one, the amplitude of $\langle z \rangle$ grows bigger and the predicted RSP converges to the ground truth one.

The same model has been trained using the WCA for the same number of epochs on the same dataset and here the results are shown. First, it can be noted in Fig.5.8 that the training and validation loss values throughout the training are following different trends with respect to the model trained with the MSE loss (Fig. 5.2)

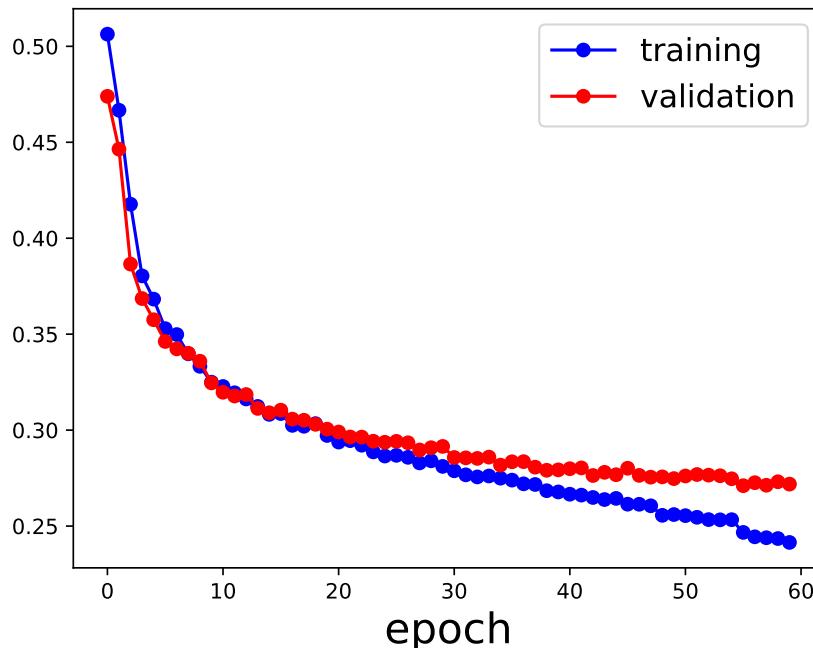


Figure 5.8: Training and validation loss curves over 60 epochs using the WCA loss function.

In this case the correct learning curve does not reach a plateau within the first 25 epochs but maintains a negative slope for longer, indicating a better learning. This suggests indeed better results when used on test data. In particular, for the same input diffraction patterns tested above in Figs.5.5 - 5.6 the model trained with the WCA yields the prediction shown in Fig.5.9 for the RSP and Fig.5.10 for the corresponding reconstructed objects.

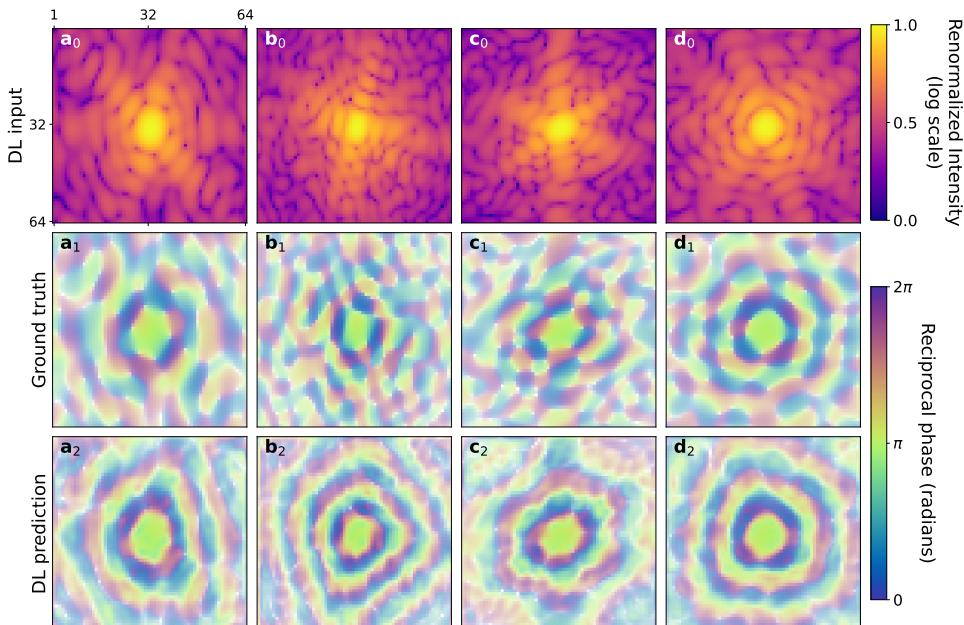


Figure 5.9: Model testing using WCA loss function. First row shows four simulated BCDI patterns, second row the ground truth RSP corresponding to the pattern and last row the DL prediction

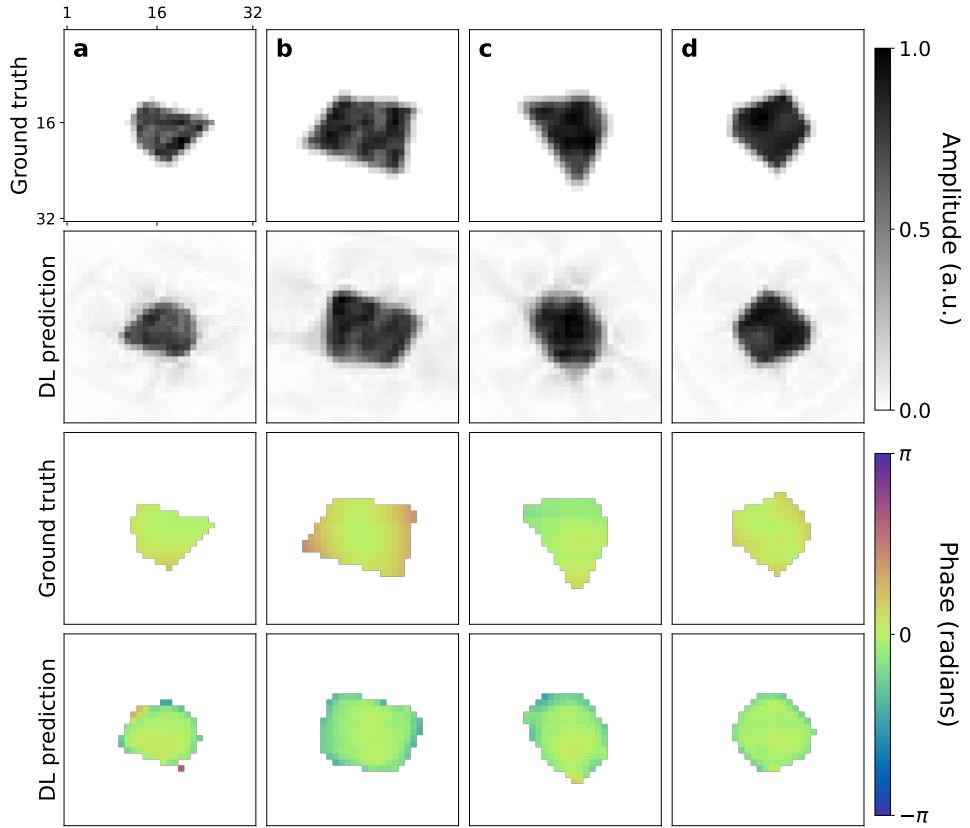


Figure 5.10: Corresponding reconstructed objects. Ground truth and predicted objects' amplitudes (first two rows respectively) and ground truth and predicted objects' phases (first two rows respectively).

The results obtained from the model trained with the WCA loss function are visually better than the MSE ones. Although not completely removed, the sign symmetry that gives rise to the superposition of the object with its twin, is less pronounced. For example, particles in Fig. 5.10(a-b-d) have a clear orientation and a shape that matches the ground truth. In all those cases though, the model has opted for the conjugate solution as the predicted object are flipped with respect to the ground truth ones. In Fig. 5.10(c) instead the symmetry is not broken and the result is still a superposition of the particle with its twin. This suggests that the symmetry breaking method implemented in the WCA, and the one proposed by Zhang and coauthors, is only partially playing a role in the actual model learning. It is interesting to notice indeed that when the training dataset or the model trainable parameters are increased, the sign symmetry is completely removed in the most difficult cases as well. Fig. 5.11 shows the effect of the dataset and models sizes for both MSE and WCA loss functions on the same simulated test data. The first important piece of information this figure shows is that the model trained with the WCA reaches higher accuracy. Moreover, it is much faster to compute since no FFT or IFFT is involved (see Fig. 5.11), thus the training time is drastically reduced. For what concerns the accuracy metric, in order to properly account for both modulus and phase, it has been calculated using

$$\left(\frac{PCC(m_{pred}, m_{GT}) + WCA(m_{pred}, \varphi_{pred}, \varphi_{GT})}{2} \right) \times 100 \quad (5.5)$$

where $PCC(m_{pred}, m_{GT})$ is the Pearson Correlation Coefficient on the object's moduli and $WCA(m_{pred}, \varphi_{pred}, \varphi_{GT})$ is the WCA function applied to the object's phase weighted by the

normalized predicted modulus. For what concerns the sign symmetry problem it is evident that while for the MSE trained model it is resolved only for a larger number of trainable parameters, for the WCA trained one it is already sufficiently overcome. As last observation, it is interesting to notice that when the model size is kept fixed and the training dataset augmented, the WCA improves the performances while for the MSE it is not the case.

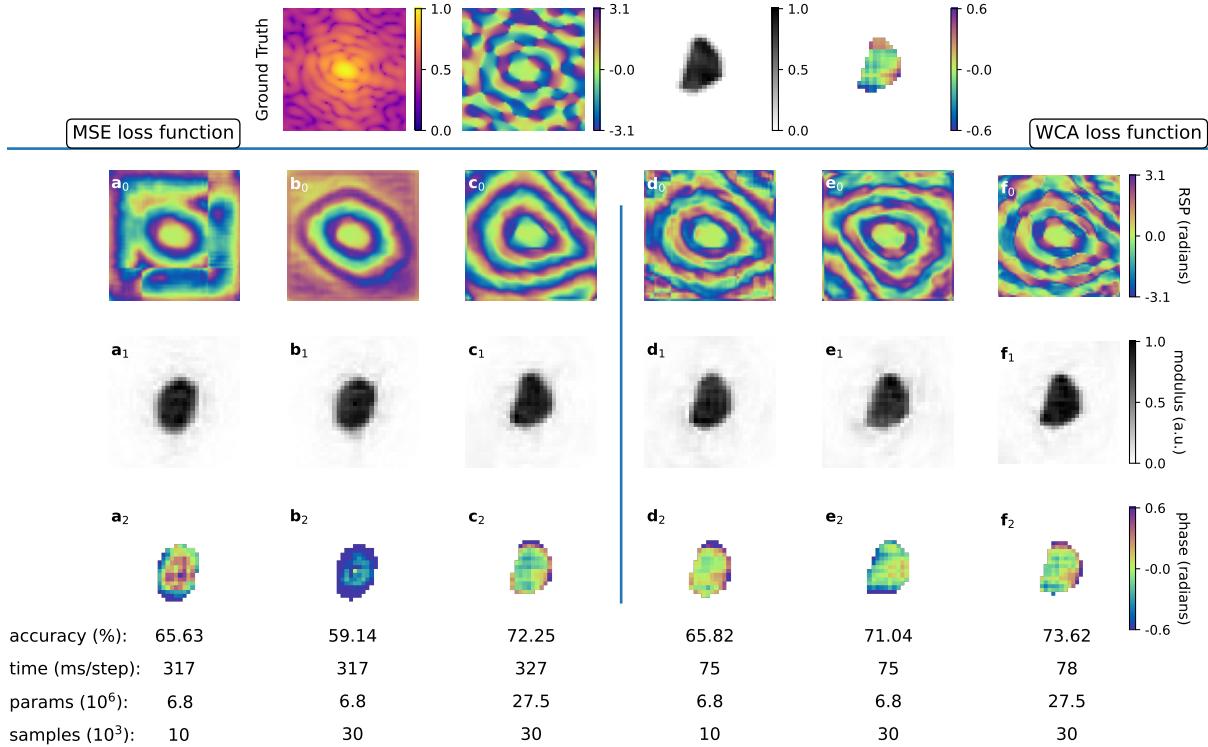


Figure 5.11: Comparison of MSE and WCA loss function for different model and training dataset sizes In the first row from left to right the input intensity, the ground truth RSP and the corresponding object (modulus and phase) are represented. a_0, b_0, c_0

are the results of the predicted RSP obtained from the model trained with the MSE loss function with the initial number of parameters and training set (a), with the augmented dataset (b) and with both model and dataset size increased (c) In third and fourth rows the corresponding reconstructed objects are displayed. d, e, f columns symmetrically shows the results obtained with the model trained using the WCA loss.

5.5 2D high strain case

In this paragraph the model training was performed on a dataset of highly strained 2D BCDI pattern simulated as described above in section 4.3.1. In this case Poisson statistic was applied to each dataset to better simulate the experimental condition. A set containing 30'000 samples was created and the “bigger” model of 27.5M parameters mentioned in Fig 5.11 was trained over 50 epochs with a learning rate of 0.001. Similarly to the low-strain case described above the same model has been trained with the MSE and WCA losses separately for comparison. The goal of this study is in fact to test the relevance of the loss function compared to the size of the model when the complexity of the task increases. The results on 4 test BCDI pattern are shown in Figs. 5.12-5.13 for the MSE one and 5.15-5.15 for the WCA.

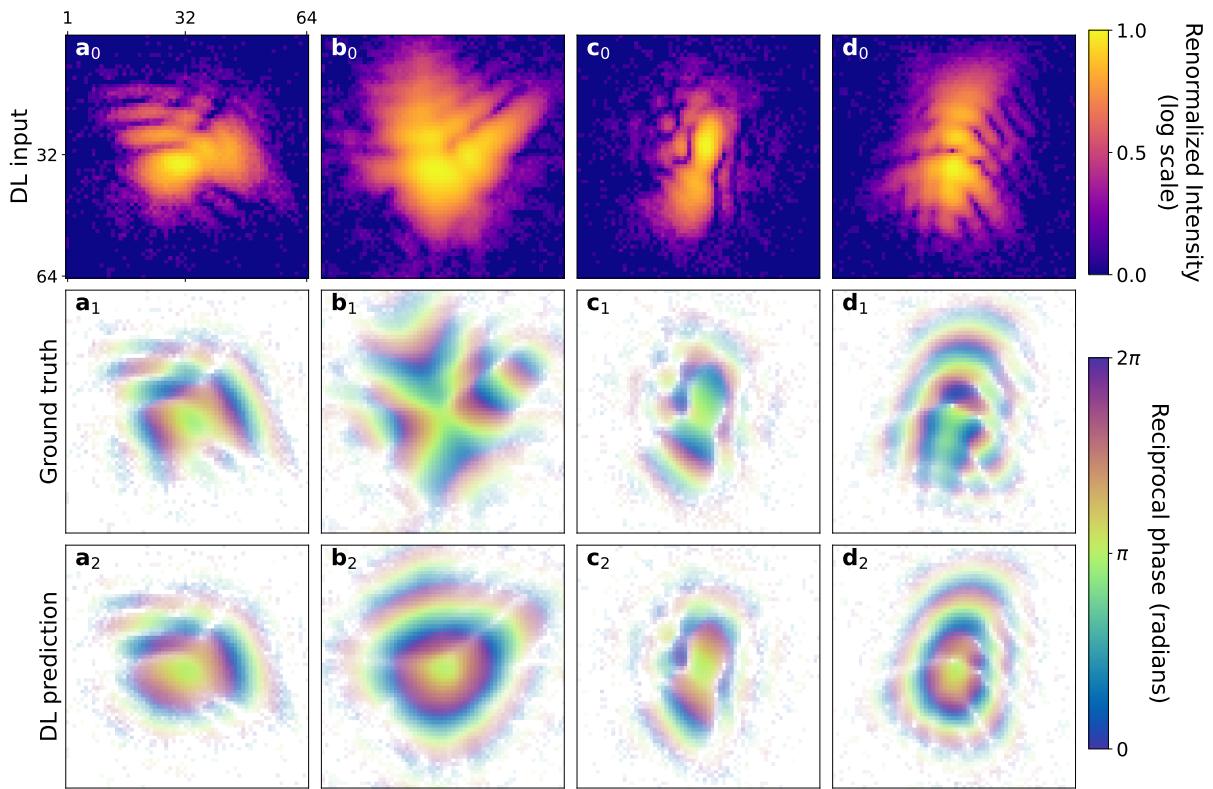


Figure 5.12: RSP predicted by the model trained with the MSE loss function calculated on both modulus and phase of the reconstructed object. First row: simulated 2D strained BCDI patterns (test dataset). Second row: corresponding ground truth RSP. Third row: predicted RSP. Once can notice that the model cannot predict correctly the RSP where the “iso-phases” do not have a circular symmetry (see **b-d**).

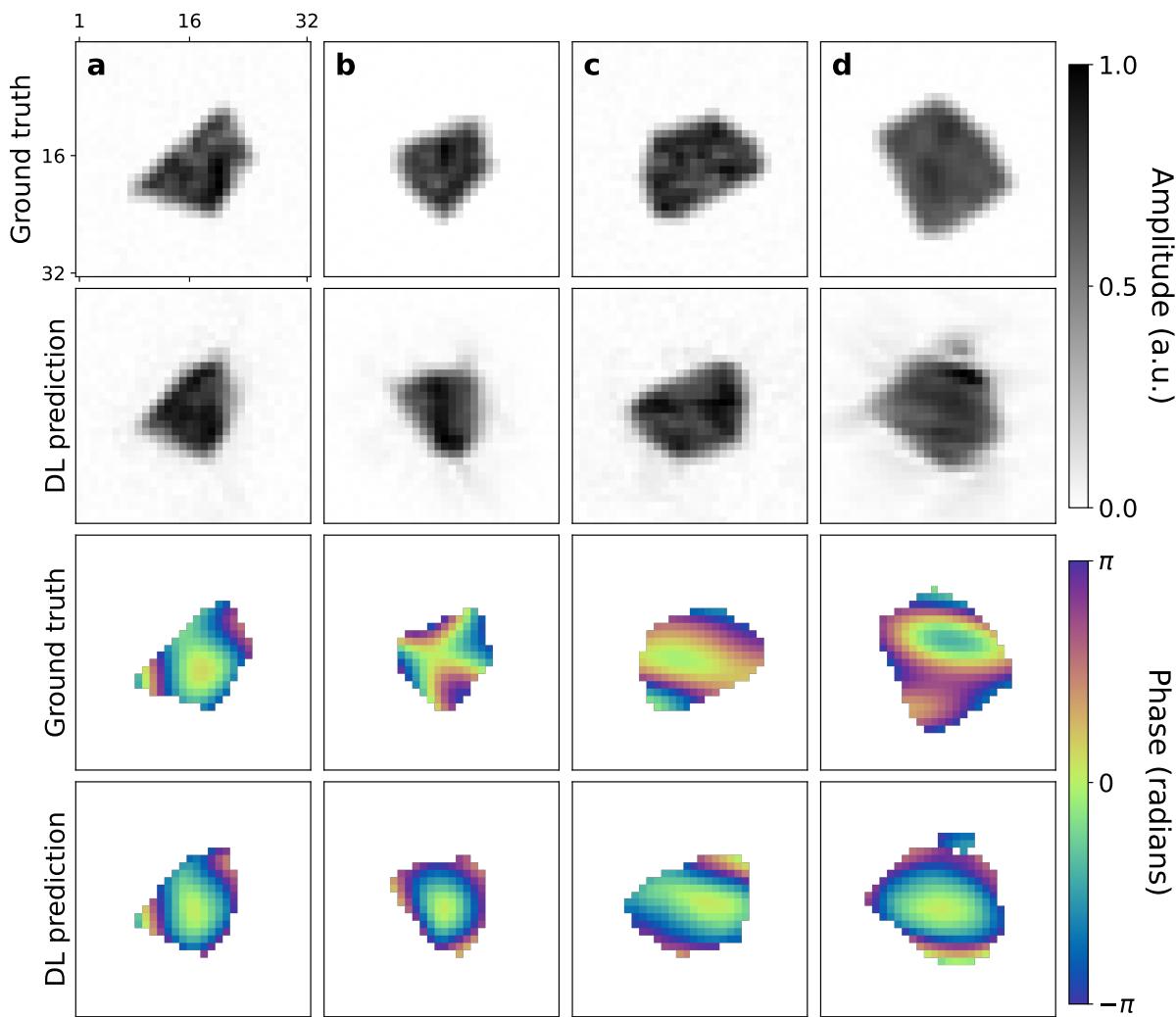


Figure 5.13: Corresponding reconstructed objects. First and third row: ground truth modulus and phase. Second and fourth row: model's results of objects' modulus and phase. Although the shape might at first sight look like the ground truth one (or the twin) the phase is often incorrect. It is curious to notice that better results are obtained when the object's phase possesses a certain degree of symmetry with respect to the center, analogously to the corresponding RSP.

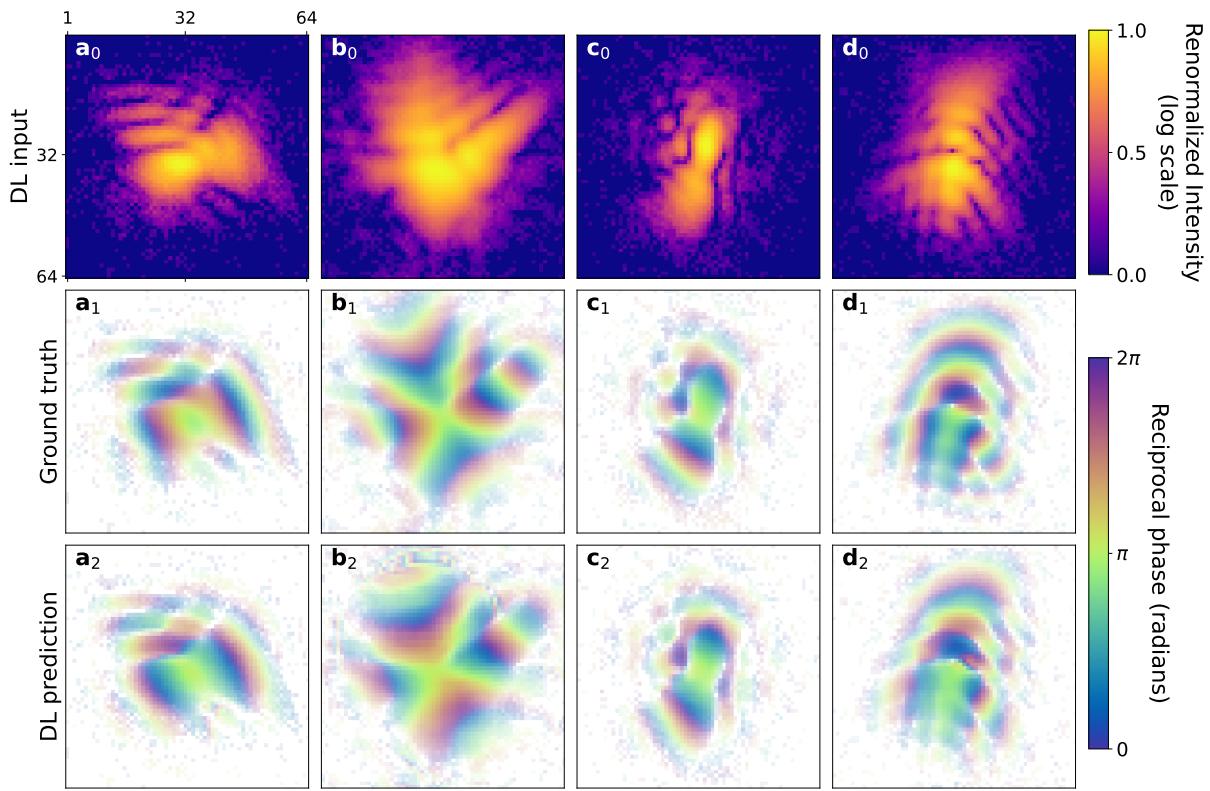


Figure 5.14: RSP predicted by the model trained with the WCA loss function. Here the model correctly retrieves the RSP for non-circularly symmetric structures as well (**b-d**)

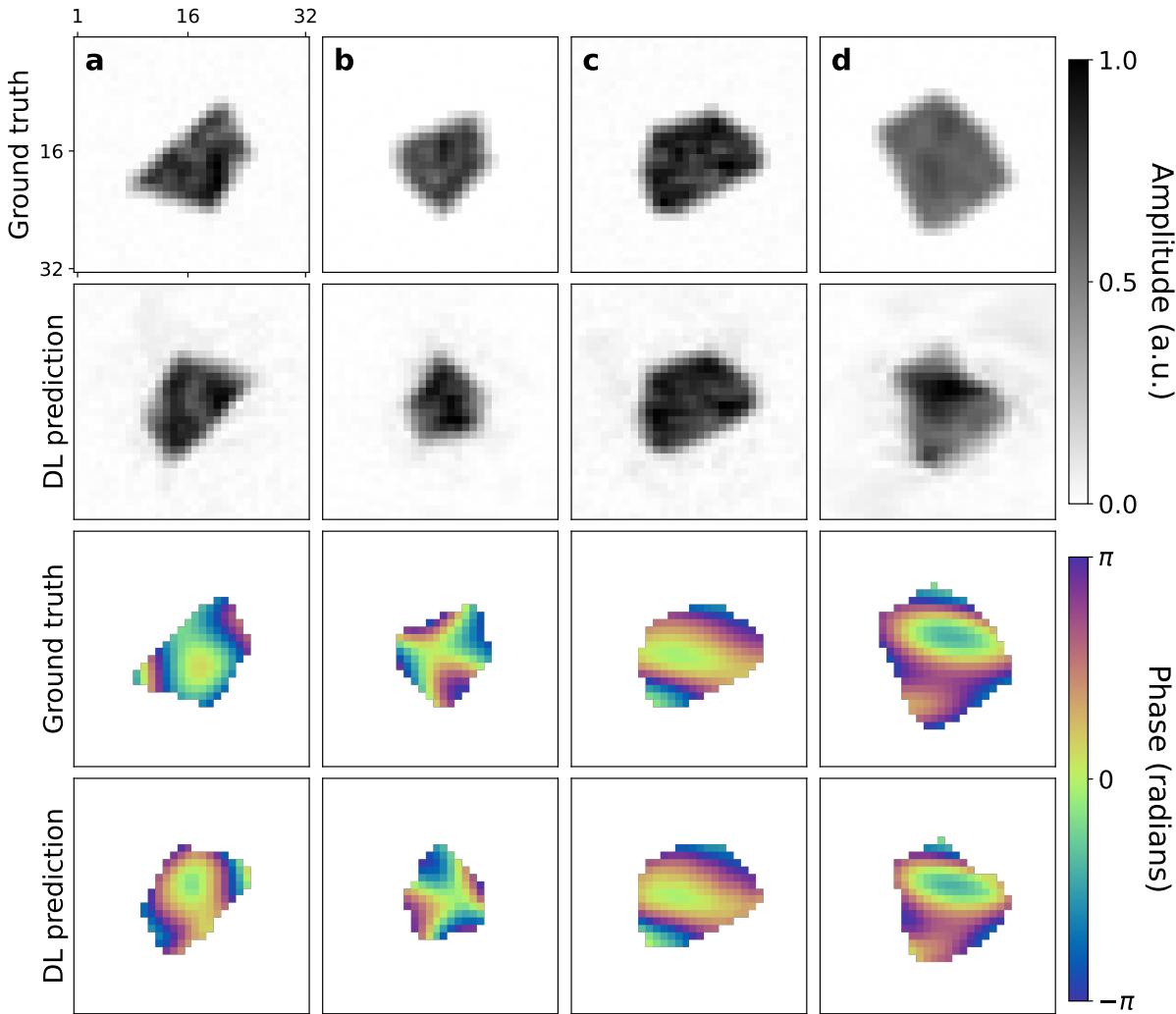


Figure 5.15: Corresponding reconstructed objects. Compared to Fig.5.13 the phase of the object is correctly recovered.

The preliminary studies on the 2D case for low-strain particles have demonstrated the possibility to recover the RSP from the diffracted intensity pattern with a U-Net like architecture without ever calculating the object in real space. Moreover, the studies on the high strain particles has shown that this model configuration is well suited for this case as well. From these promising results, it was decided to investigate the mapping intensity-RSP for portions of the reciprocal space.

5.6 Phasing patches: 3D case low strain

In this section of the manuscript the DL prediction of “patches” of the RSP will be explored and discussed. Three-dimensional BCDI pattern of low strained particles were used to conduct this study. Although this patching approach has not given satisfactory results for the PR, it is nevertheless reported in the manuscript as study on the *local* rather than *global* relationship between the diffracted intensity and the RSP. It is indeed known that there exist a unique mapping, barring some trivial RSP symmetries, between the diffracted intensity and the RSP in

3D [132]. What is interesting to investigate is whether this relationship exists also for subsets of the reciprocal space, and in particular if it can be retrieved by a DL model. (From now on the term “patches” will be used to refer to cubic subsets of the reciprocal space).

When deciding to work with patches, there is a number of questions that arise and the answer to which is not straightforward nor unique in many cases. Namely: What size is best? Can the patches be extracted at random positions or should there be an order? What about the normalization of the intensity range inside the patch? How are the patches stitched together into the full RSP eventually? How are the phase symmetries taken into account during the stitching? Here I will present the approach that allowed me to address these questions.

5.6.1 The choice of the size

Similarly to the inpainting case, 32 pixel-side cubic patches, cropped out of 128 pixel-side simulated BCDI patterns were considered. The choice is supported by the following reasons:

- The good results obtained for the inpainting case suggested that the amount of information contained inside a 32 pixel-side patch of reciprocal space is enough for the model to grasp spatial correlations.
- The average oversampling ratio of BCDI experimental data is such that in a 32 pixel-side volume a sufficient amount of fringes is contained, meaning intuitively that the model can predict the corresponding RSP.
- An even number multiple of 2 is usually considered GPU-friendly since it facilitates the shared calculations across different threads.

5.6.2 Patches division and stitching

At first, the patches were thought to be extracted randomly from the full BCDI pattern as for the inpainting case. However, by doing so the RSP of each patch would in principle have different offsets and different wraps than the neighbors and this would complicate the stitching of the patches back into the full RSP. For this reason, and considering the approximate spherical symmetry of the average BCDI pattern it was decided to crop patches radially, starting from the region around the center of the Bragg peak and the progressively moving outwards to higher q-values. In this configuration, an integer step (10 pixels in our case) was chosen beforehand and the first patch around the center of the Bragg peak was selected together with all the patches centered in distances of integer multiples of the chosen step. Fig. 5.16 shows a simplified schematic of the patches extraction. For the DL model training the patches of the intensity pattern need to be selected as well as the for the corresponding RSP for ground truth comparison.

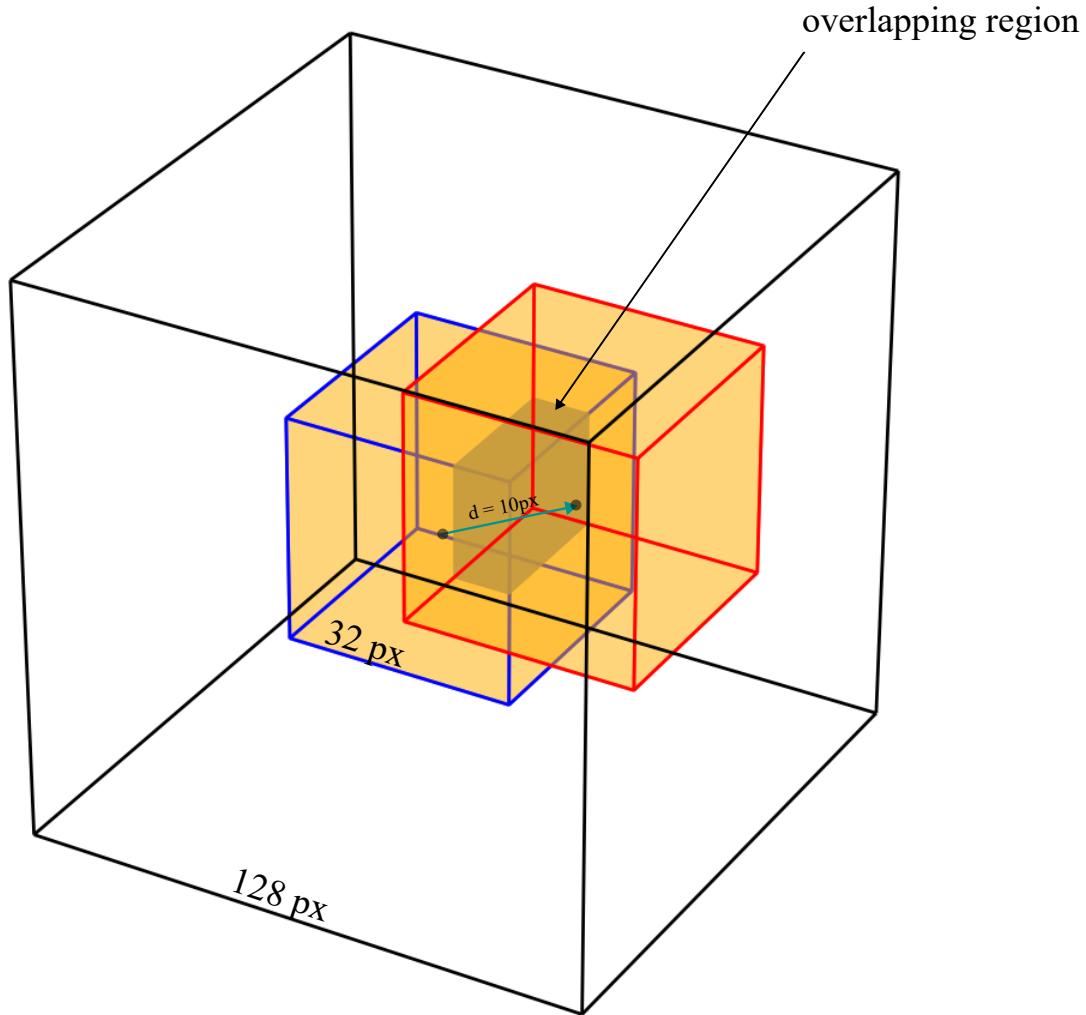


Figure 5.16: Schematic of the cropping of patches. From the full BCDI pattern (white 128 px-sided cube) the first patch is cropped out in the center and (orange 32 px-side cube with blue outline). Other patches are extracted radially from concentric shells separated by a 10 pixels step. Here only one patch from the first shell at distance 10 pixels from the center of the peak is displayed for simplicity. The gray shaded area highlights the overlapping volume between the two patches.

Being the step size smaller than the semi-diagonal of the 32 pixel-sided patches, it follows that the patches of adjacent cells have overlapping volumes. These common regions can have a twofold purpose. Firstly, they reduce the complexity of the stitching procedure since when this is executed progressively starting from the central patch, the sign and the offsets of the RSP are unambiguously fixed for all the following ones. Secondly, during the DL model training, for patches belonging to the outer shells, the overlapping volume of RSP belonging to the innermost adjacent shell can be provided as initial guess along with the input intensity patches. This of course cannot be exploited for the central patch that necessarily has to be predicted without initial RSP guesses.

The last question to be answered concerns the normalization. Since each patch is processed independently of the others by the DL model, it was decided to normalize each patch between

0 and 1 (always in log scale).

To summarize, the final design implied the use of two distinct training datasets and two different CNNs. The first dataset was dedicated to the central portion, therefore the first CNN was provided with 3D intensity patches in input (normalized log scale) and corresponding RSP patches as ground truth labels. A second training dataset containing patches from outer shells (5 concentric ones for a 128 pixel-sided full BCDI pattern) was created. Here each file was made of the pair intensity-RSP initial guess - from the closest neighbor patch belonging to the innermost shell - as input, and the full RSP ground truth patch corresponding to the input intensity. This second dataset was used to train a second CNN identical to the first one. One observation regarding the datasets is that there is an intrinsic imbalance between the number of central patches and the outer ones. In fact, for a single full BCDI pattern, the number of patches in the first shell is 1, while the number of outer portions can go up to several hundreds. Moreover, the central patch is the most important one as it contains a low resolution representation of the particle in real space. In order to balance the training, the first dataset was augmented with more simulated data.

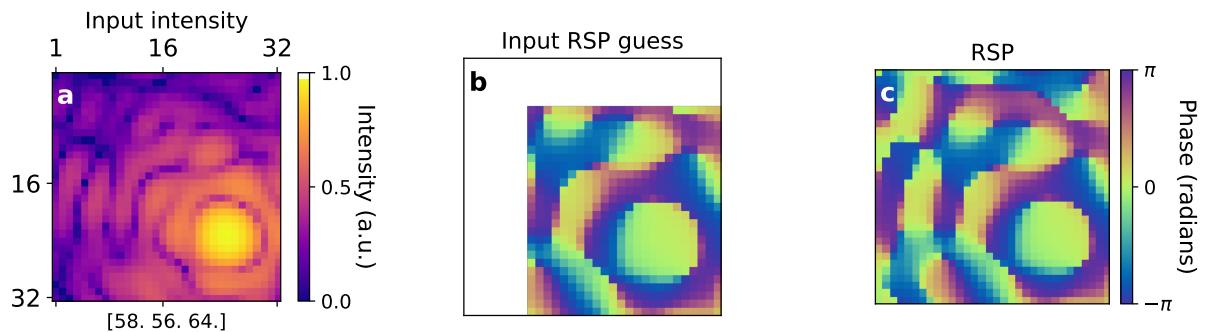


Figure 5.17: Example of input-ground truth pairs for the outer patches model. **a** Central slice of the input intensity for the patch cropped from the first shell at position [58,56,64] (the central patch is at [64,64,64]). **b** Initial RSP guess deriving from the overlapping region of the intensity patch with the central one. The blank area represents the part that needs to be predicted. **c** Ground truth RSP corresponding to the intensity patch in **a**.

5.6.3 Model architecture

The model architecture is similar to the one used for the inpainting case, with a U-Net like structure. 5 encoder blocks reduce the feature map to 512 one-dimensional vectors in the bottleneck and 5 decoder blocks upsample the feature map back to the original size. Skip connections are used as well and similarly to the 2D case for Phase Retrieval, the last layer has been left with no activation function. The total number of trainable parameters is of the order of 3.5 millions.

For the second CNN the layers and features are identical to the first one but three modifications were made. In particular, (i) the input tensor was composed of the intensity patch concatenated with the initial RSP guess and a binary mask marking the RSP guess voxels from the others. (ii) the mask was used at the exit of the decoder as well to select the new predicted voxels only for the backpropagation. (iii) The WCA loss function has been restricted only to the positive sign of the RSP since no sign ambiguity is left when fixing an initial guess.

5.6.4 Results on patches

The first model has been trained over 50 epochs on 10'000 samples of central $32 \times 32 \times 32$ pixel-size patches cropped out of $128 \times 128 \times 128$ pixel-size full BCDI simulated pattern using the WCA loss function. Fig.5.18 shows good learning curve which is supported by the results on test data, illustrated in Figs. 5.19 - 5.20

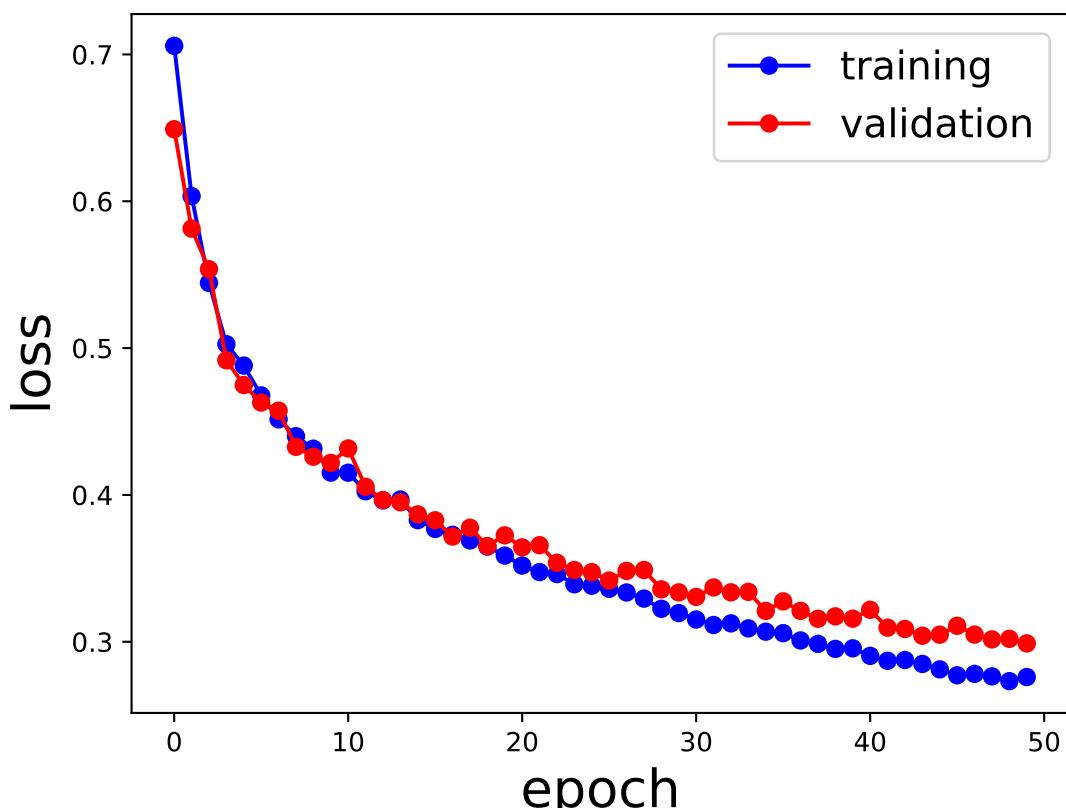


Figure 5.18: Training and validation loss curves over the 50 epochs long training. The plots indicate a good learning of the model because both training and validation losses are decreasing monotonically with the same pace.

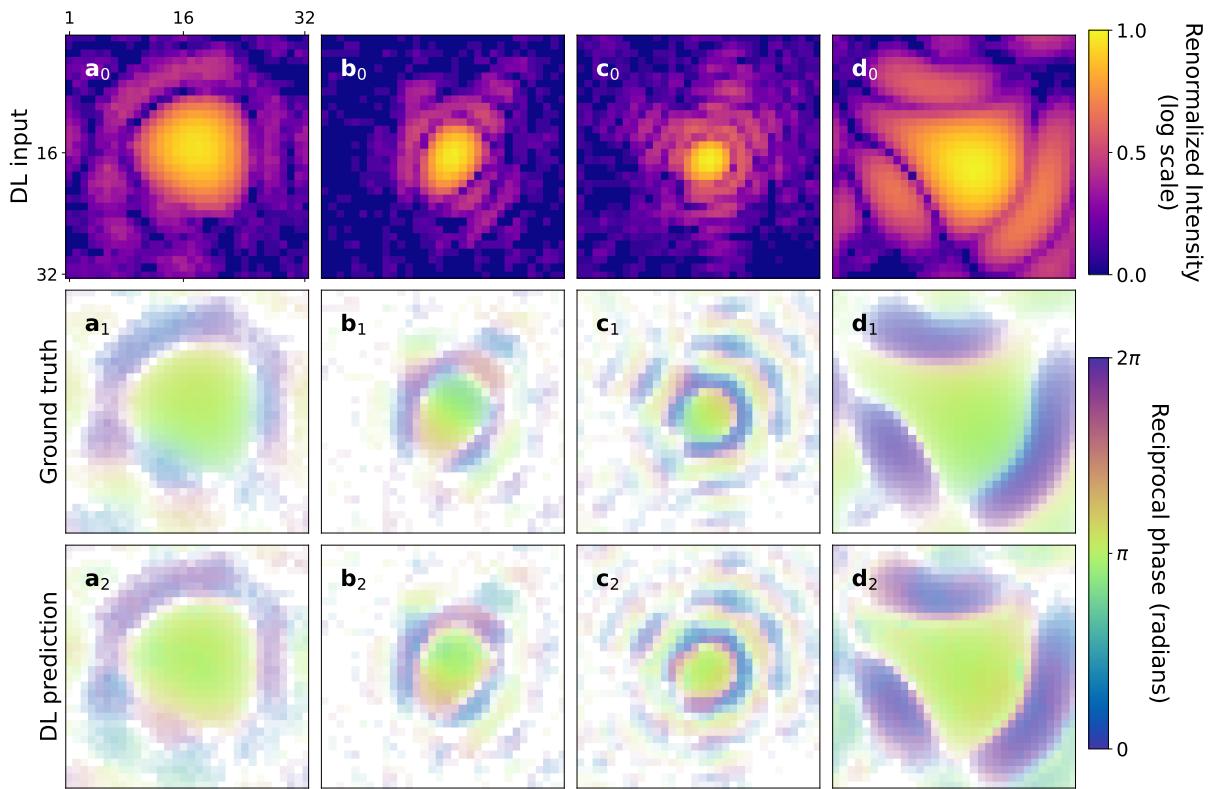


Figure 5.19: Slices of the central patches. First row shows four different examples of BCDI patterns cropped around the center of the peak. The small oversampling ratio of **b**₀ and **b**₁ makes such that the central portion already contains the full useful signal and shows the diversity of the dataset. Row from **a**₁ to **d**₁ shows the corresponding ground truth RSP while the last row shows the RSP predicted by the DL model.

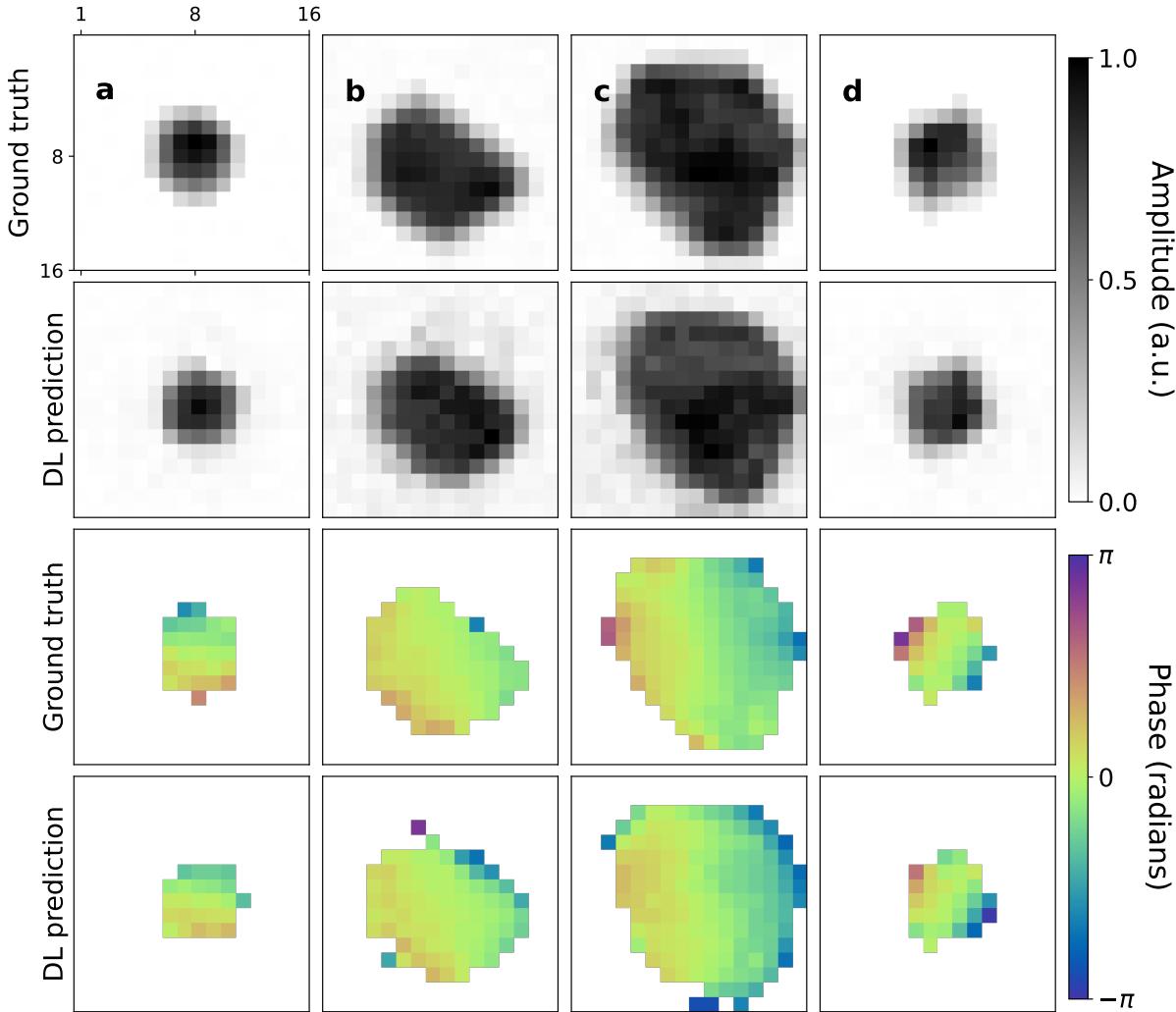


Figure 5.20: Corresponding objects. First and third rows show the modulus and the phase of the ground truth objects while second and fourth rows show the predicted ones. Although the low resolution due to the limited reciprocal space window, the model proves to correctly find the shape and phase distribution of the particle.

From the results obtained after the training of the model dedicated to the central portion of the simulated BCDI patterns, one can conclude that the model is capable of retrieving the correct RSP for the low strain case, meaning that the leap from the 2D case to the 3D case does not imply unforeseen complications. Moreover, given the diversity of the training dataset the model manages successfully for full peaks contained in a small patch.

The second CNN was trained on patches extracted from outer shells over 50 epochs on a dataset containing 50'000 samples. Fig. 5.21 illustrates some relevant results. In particular one can observe that the model can predict the RSP in the missing regions providing a relatively smooth transition between the “known” and “unknown” parts. This result is particularly interesting as it proves that a CNN trained with the WCA loss function learn the map that links a portion of diffracted signal with the corresponding RSP with no information on the particle in real space nor the position of this portion with respect to the center of the diffraction peak. It also shows that the size of the patches is contains a sufficient amount of information for this

map to be learned. However, one can also notice that some “noise” is present in the predicted regions. These discrepancies from the ground truth become detrimental during the stitching procedure.

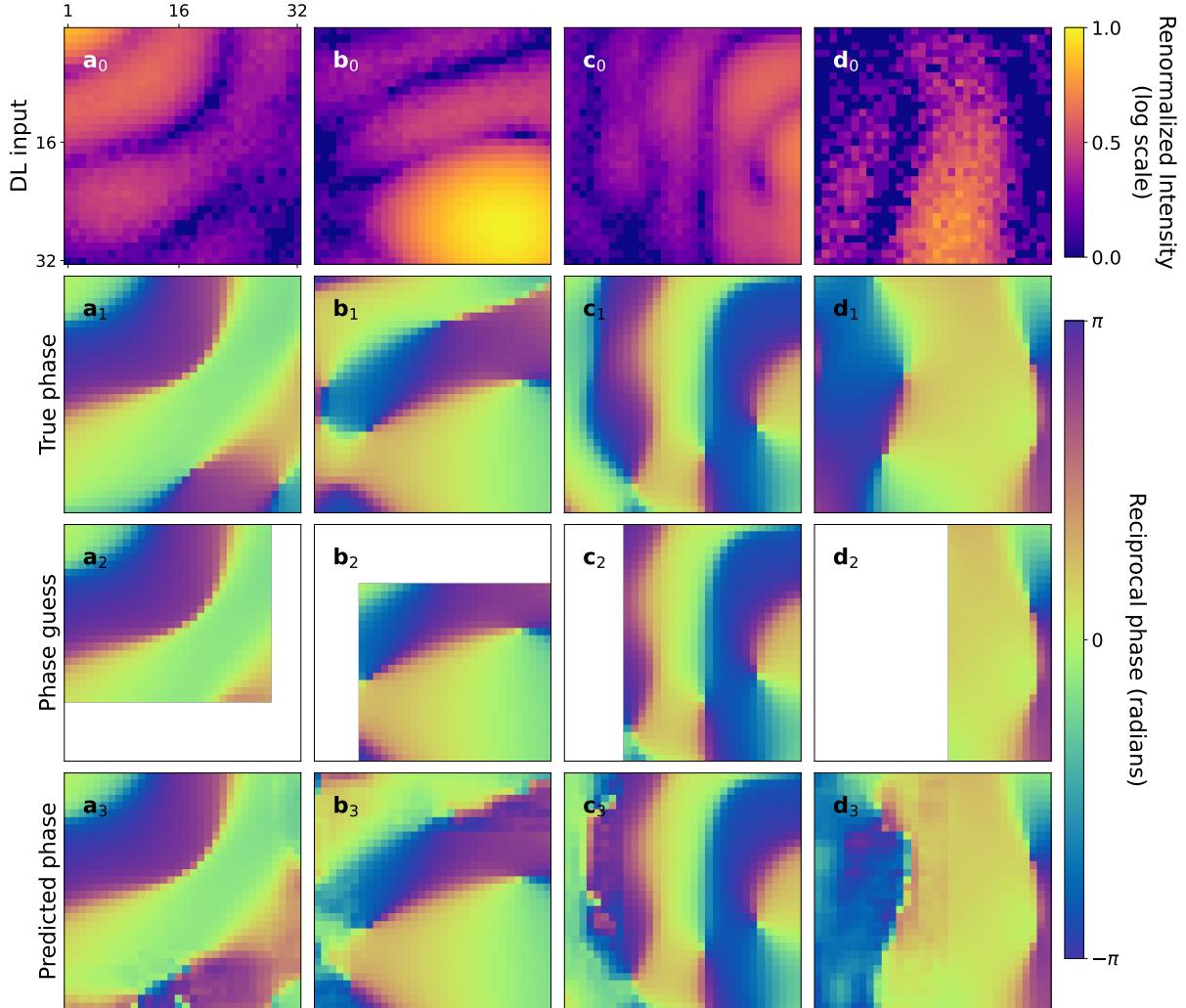


Figure 5.21: Examples of RSP prediction for outer patches. The first row shows the central slice of some patches extracted far from the central part of the BCDI peak. The second row shows the ground truth RSP, the third row the RSP initial guess obtained from the overlap with the nearest neighbor of the innermost shell, and the last row shows the DL output. The DL prediction is limited to the missing region.

5.6.5 Results on the full RSP

Here the results of the combination of the two CNNs are presented for the low-strain case. Once completed the training the model was tested on full simulated and experimental BCDI pattern. In order to properly retrieve the full RSP corresponding to the diffracted intensity a stitching algorithm for the predicted patches was designed. The stitching takes place progressively starting from the central patch and updating the full RSP array shell by shell. Once the prediction of the central patch RSP is obtained from the first CNN, the patches of intensity belonging to the first shell at distance 10 pixels are extracted and the overlapping regions between each

patch with the central one are calculated. The predicted central patch RSP for each overlapping region is therefore located in the initial guess RSP array that is given to the model as input paired with the intensity patch. Subsequently, the full batch of pairs $(I, \varphi_{guess})_{shell=1}$ is sent through the second CNN and the corresponding RSP output is obtained. At this stage two main issues have to be considered, namely: (i) during the training of the second CNN the initial guess RSP was taken from simulated *ground truth* RSPs while here it is taken from the model's previous prediction itself. It is for this reason that any small unavoidable discrepancies between the predicted and ground truth RSP can lead the model to further errors. (ii) When a round of RSP is predicted there are overlapping regions between patches of the same shell and patches of the previous batch. The most straightforward way to perform the stitching of the patches into the full RSP is to overwrite each time the results. However, although a better approach based on the average of the overlapping prediction was implemented, the issue did not seem to be fully resolved. A first test was conducted with this simpler solutions for both issues, and a more robust approach is briefly discussed at the end of the section.

The crop-predict-stitch method is hence repeated until the last shell. The number of shells and predictions scales with the size of the dataset, and it is however always restricted to a spherical region around the Bragg peak. It is therefore less accurate for non-cubic data. Here, Fig.5.22 shows an example of full RSP stitching for a cubic 128 pixel-sided simulated BCIDI pattern, while Fig.5.23 reports the analogous result for an experimental data phased with PyNX.

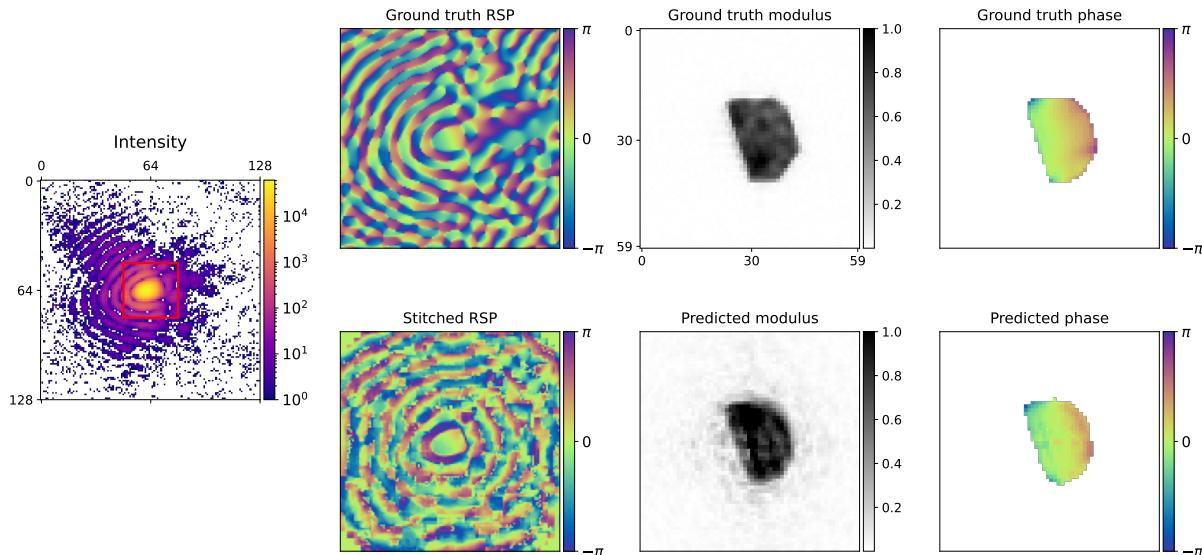


Figure 5.22: Results of the stitching of RSP predicted patches for 3D simulated data (central slice displayed). The red square on the intensity figure represents the central patch.

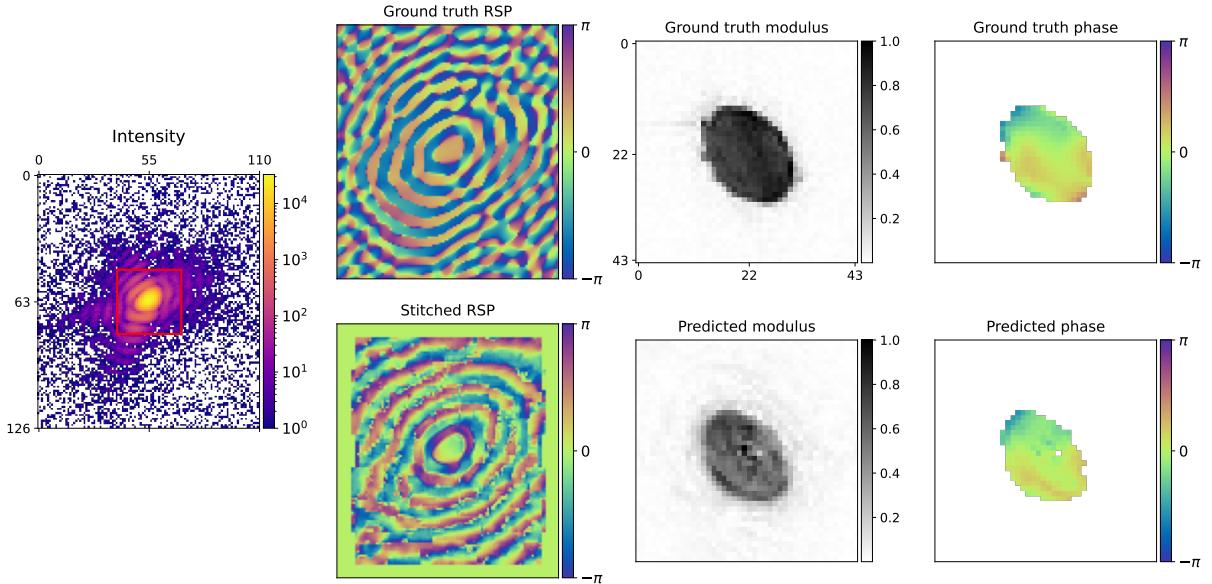


Figure 5.23: Results of the stitching of RSP predicted patches for 3D experimental data (central slice displayed).

What emerges from the above results can be summarized as follows:

- (i) the accurate prediction of the central RSP patch is fundamental for retrieving the low resolution estimate of the object
- (ii) the stitching is problematic already from the first shell, most likely because of the two issues pointed out above (initial guess from model prediction and RSP averaging of overlapping regions)
- (iii) the outer RSP patches correctly infer the oversampling ratio as the “thickness” of the RSP oscillations matches the one of the diffracted intensity, nonetheless, the overall orientation seems to prefer a circular.
- (iv) from the reconstructed object’s modulus a non-physical more intense spot in the center of the array had to be filtered out. The occurrence of this spike indicates the presence of wrong zero frequency components in reciprocal space, hence wrong zero RSP values. Fig. 5.24 shows the object’s modulus before the removal of the high intensity spike located in the center.

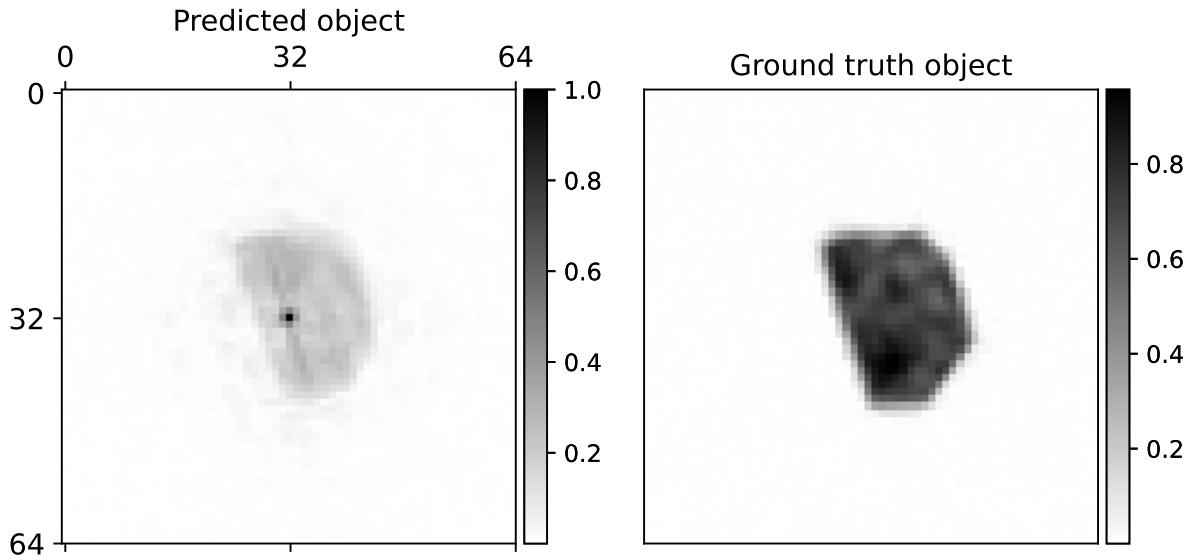


Figure 5.24: Predicted object's modulus before the filtering of the zero-frequency component.

5.7 Patches: 3D case high strain

For completeness the same model has been trained on a dataset containing highly strained simulated diffraction patterns. Each BCDI pattern has been simulated following the procedure explained above in Sec. 4.4.1 and 5.6.2 for a total amount of 50'000 for both central and outer patches. The two CNN have been trained for 50 epochs each and then tested on new simulated data.

For what concerns the first CNN trained on central patches it was possible to deduce a more difficult learning from the loss curves. While in Fig. 5.18 the validation loss reaches 0.3 at the end of the training, for Fig. 5.25 it only drops to 0.6 with a significant divergence between training and validation starting from the 20th epoch. A higher loss value is nevertheless expected because of the more complex intensity-RSP mapping for the high strain cases, while the increasing gap between training and validation loss is a signature of poor generalization, early symptom of overfitting.

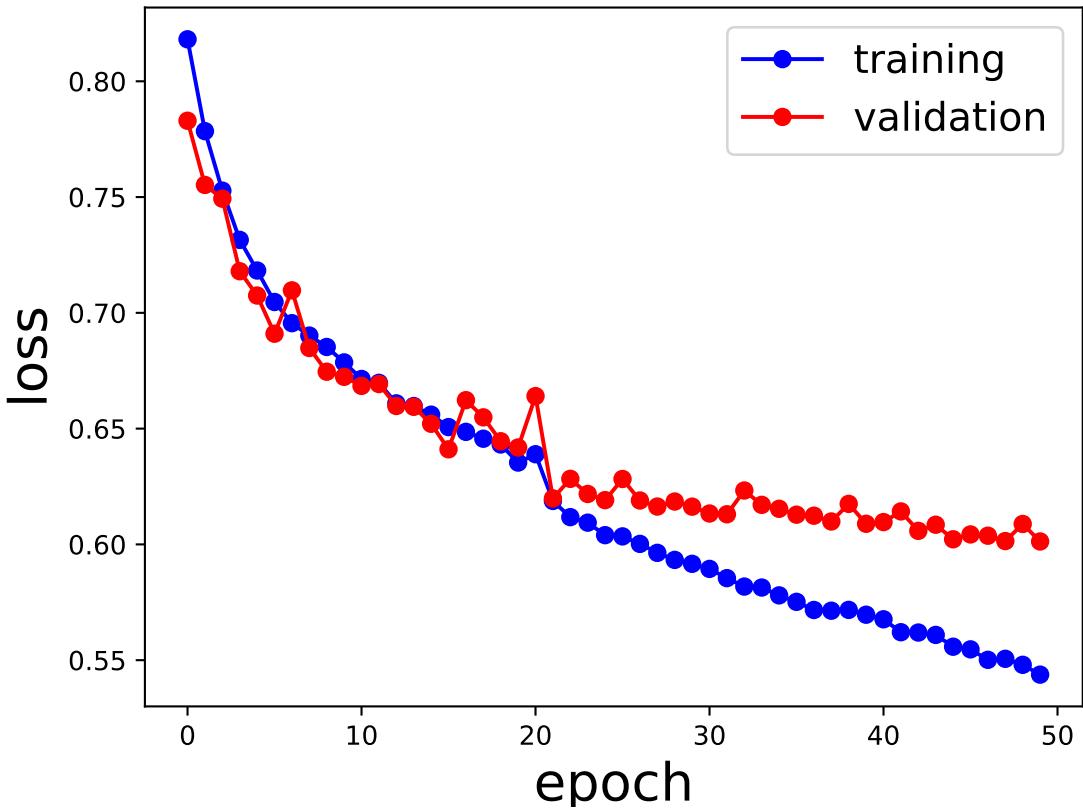


Figure 5.25: Training and validation loss trends for the training of the first CNN on central patches of highly strained BCDI patterns. Higher loss values can be observed when comparing with the low strain case (Fig. 5.18) and a beginning of overfitting from the 20th epoch is also visible.

The results on test data show in fact that the model not always manages to correctly predict the RSP, especially when the iso-phase regions are not spherically symmetrical (Fig. 5.26c). It however succeeds to retrieve the correct RSP oscillations (Fig. 5.26a) inside the central fringes elongated by the high strain. The reconstructions in real space, shown in Fig. 5.27, confirm satisfactory result for the first column and poor for the third column.

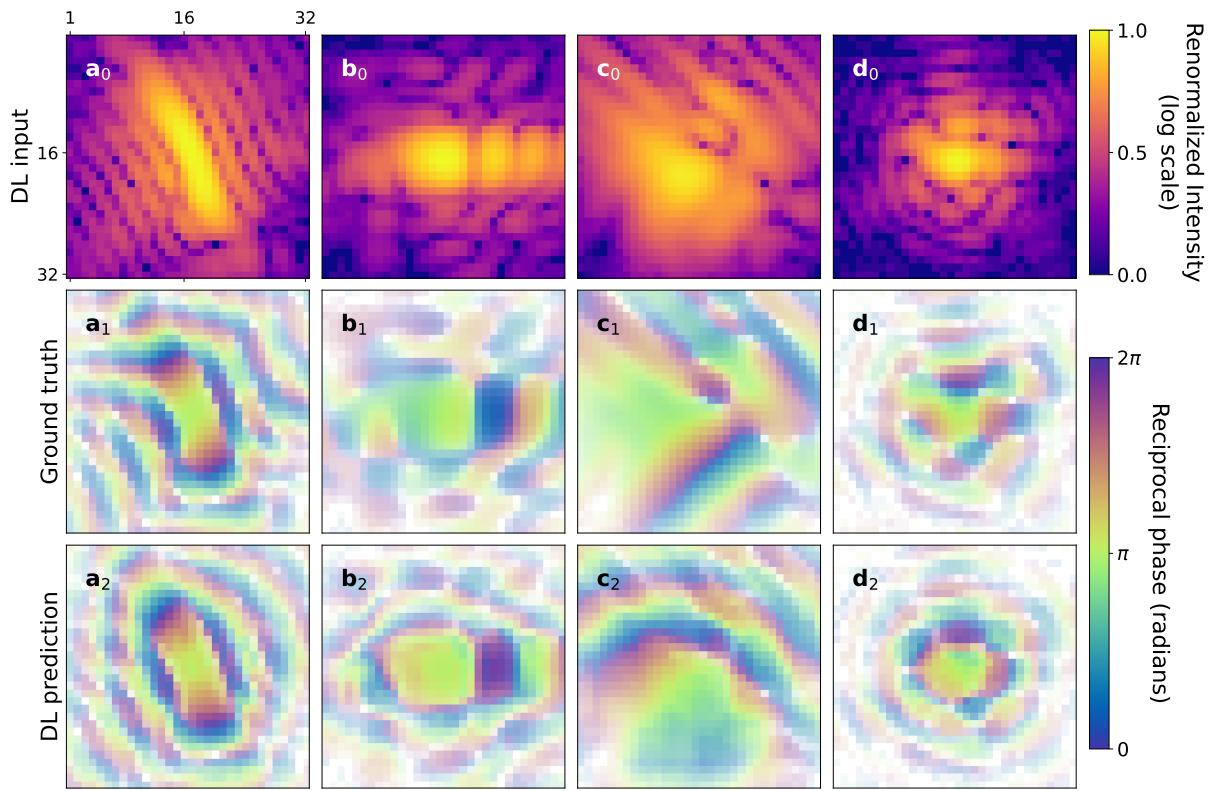


Figure 5.26: Slices of the central patches. First row shows four different examples of simulated high-strain BCDI patterns cropped around the center of the peak. Ground truth and predicted RSP are shown in second and third row respectively. The model manages to estimate the correct RSP when this shows spherically symmetric iso-phase regions while struggles more for different symmetries.

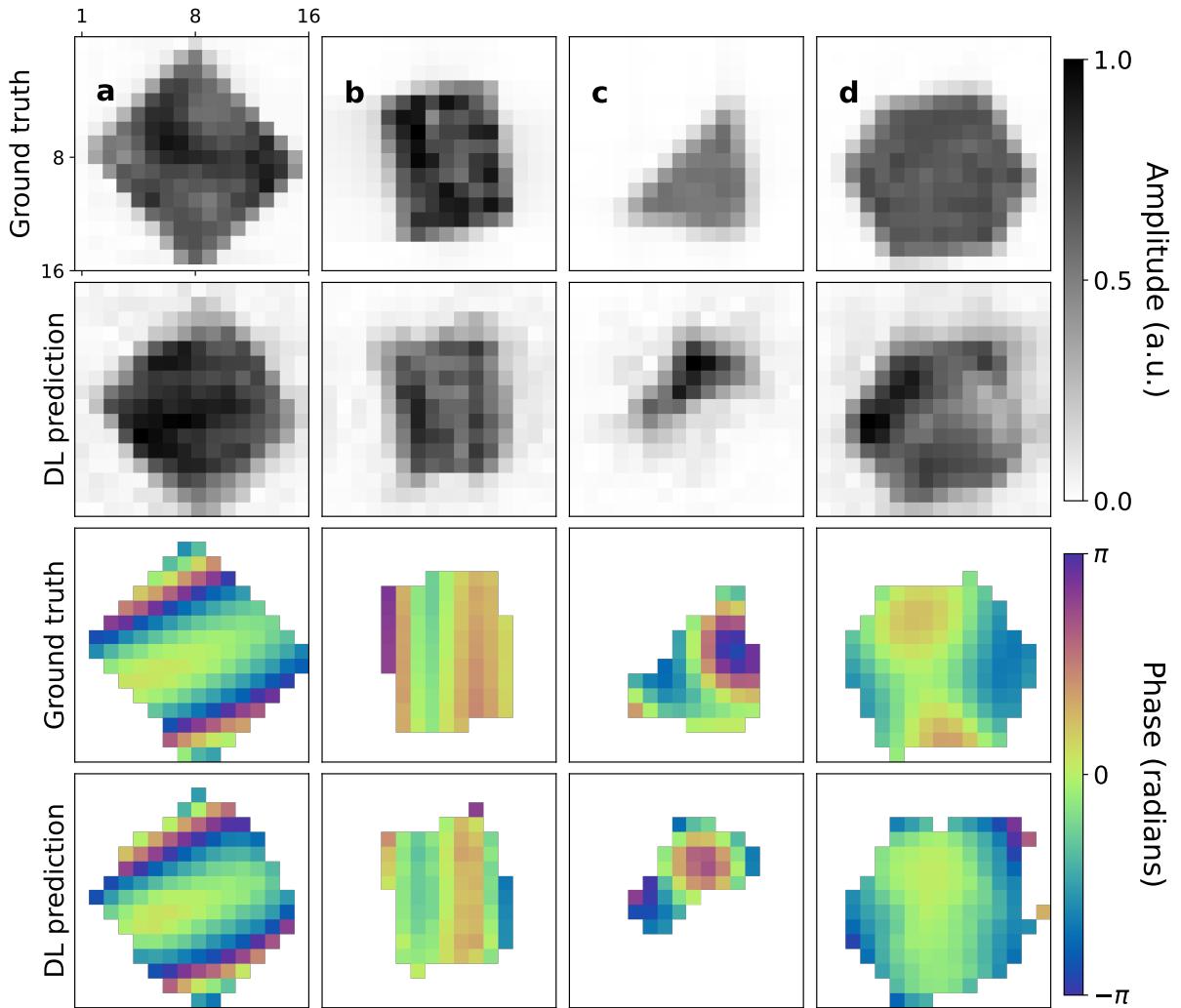


Figure 5.27: Corresponding reconstructed objects. Except for case **c** the predicted RSP is good enough to well estimate the size, shape and phase of the real space object.

Regarding the second CNN trained on patches instead, one can observe that the performance is not severely affected by the high-strain. Strong discrepancies between predicted and ground truth RSP are mostly present where there is no intensity signal, thus not relevant (see Fig.5.28). The good accuracy of on the outer patches predictions suggests that the crucial and more challenging mapping to retrieve involves the central patch mostly.

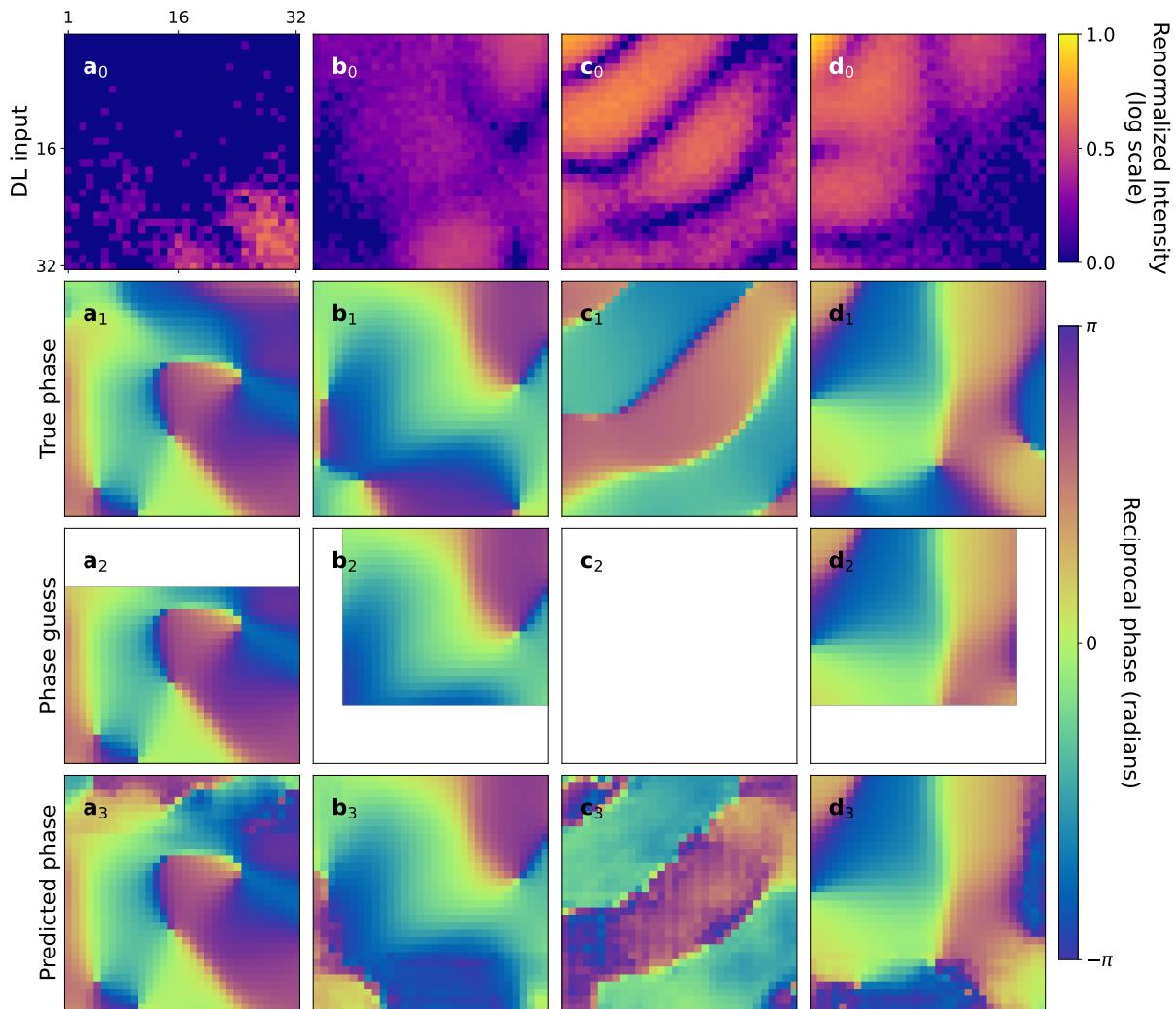


Figure 5.28: Examples of RSP prediction for outer patches cropped from simulated high-strain data. Similarly to the examples shown in Fig. 5.21 for the low-strain case, the model yields relatively correct outputs. Worse predictions are observed where low intensity signal is recorded, therefore less important during the reconstruction.

For completeness, it is shown here (Fig. 5.29) the result of the full RSP stitching and the corresponding reconstructed object for the high-strain case as well. A simulated test data has been used for the ground truth comparison. It is clear that the stitching algorithm is performing poorly as observed for the low-strain case. However, the central RSP patch is fairly similar to the ground truth and therefore the low resolution reconstructed object shows roughly the correct shape and phase.

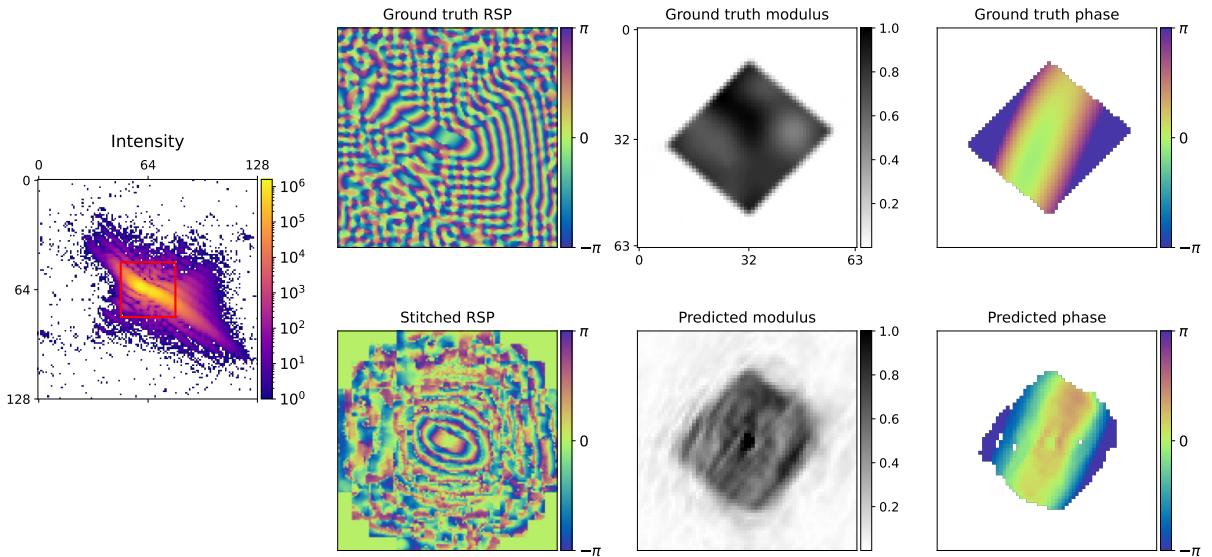


Figure 5.29: Results of the stitching of RSP predicted patches for 3D simulated “high-strain” data (central slice displayed).

5.7.1 Discussion

Although failed, this study on the prediction of the RSP in smaller patches has led to better understanding of the problem and nevertheless unveiled some interesting insights. For instance, it showed that the retrieval of the mapping between patches is possible with a CNN trained with the WCA loss function, untying even more the relationship with the real space object. Moreover, it emerged that the main difficulty of this approach is given by the stitching of the RSP patches into the full array. As mentioned above, the hypothesized reasons for this problem are (i) the fact that the model “sees” simulated ground truth RSP guesses during the training and predicted ones during inference, and (ii) the averaging of RSP predictions for overlapping voxels. In order to overcome these limitations other approaches were contemplated but never realized for lack of time. In particular, it was imagined a way to extract, analyze and stitch the patches inside the model into a sort of Recurrent Convolutional Neural Network (RCNN) that would keep track of the previous innermost shell thanks to a dedicated convolutional Long-Short Term Memory (LSTM) [133]. By doing this the model would always be exposed to its own RSP predictions as initial guess for outer patches and the RSP average over overlapping voxels could be replaced with a convolutional layer with non-linear activation function. While the attempts of setting up such model are not reported here, it is mentioned the idea as possible inspiration for future works.

To conclude, main finding of this study on patches is that it is crucial for a good object estimate to accurately predict the RSP in the vicinity of the center of the Bragg peak, and that the CNN model trained with the WCA can accomplish this task for highly strained patterns as well. It was therefore decided to invest the efforts into a regular CNN for the prediction of the RSP of 3D highly strained BCDI patterns with the intermediate size of $64 \times 64 \times 64$ pixels.

5.8 Model design: 3D case high strain

The model here described and the results obtained on simulated and experimental 3D BCDI patterns can be found entirely in the paper "Phase Retrieval of Highly Strained Bragg Coherent Diffraction Patterns using Supervised Convolutional Neural Network" ...

The architecture that was employed is an adaptation of the 3D U-Net employed for smaller patches Fig. 5.30 As learned from the preliminary study on the 2D case, to better interpolate the diverse distribution of distorted BCDI pattern, the number of trainable parameters and samples in the training dataset was increased significantly. Additional encoder and corresponding decoder blocks were added for a total of 145 million parameters. Moreover, similarly to the model used for the gap inpainting, dilated convolutions were adopted in the first two encoder blocks in order to improve the receptive field of the model.

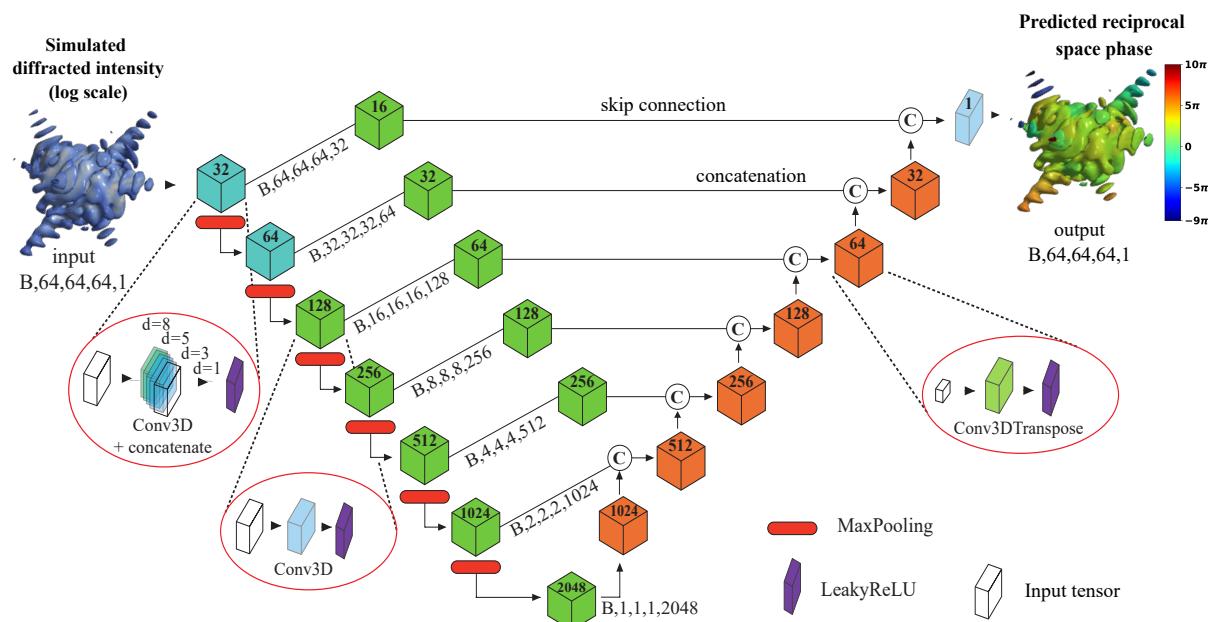


Figure 5.30: Schematic of the 3D U-Net model employed for the RSP prediction of highly strained patterns.

95'000 simulated BCDI patterns have been created following the procedure described in Sec.4.4.1 on a cubic 64 pixel-sided grid for different particle's shape, strain distribution, oversampling conditions and noise levels. Another smaller dataset containing 4'000 samples was created instead for testing the model.

The model was trained with the WCA loss function for 60 epochs with a learning rate of 10^{-4} . To speed up the process, the training has been conducted using two NVIDIA TeslaV100-SXM2-32GB GPUs using the MirroredStrategy feature for synchronous training across multiple devices provided by Tensorflow library. This measure allowed to reduce the training time from 2 hours per epoch to 30 mins.

5.8.1 Results: simulated data

Once completed the training, the model has been tested on simulated data. Here, Figs.5.31 - 5.32 illustrate some examples of comparison between ground truth and predictions for both the RSP and the reconstructed objects respectively.

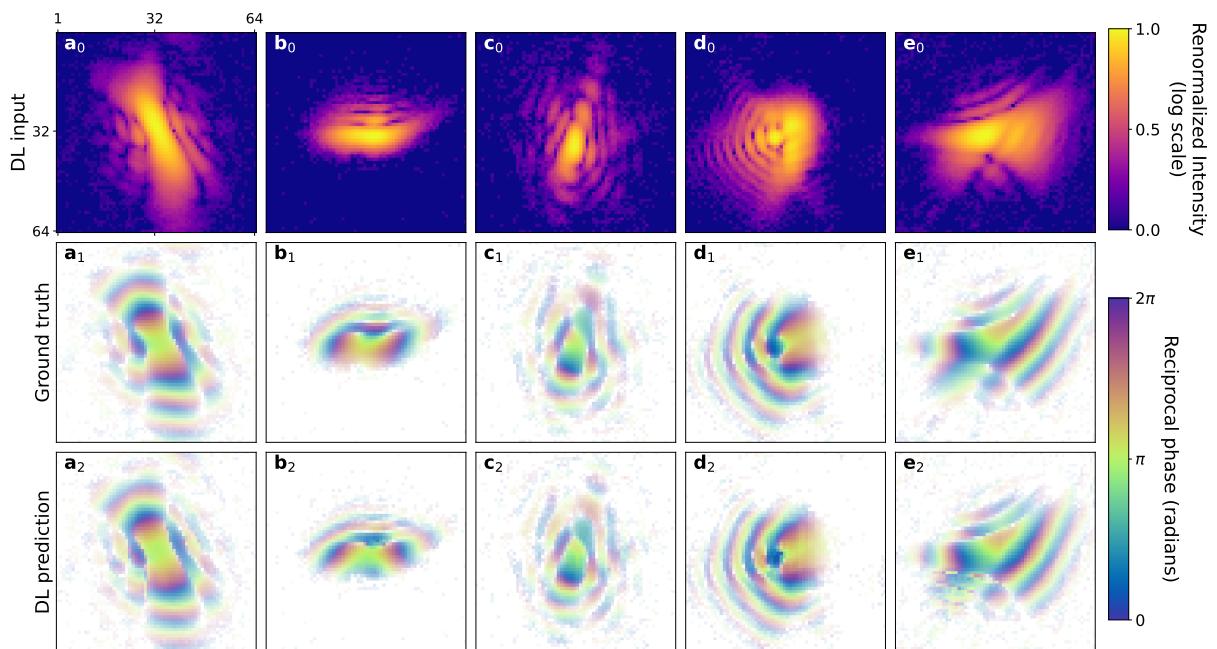


Figure 5.31: $a_0 - e_0$) Central slices of simulated input intensities from the test dataset. $a_1 - e_1$) Corresponding ground truth RSP. $a_2 - e_2$) Corresponding slices taken from the Deep Learning (DL) model prediction of the RSP.

The model correctly predicts the RSP oscillations inside the fringes also when these have been distorted or merged into a single continuous intensity stripe (Fig. 5.31 a-b) because of the high strain along the corresponding axis in real space.

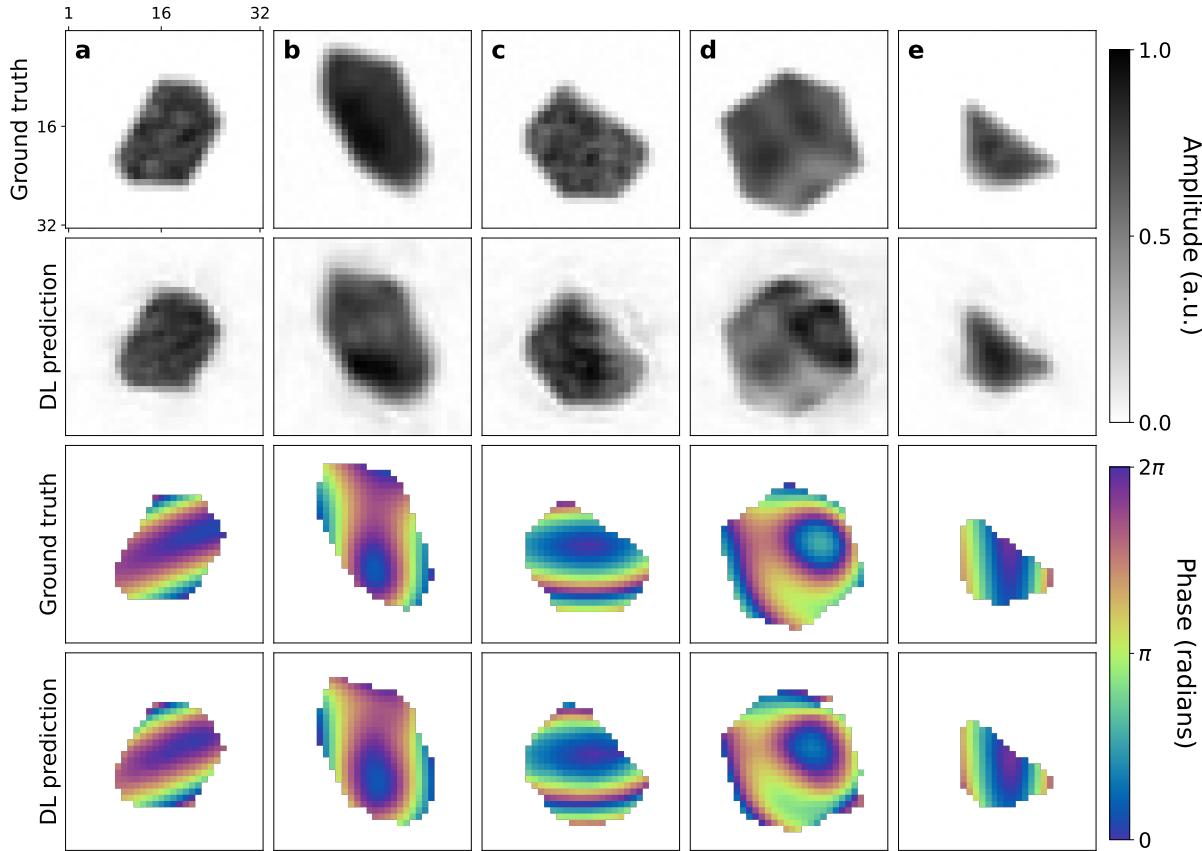


Figure 5.32: Reconstructed objects from the diffraction patterns in Fig. 5.31. **a-e)** Central slices of both the modulus and phase for the ground truths and the DL reconstructions.

Except for some noise and inhomogeneities in the objects' moduli, the reconstructions from the predicted RSP achieve good accuracy on simulated data, for different particle shapes and strain distributions. It is worth mentioning that the presence of noise affecting the objects' shapes is an effect of the loss function that is not computed in real space. The model is never directly shown that the real space object are compactly supported, as it comes as consequence of the correct RSP prediction. However, while small discrepancies of the RSP prediction result in noise on the object shape, an overall accurate RSP prediction ensures the retrieval of both the correct shape and phase of the object. This fact is of primary importance when considering the use of the DL prediction as starting point of iterative refinement with conventional algorithms. It is indeed easier to reach convergence from a low resolution noisy but accurate estimate of the object rather than a clean but inaccurate one.

5.8.2 Results: experimental data

Given the satisfactory results obtained on simulated data the DL model was tested on experimental data as well. Two relevant examples of BCDI patterns collected at the ID01 beamline of the ESRF-EBS are considered here. The first pattern (Fig. 5.33 a) is given by a platinum nanoparticle on Yttria-stabilized zirconia (YSZ) (Particle 1) while the second (Fig. 5.35 b) is a dewetted platinum/palladium bilayer on a sapphire substrate (Particle 2).

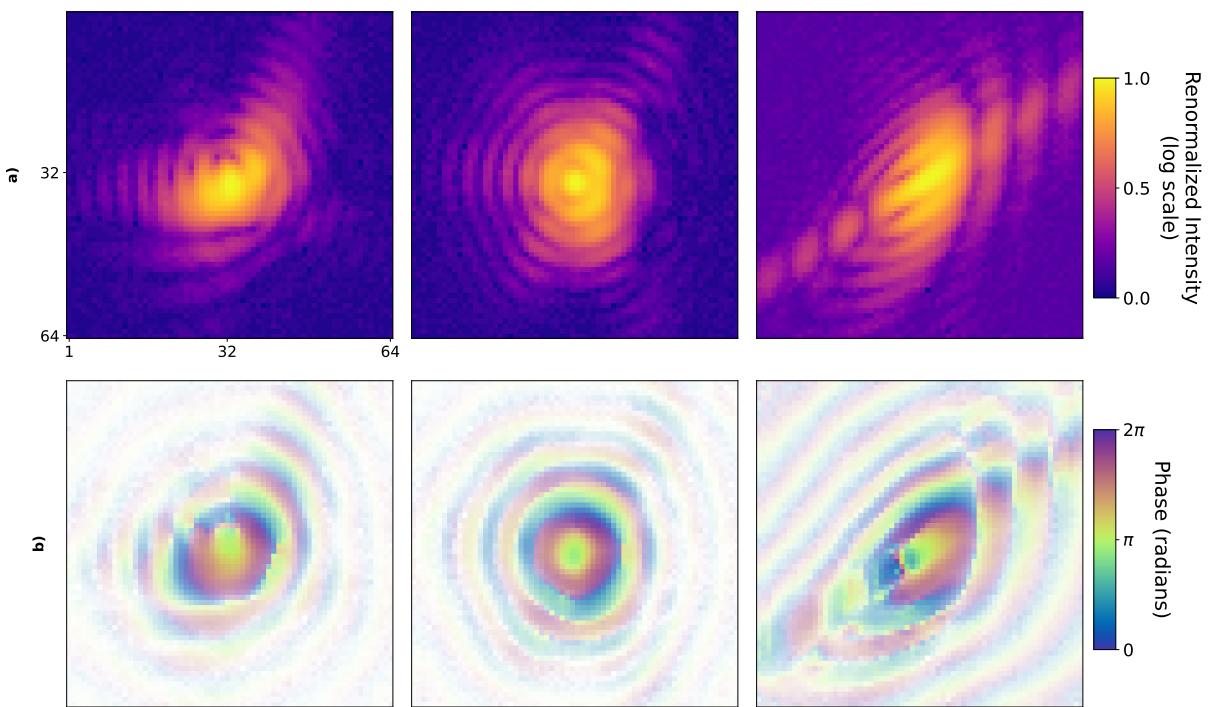


Figure 5.33: **a)** Central slices of the experimental diffraction pattern from Particle 1. **b)** Corresponding slices of the DL model RSP predictions.

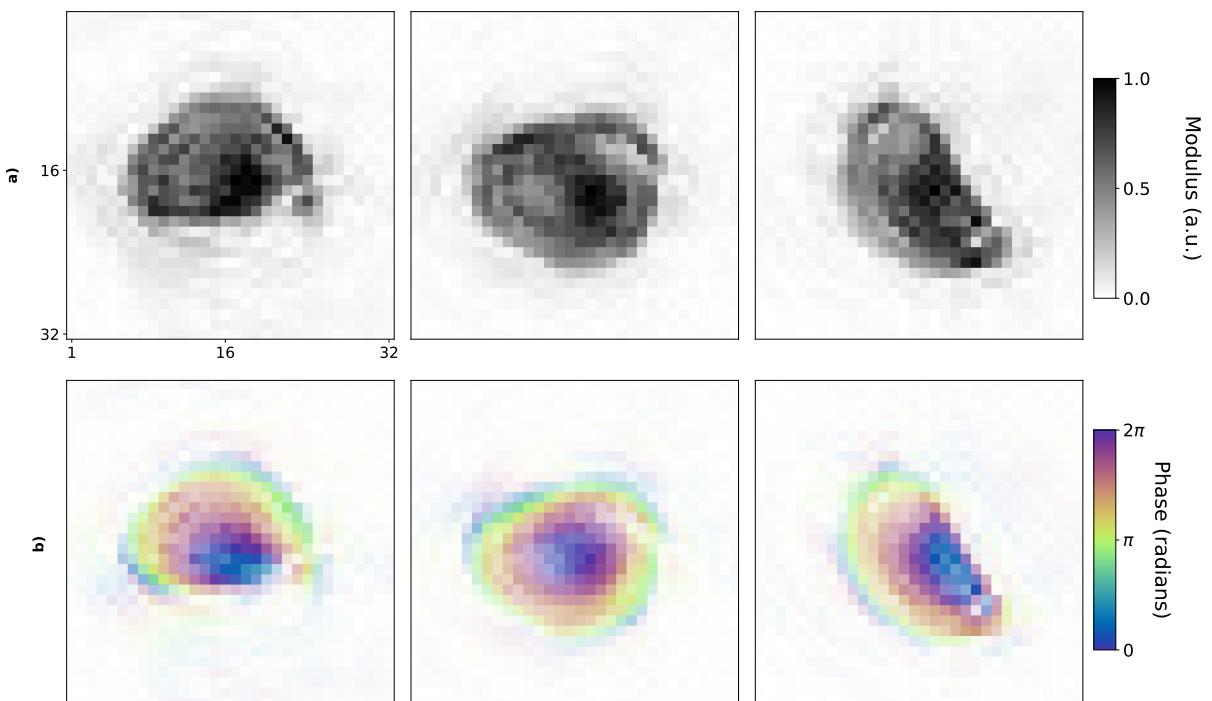


Figure 5.34: Central slices of the reconstructed Particle 1 modulus **(a)** and phase **(b)**

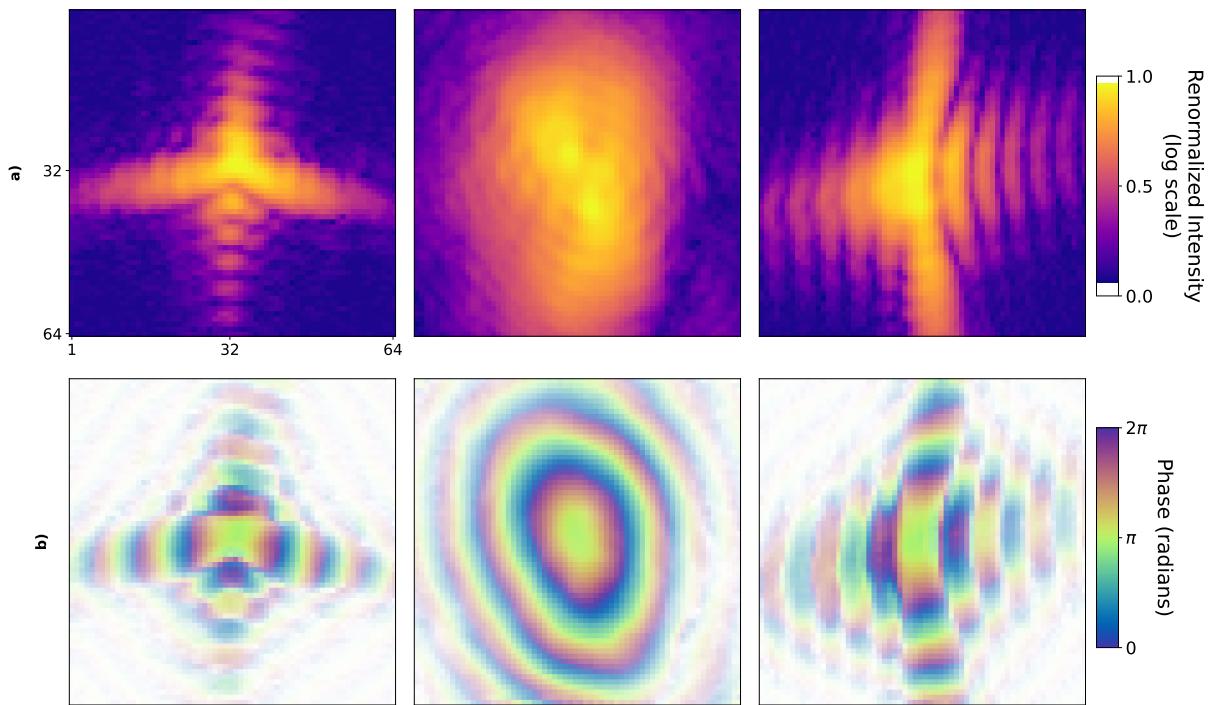


Figure 5.35: a) Central slices of the reconstructed Particle 2. b) Corresponding slices of the DL model RSP predictions.

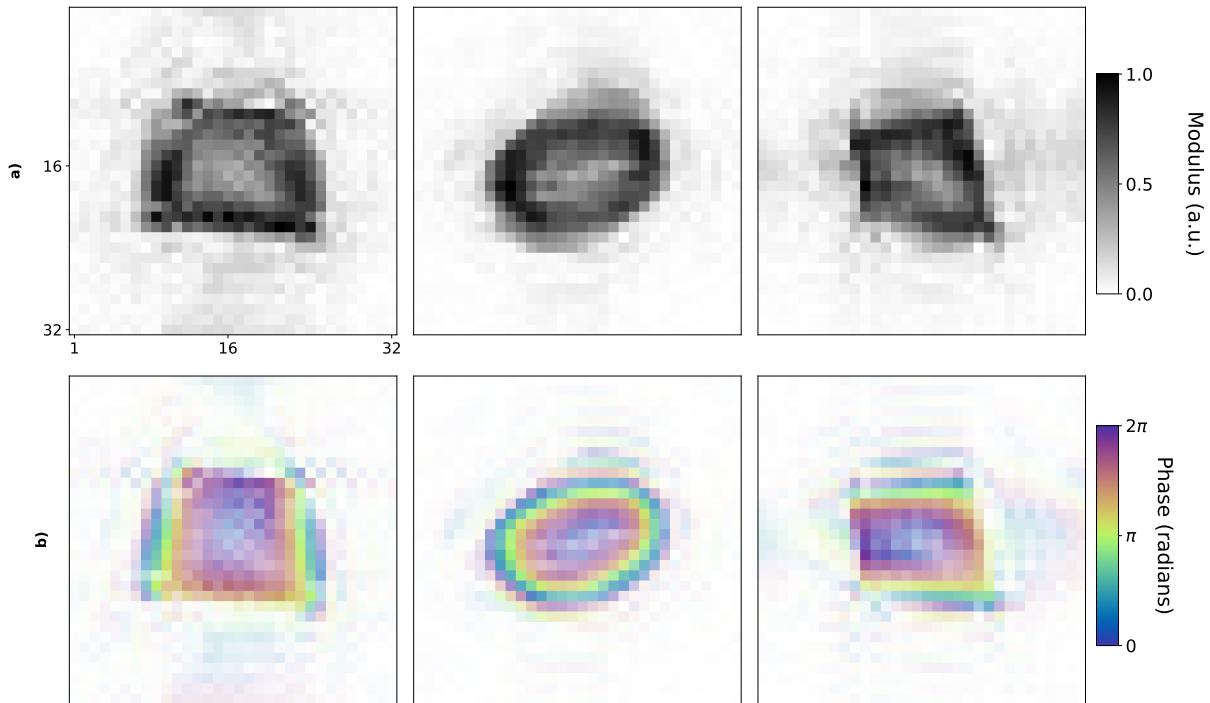


Figure 5.36: Central slices of the reconstructed Particle 2, modulus (a) and phase (b)

Fig. 5.34 and 5.36 show the reconstructed object obtained from the predicted RSP. Despite the low resolution and the presence of noise it is possible to recognize the shape of realistic particles and phases.

In these cases of PR of experimental data it is impossible to establish a comparison with a ground truth because not available. In fact, 60 independent runs of standard iterative algorithm have been launched in order to reconstruct these datasets but no satisfactory results were obtained. Precisely, the recipe of 400 HIO + 1000 RAAR + 300 ER and different thresholds for the support estimation were used for each run. Moreover, the 3 best results according to the Free LLK metric [134] were combined with mode decomposition to improve the quality of the reconstruction. However, in both cases the obtained reconstructions were not satisfactory as holes were present (Fig.5.37), or the support was excessively shrunk (Fig.5.38).

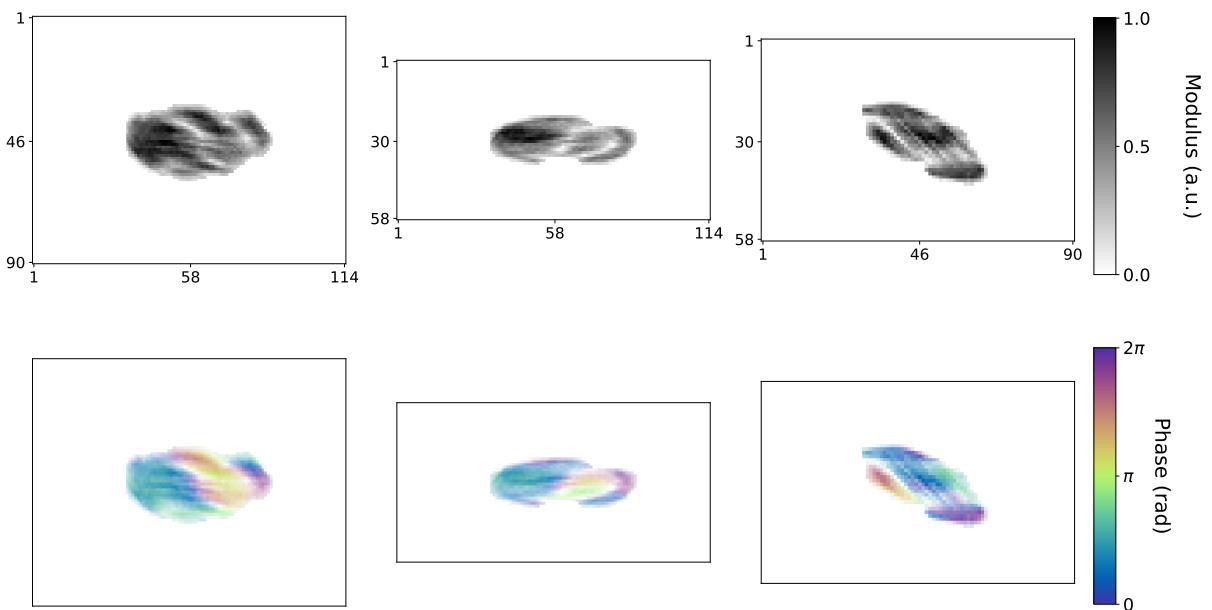


Figure 5.37: Combination of the 3 best reconstructions of particle 1 out of 60 independent runs with PyNX (central slices). Although the shape is guessed the presence of holes in the modulus denotes a poor quality of the result.

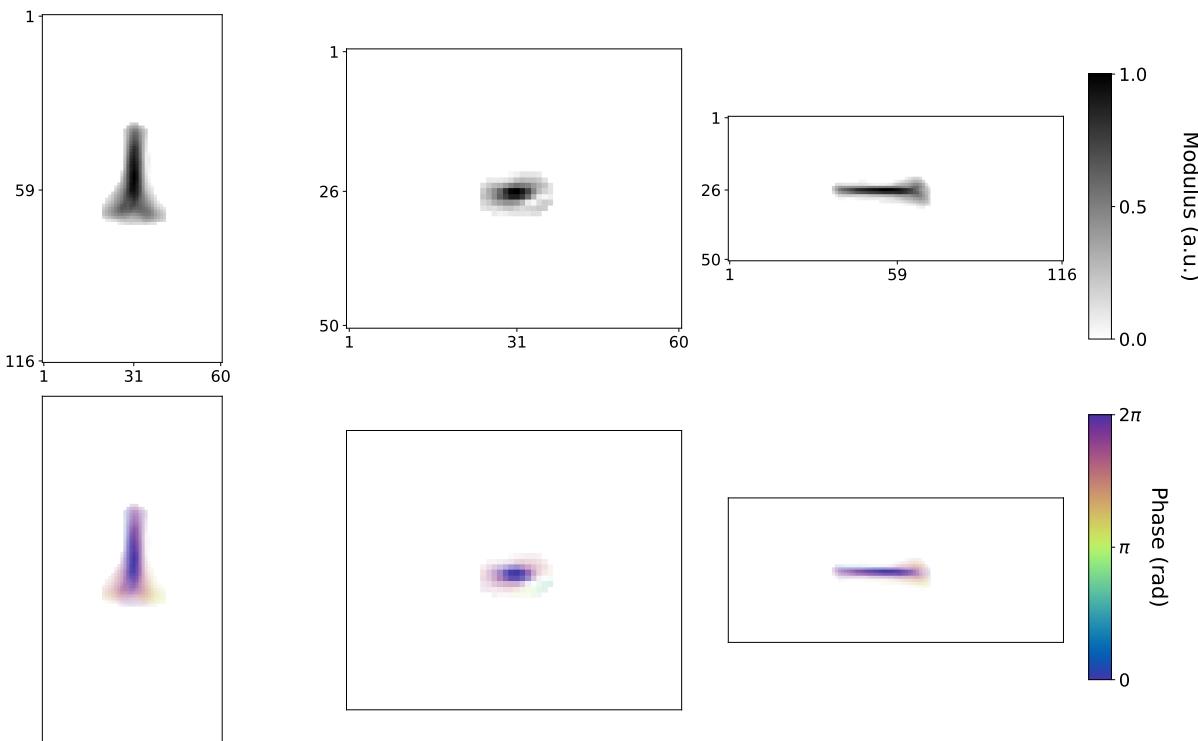


Figure 5.38: Combination of the 3 best reconstructions of particle 1 out of 60 independent runs with PyNX (central slices). The high strain induced by the substrate deceives the standard PR algorithm that excessively shrinks the object's support along the axes perpendicular to the highly strained ones.

5.9 Refinement with iterative algorithms

The use of the DL model for the phasing of experimental data has to be intended as a pre-processing step that can provide a starting point for further iterative refinement. Classical ER algorithms can be used to polish the DL estimate, simplifying the convergence. At this stage it is worth mentioning that, since the DL model was trained on 64 pixel-size cubic volumes, it was necessary to crop and resize the experimental diffraction patterns. This procedure requires special attention as for different cropping/resizing parameters the DL prediction varies. In particular, it is curious to notice that the cropping/padding operations in reciprocal space correspond to interpolations in real space and vice versa. For this reason the procedure illustrated in Fig.5.39 had to be adopted to adapt the dataset to the DL format and then bring back the predicted object to the size relative to the original dataset one. This step is fundamental when the DL object is then used as starting point for further refinement with classical algorithms. Because of the large diversity of the region of interest (ROI) from one experimental dataset to the other, the manual intervention is for the moment needed to adjust the cropping and binning parameters such that the data fed into the DL model resembles the simulated data used for training.

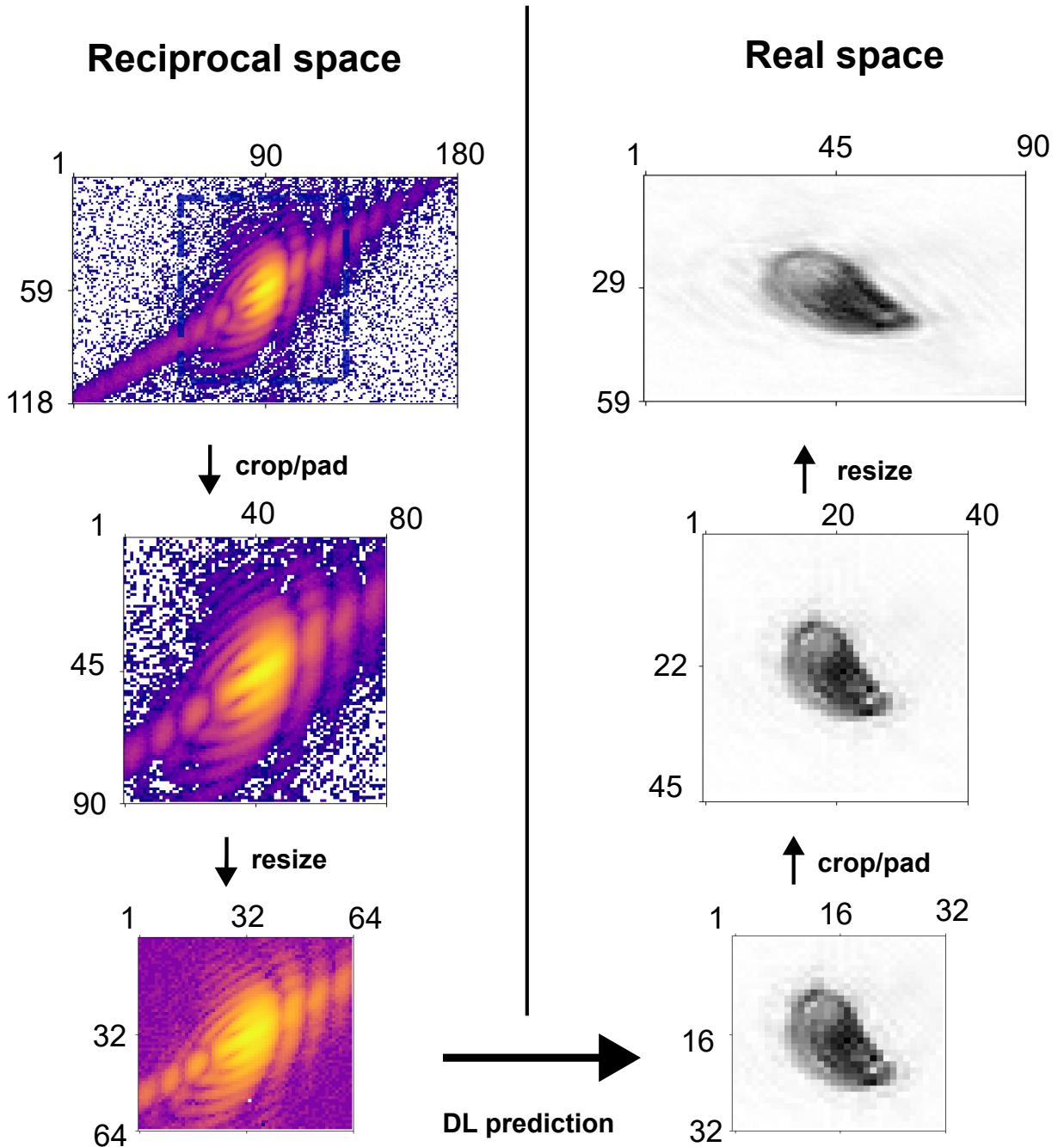


Figure 5.39: Manipulation of the datasets. Here the experimental BCDI pattern is firstly cropped around the COM of the Bragg peak, cutting out parts of the signal. The data is then interpolated into a 64 pixel sided cubic grid and transformed in normalized log-scale. The object obtained after the DL RSP prediction is then padded and resized back to the original shape such that it can directly be plugged into an iterative algorithm for refinement.

If the DL predicted object is a good low-resolution guess of the solution, few steps of ER are usually enough to reach convergence. ER is indeed the simplest alternating projection method and converges linearly to the local minimum [79] so it represents the optimal choice when the DL prediction places the starting point around the minimum. In this specific case 300 iterations of ER were performed and, since the estimated DL object is assumed to be close to the solution, only the pixels at the border the support were allowed to be updated. Figures 5.40 and 5.41

show the results obtained after refinement presented above.

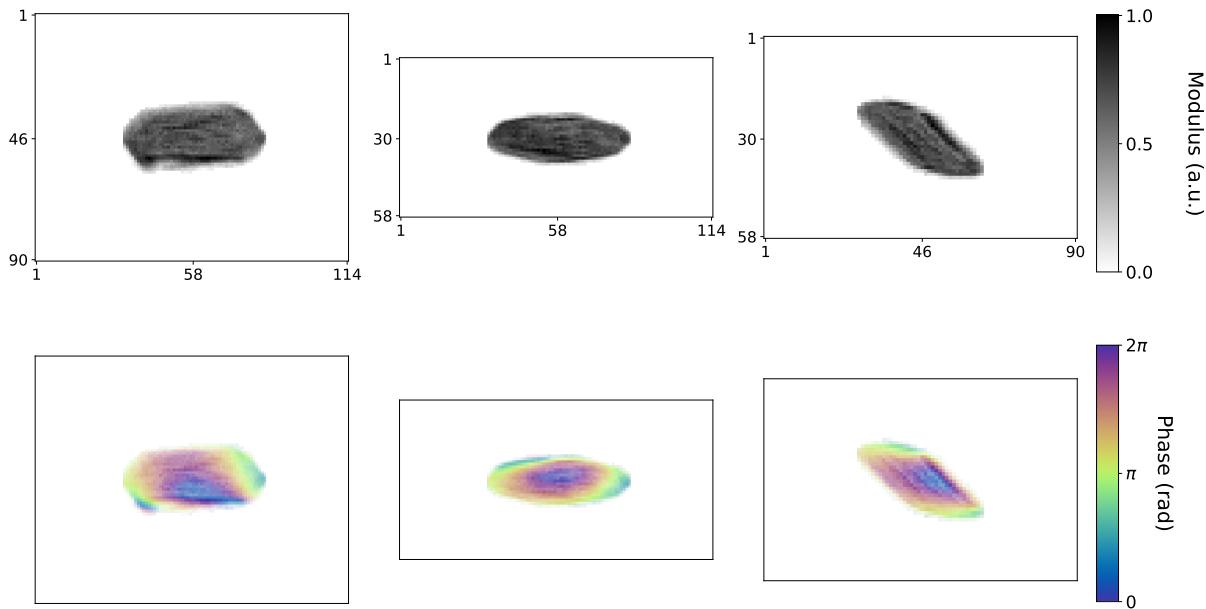


Figure 5.40: Central slices of Particle 1 after 300 cycles of ER for refinement of the DL initial estimate.

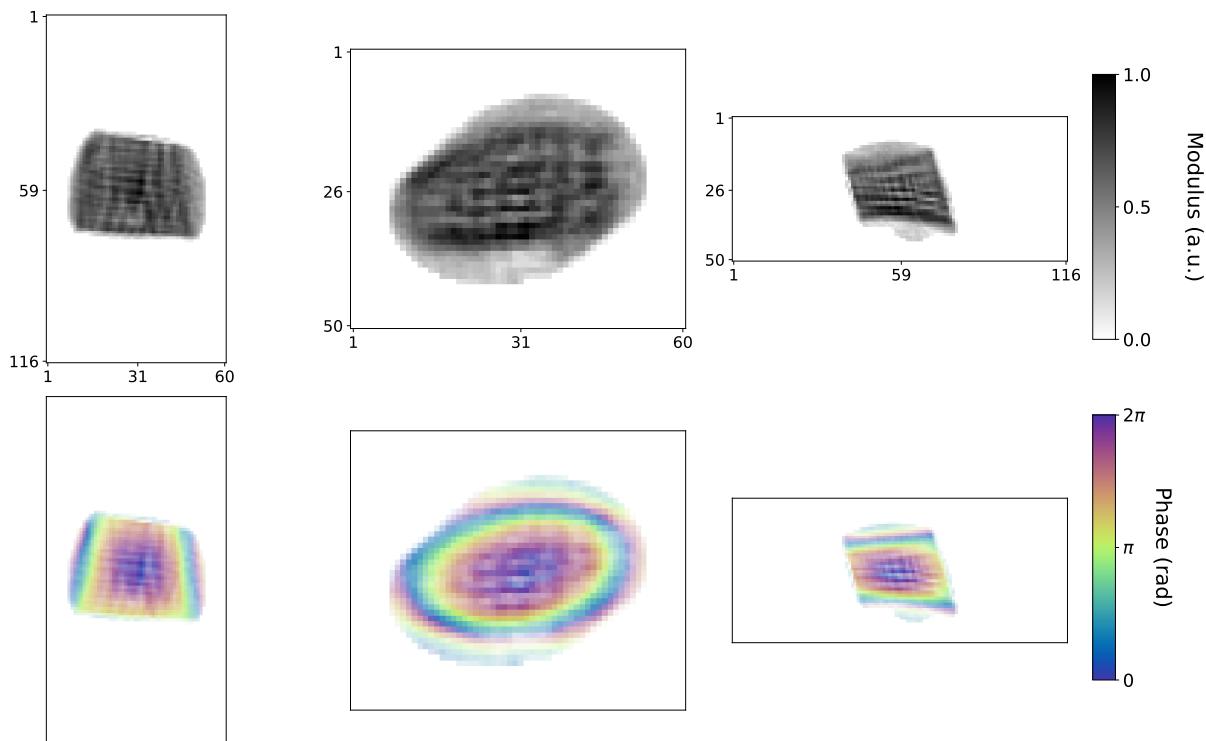


Figure 5.41: Central slices of Particle 2 after 300 cycles of ER for refinement of the DL initial estimate.

The improved homogeneity of the modulus and the more physical shape of a Winterbottom particle are a clear sign of better reconstructions for both cases. One can observe that the shape and phase of the final reconstruction are not remarkably different from the DL estimates. This proves that the DL model is able to generalize correctly to experimental data as well and that

the following ER steps are a good choice for refinement.

While for Particle 1 it is obvious that the DL + PyNX solution is better than the PyNX one because of the absence of “holes” in the modulus, for Particle 2 it can be harder to believe that the DL + PyNX approach actually yields an improved solution. For this reason it is worth calculating the diffracted intensity from the reconstructed object and its fidelity with respect to the observed one. Here, Fig. 5.42 shows that the calculated intensity obtained from the DL + PyNX is more accurate than the one obtained from the PyNX - only solution.

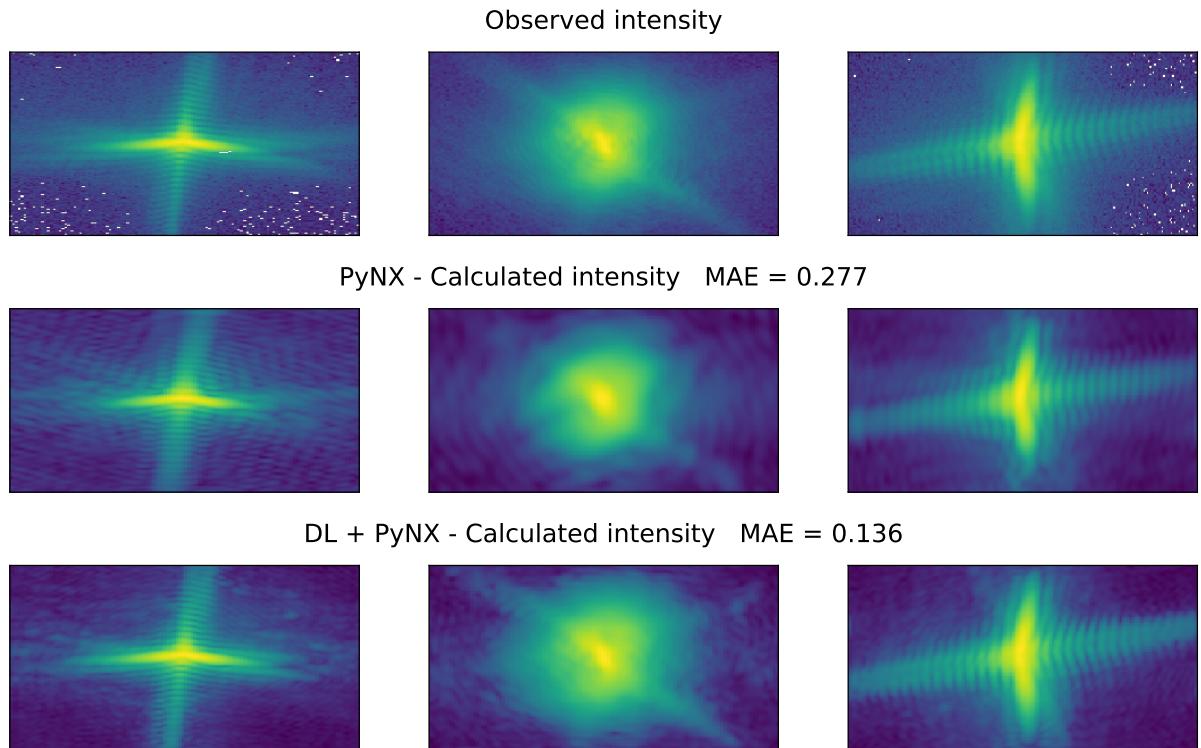


Figure 5.42: Projections along the three axis of the observed and calculated diffraction patterns relative to Particle 2. The mismatch (MSE) of the one calculated from the particle reconstructed using PyNX only, is higher than the one calculated from the particle obtained with the DL + ER method. This proves that the latter is the correct solution.

Beside the higher quality of the results it is worth mentioning the significant reduction of the wall-clock time. Table 5.2 summarizes the time taken for the 3 different methods. A 40X to 50X speed-up is recorded for these dataset sizes and the time saving increases for larger ones.

5.10 Performance assessment

In this paragraph the results of the DL model presented above, tested against different strain configurations and magnitudes are discussed. The scope of the study is to assess the model’s performances for these different cases, to evaluate when it works best and ultimately to estimate appraise the gain in accuracy when coupled with ER refinement. The first test was prepared simulating a Winterbottom shaped particle, similar to the ones used for the training set, with an

	Particle 1	Particle 2
Data size	(118,180,230)	(110,160,200)
DL inference	3.04 s	2.21 s
PyNX: 60 runs	227.41 s	123.55 s
DL + PyNX: 1 run	4.73 s	3.08 s

Table 5.2: Computation times for the three different methods. DL RSP prediction and inverse FT are considered for DL time. PyNX - only time includes 60 runs of 400 HIO + 1000 RAAR + 300 ER performed in parallel by PyNX. The best reconstructions selection and following mode decomposition is not considered. DL + PyNX time includes the full DL time plus the reshaping of the initial guess to the original size and a single run of 300 ER.

applied phase built with two Gaussian functions with two different and increasing amplitude ranges. The corresponding BCDI pattern has been simulated for each case, keeping the same oversampling ratio and noise level across the simulations. For each calculated diffraction pattern the RSP has been predicted using the DL model and corresponding objects have been obtained with inverse FT. At this point the accuracy of the prediction was calculated with using the formula in 5.5 and the results are shown in Fig.5.43.

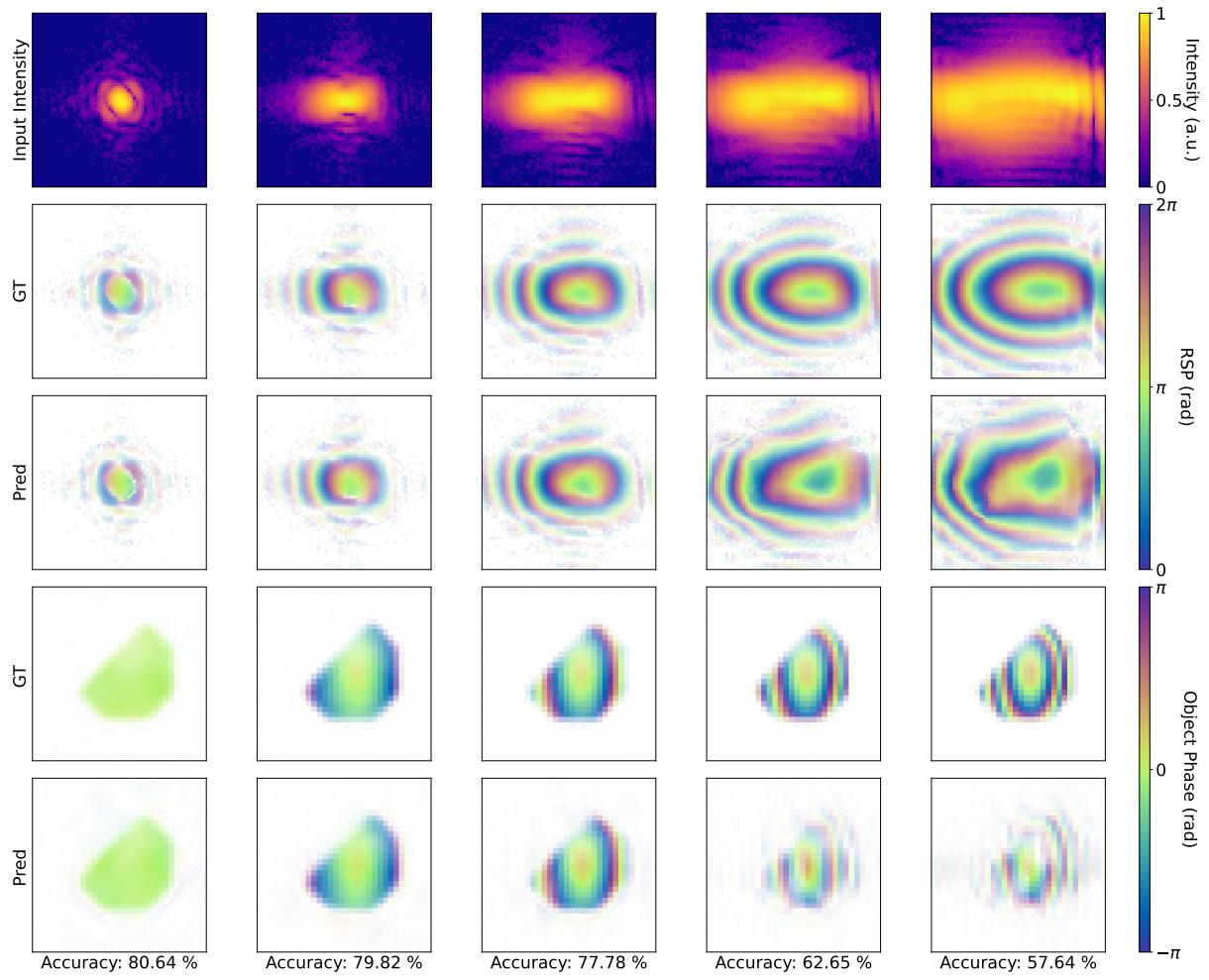


Figure 5.43: Evolution of the BCDI pattern and corresponding RSP and object for higher strain calculated with the sum of two Gaussian functions of increasing amplitude. Third and fifth row show the DL predicted RSP and the reconstructed object’s phase respectively. As expected the prediction worsens for high phase ranges but it overall maintains a similarity with the ground truth.

As expected the accuracy drops as the phase range increases. It is interesting to notice that despite the worsening of the prediction with the increasing strain the object’s phase structure inside the support recalls the ground truth phase. This detail is fundamental for the ER refinement as the initial guess, notwithstanding the inhomogeneous and “shattered” support, represents a good estimate of the solution. Objects with similar cleaner supports of incorrect shapes and phases are much worse starting point for iterative refinement since far from the solution.

The same procedure has been repeated for a different strain configuration, this time constructed with two cosine functions with increasing amplitude. The phase distribution induced by these functions is rather different, more in the spatial structure than the amplitude. Fig. 5.44 reports the results showing that the model accuracy is significantly poorer than the previous case. It is worth reminding that the training set was composed of equal amounts of particles simulated with Gaussian and cosine phase profiles, meaning that the lower accuracy scores are not due to some possible imbalance of the training. The phase range inside the particle, that can be estimated visually by the amount of wraps inside the support, is also in the same order

of magnitude for both cases meaning again that the reason for the poorer results is of other nature.

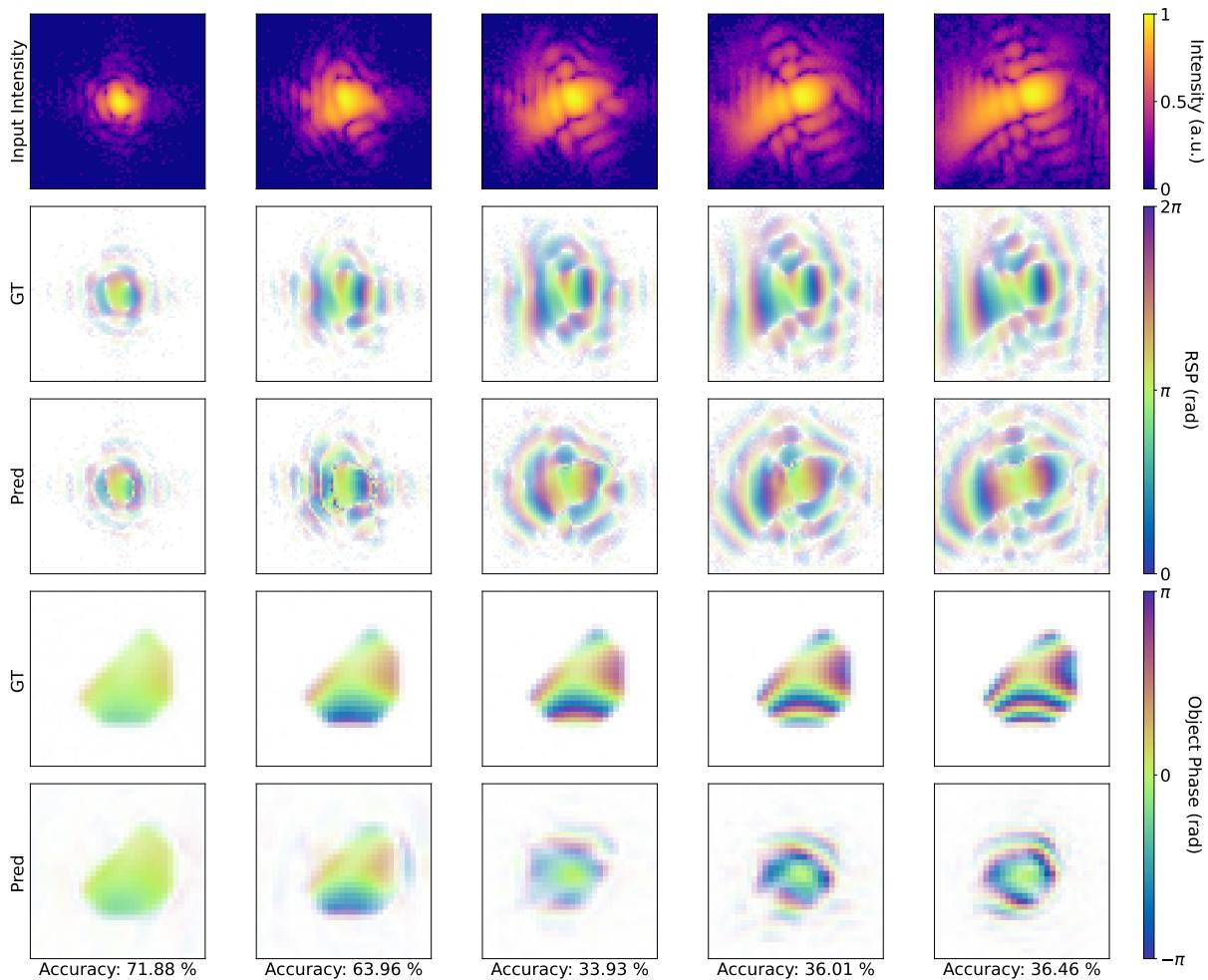


Figure 5.44: Evolution of the BCDI pattern and corresponding RSP and object for higher strain calculated with the sum of two cosine functions of increasing amplitude. Third and fifth row show the DL predicted RSP and the reconstructed object's phase respectively. The model rapidly struggles with this type of phase field.

Similarly, the model seems to struggle more for phase fields simulated with using Gaussian correlated random field (same as in 4.4.1) than for the first case. Again the reason doesn't seem to be related to the different phase range nor population imbalance in the training set (Fig. 5.45)

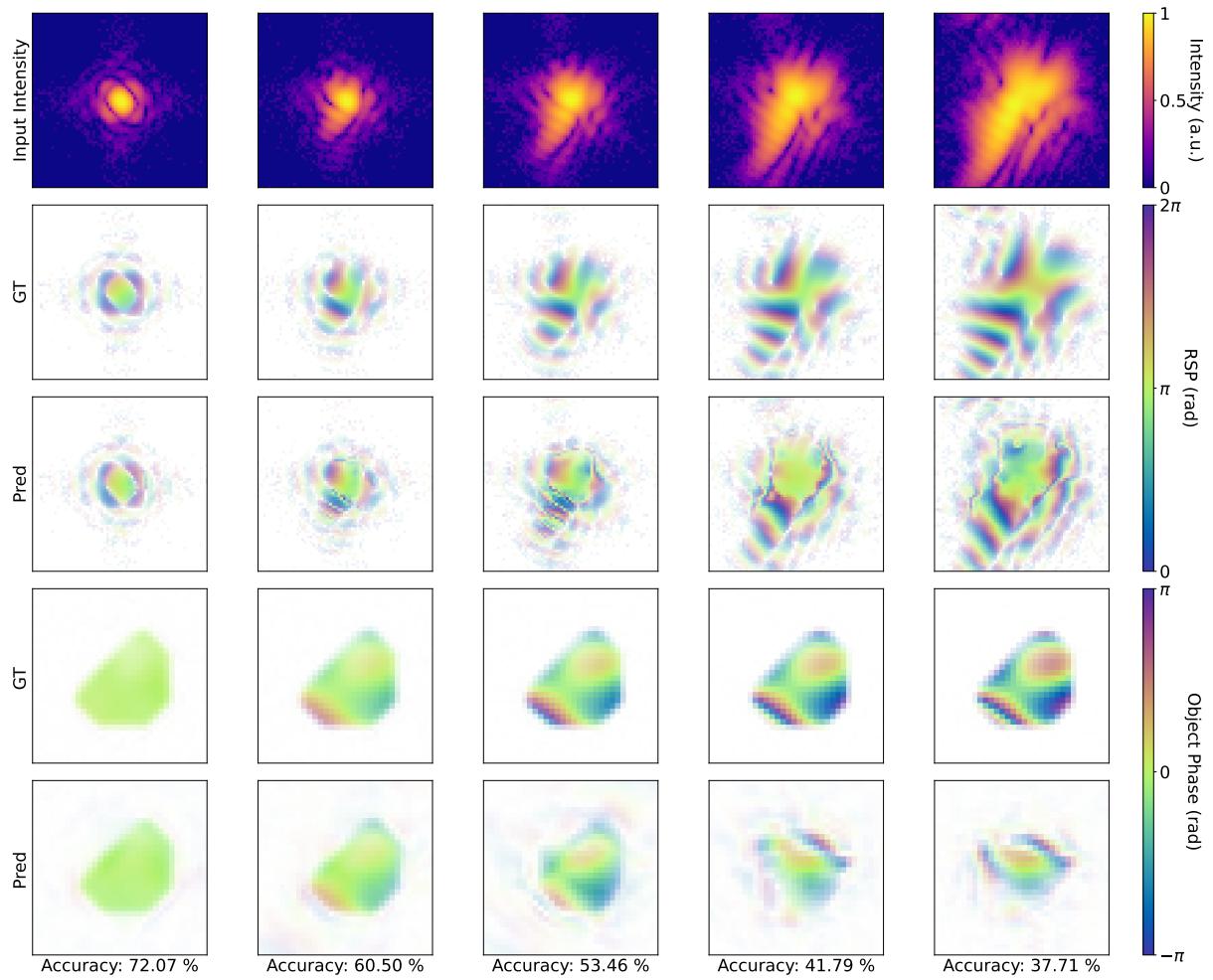


Figure 5.45: Evolution of the BCDI pattern and corresponding RSP and object for higher strain applied by using a Gaussian correlated random field of increasing amplitude. Third and fifth row show the DL predicted RSP and the reconstructed object's phase respectively.

To better assess the performances of the model on the three different cases, the same procedure has been repeated on a bigger and more diverse population. Namely, 20 randomly shaped particles have been simulated and for each of them an increasing strain field has been applied in 10 different steps, for the three type of distributions. The results of the model are shown in Fig. 5.44

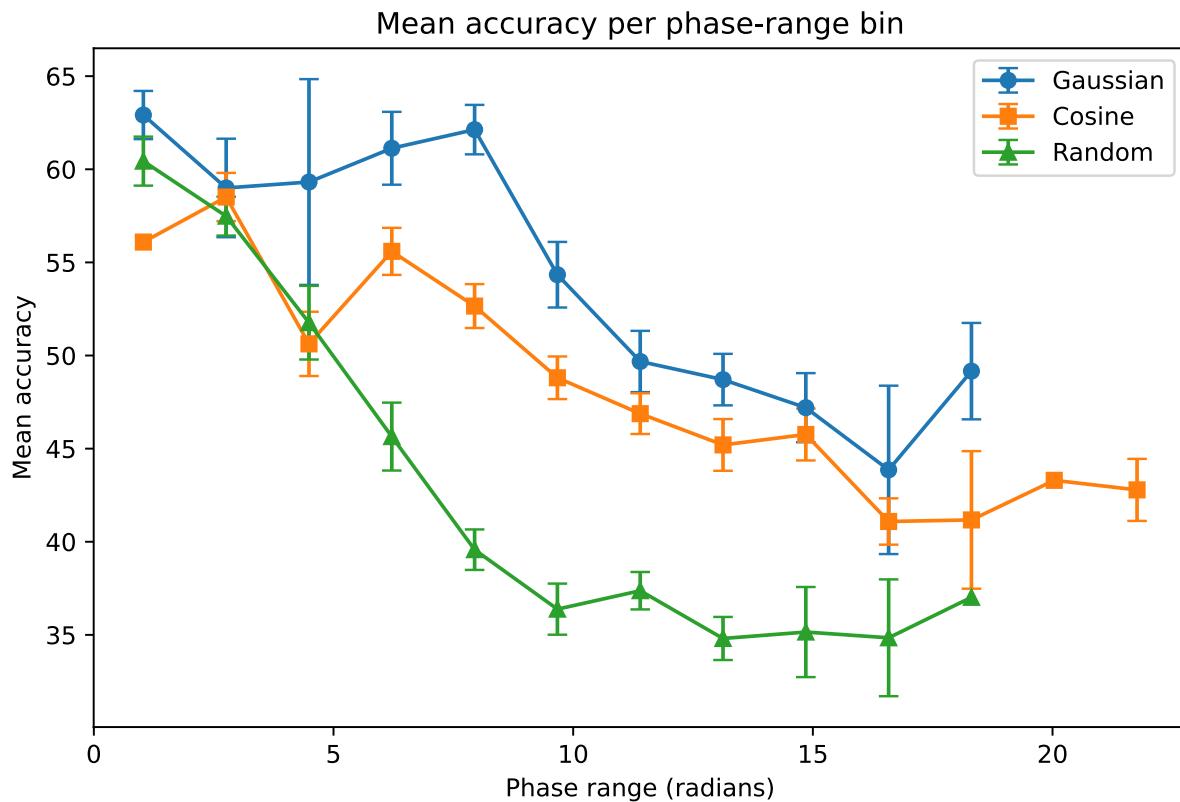


Figure 5.46: Mean accuracy of the DL model for different phase ranges and types. The phase field simulated with two Gaussian functions yield RSPs that are better predicted by the DL model than the ones obtained with two cosine functions or random Gaussian fields.

Another study that has been conducted aimed at estimating the accuracy gain of the DL model coupled with ER refinement compared to the sole iterative algorithm. In this case a single Winterbottom particle has been selected for simplicity, and an increasing phase field was applied in 50 steps for the three different types. At this point the accuracy scores have been calculated for the DL model only first. Separately, a single run of 400 HIO + 1000 RAAR + 300 ER has been performed using PyNX for each of the 50 diffraction patterns. At last, the DL model predicted objects have been used as starting point for 300 ER refinement, always with PyNX. In all cases, it was made sure that the solution found by either the DL or the iterative algorithm wasn't the twin of the ground truth. If so, the object was flipped before calculating the accuracy.

The analysis has been repeated for the three different phase fields used in the above cases and the results are shown in Figs. 5.47, 5.48, 5.49.

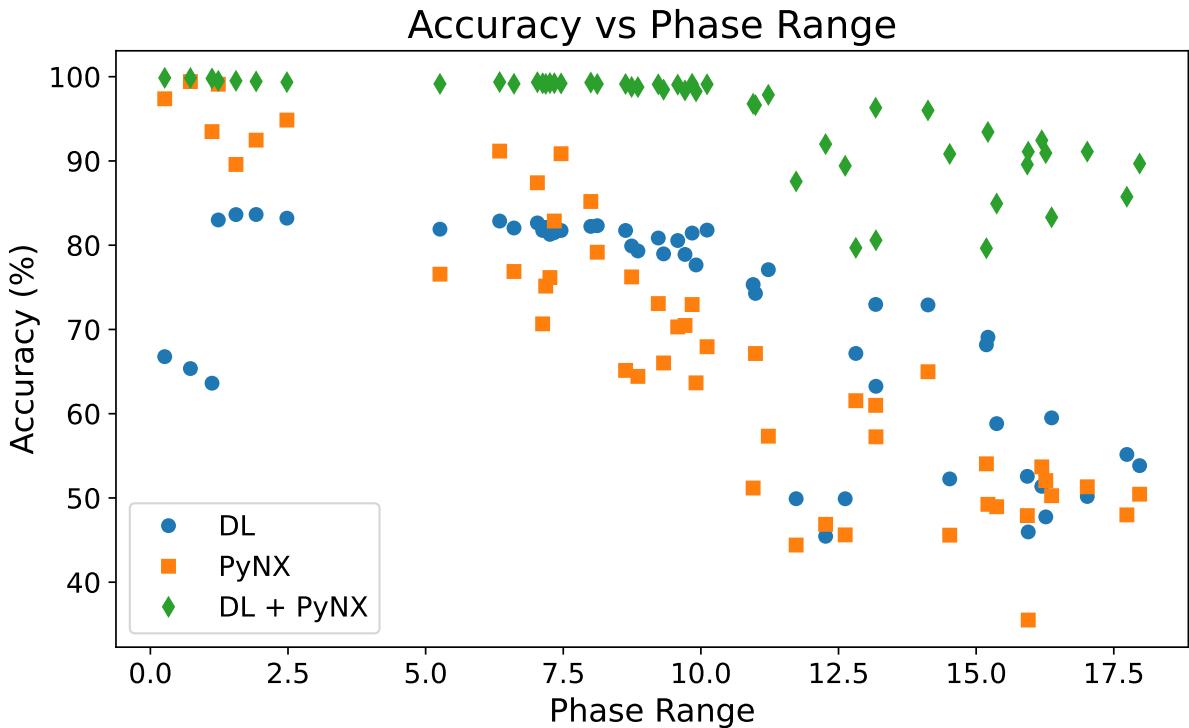


Figure 5.47: Comparison of the accuracy scores of the DL model prediction, iterative PR and DL + ER approaches for objects with phase fields simulated with two Gaussian functions with increasing amplitude. The DL + ER reconstructions always achieve higher accuracies than the other methods, also in those cases in which the DL ones are low.

While for lower phase ranges the DL prediction is worse than the two other methods, the main result of Fig.5.47 is that in all cases the accuracy of the DL + ER method is always superior, meaning that the DL model can significantly aid the PR process also when the first estimate is not very close to the solution (down 50% accuracy in the figure.).

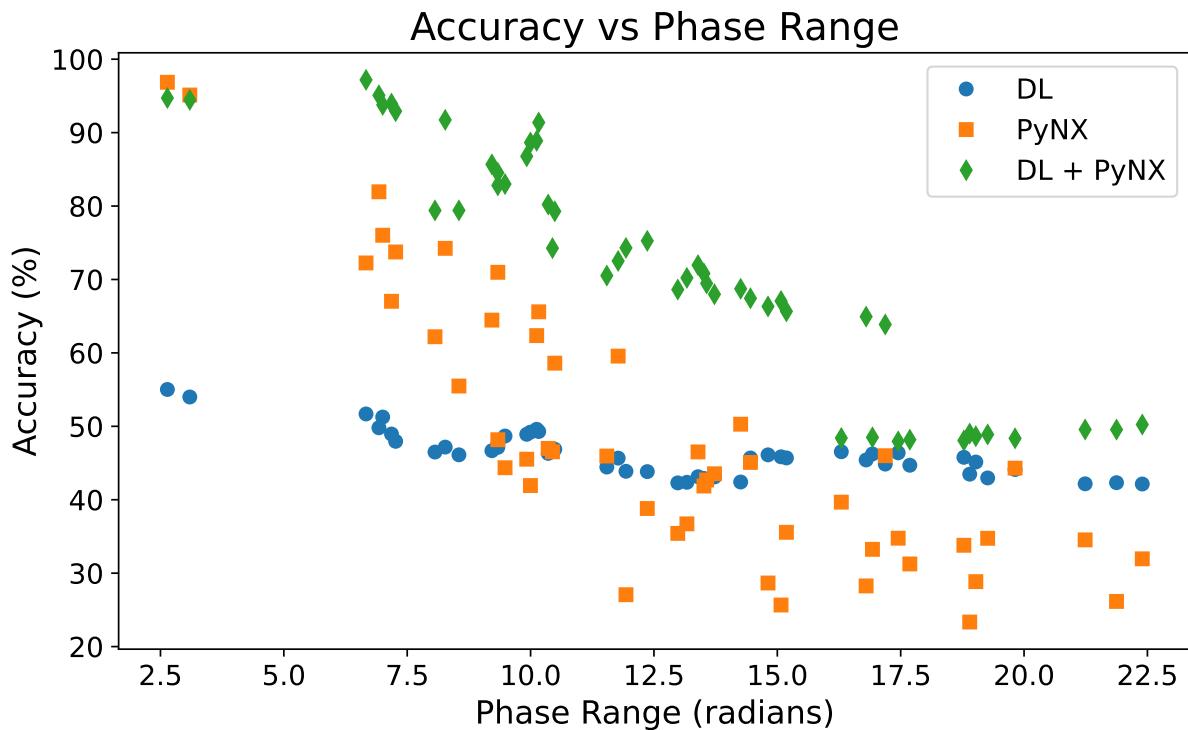


Figure 5.48: Comparison of the accuracy scores of the DL model prediction, iterative PR and DL + ER approaches for objects with phase fields simulated with two cosine functions with increasing amplitude.

Similarly, the DL model proves to be significantly helpful at providing a good initial guess for the ER refinement when the object's phase is simulated with cosine functions.

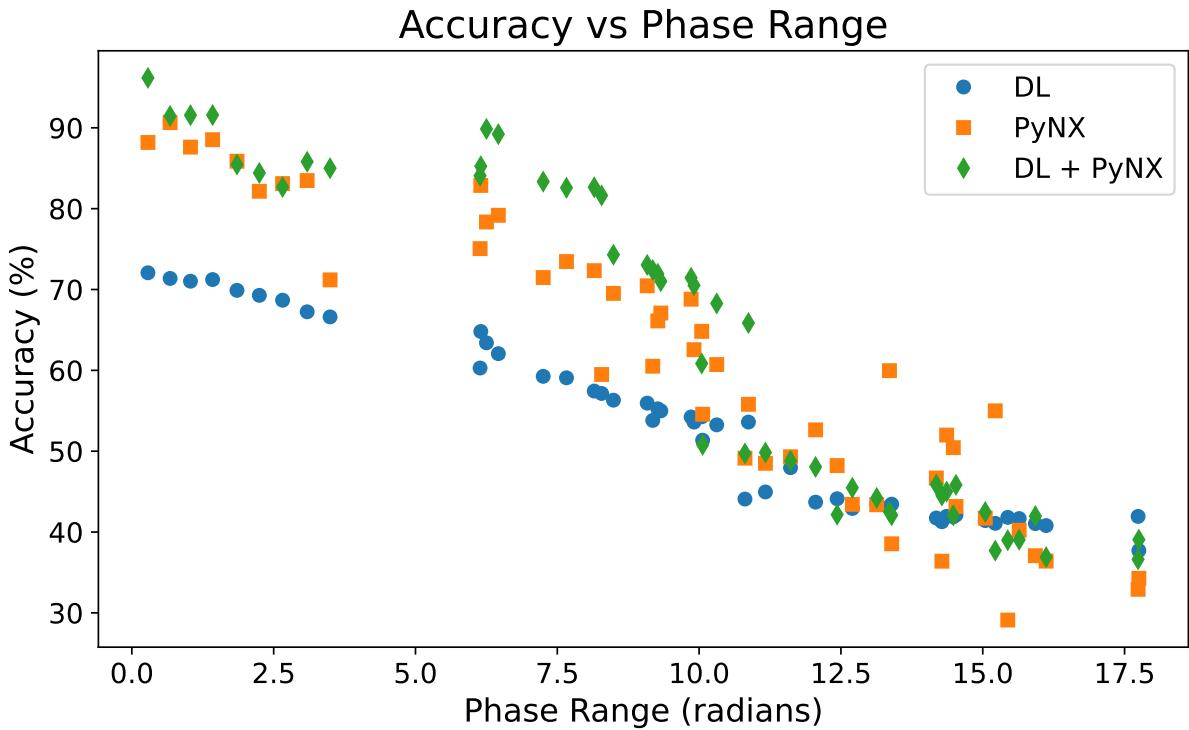


Figure 5.49: Comparison of the accuracy scores of the DL model prediction, iterative PR and DL + ER approaches for objects with phase simulated with a Gaussian correlated random field with increasing amplitude. The combination of DL + ER yields better results up to a phase range value of 10 radians. For higher values the poor DL prediction does not provide a good enough estimate of the solution thus it does not help the convergence of the ER method.

In this case as well, the DL model, although when used alone yields relatively low accuracy scores, improves the quality of the solution when coupled with some ER cycles for refinement. At the same time it is interesting to notice that when the accuracy of the DL prediction is too poor - most likely when, as in Fig.5.45, the object's phase differs too much from the ground truth one - the ER cycles do not improve the result as the initial estimate is wrong.

As last study on the performances of the DL model, the reasons behind the imbalance of accuracy scores for different phase fields are investigated on a qualitative level. From the visual assessment of the DL prediction one can observe that the model performs best when the RSP is characterized by “iso-phase” lines with spherical symmetry. This effect was already observed in the 2D case in which the WCA loss managed to overcome this limitation. In the 3D case this effect is more pronounced and the model still struggles to break this structural symmetry despite the large number of trainable parameters and training samples. A possible explanation is given by the symmetry of the diffraction pattern used in input. The averaged spatial intensity distribution of the ensemble of training samples possess a spherical symmetry, decaying with a power law radially. As a consequence, the model replicates a similar distribution in the predicted RSP. This result is clearer when the direct output of the model, in the form of an unwrapped RSP, is inspected.

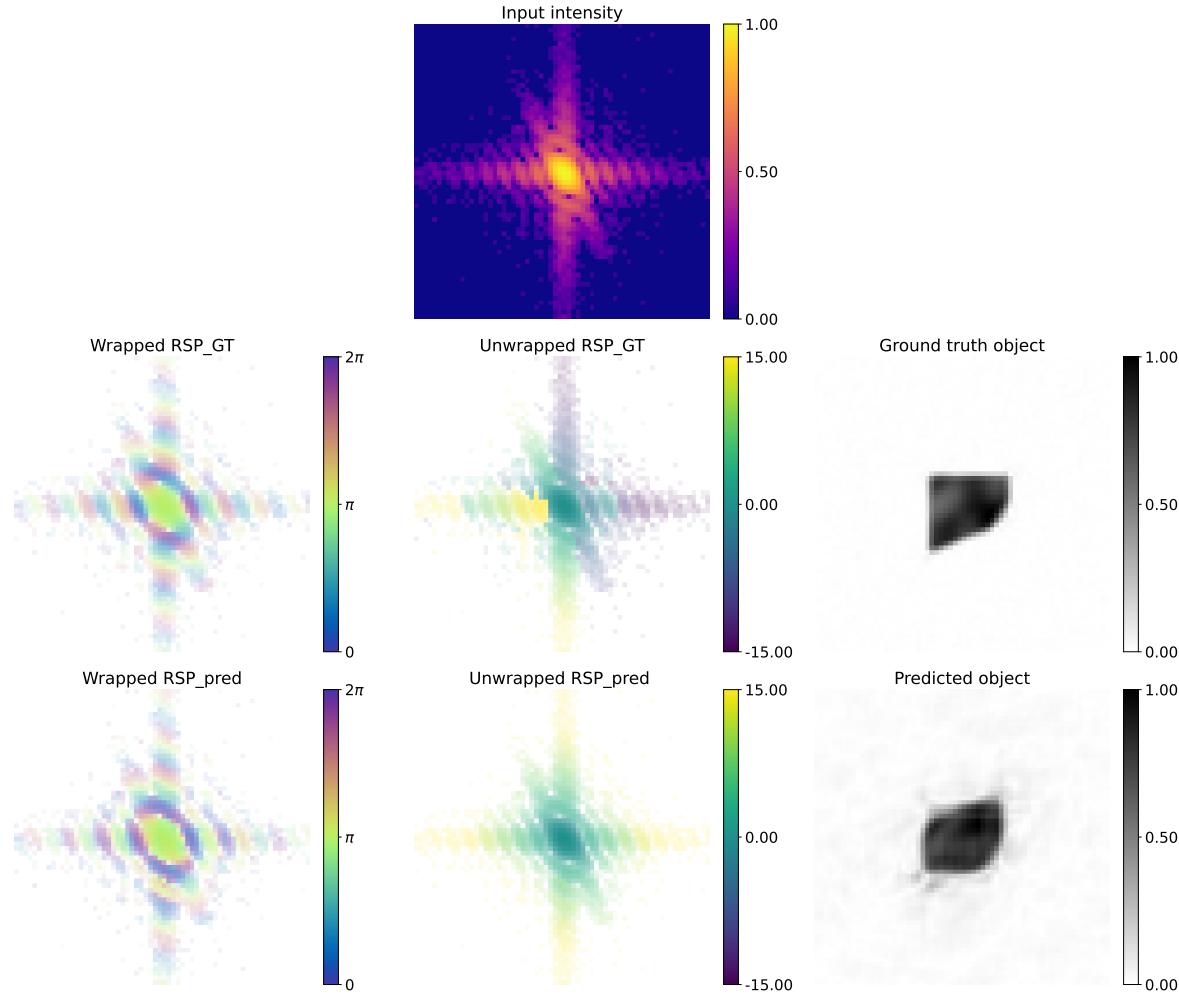


Figure 5.50: Example of DL prediction for a low strain randomly shaped particle. The non-centrosymmetry of the object's shape and the small strain produce a symmetric diffraction pattern with a RSP that when unwrapped possesses a non-spherical symmetry. This deceives the model that wrongly predicts a symmetric RSP, resulting in an incorrect reconstructed object.

When comparing the unwrapped phases one can observe that while the ground truth, along the main streaks, grows monotonically from one to the other end, the predicted one, which is the direct output of the model, grows isotropically along all radial directions. The consequence is a wrong result of the retrieved object as visible in Fig. 5.50. It is worth noticing that the example reported in the figure is a low-strain case, meaning that the main obstacle for the model is given by the structural symmetry of the RSP rather than the strain. A support of this thesis, to the same object a stronger phase obtained with two Gaussian functions has been applied and the same test has been conducted. The results illustrated in Fig. 5.51 show that the RSP symmetry has changed to a more “spherical” one thanks to the strain. Therefore, the predicted RSP is more accurate as well as the reconstructed object’s shape.

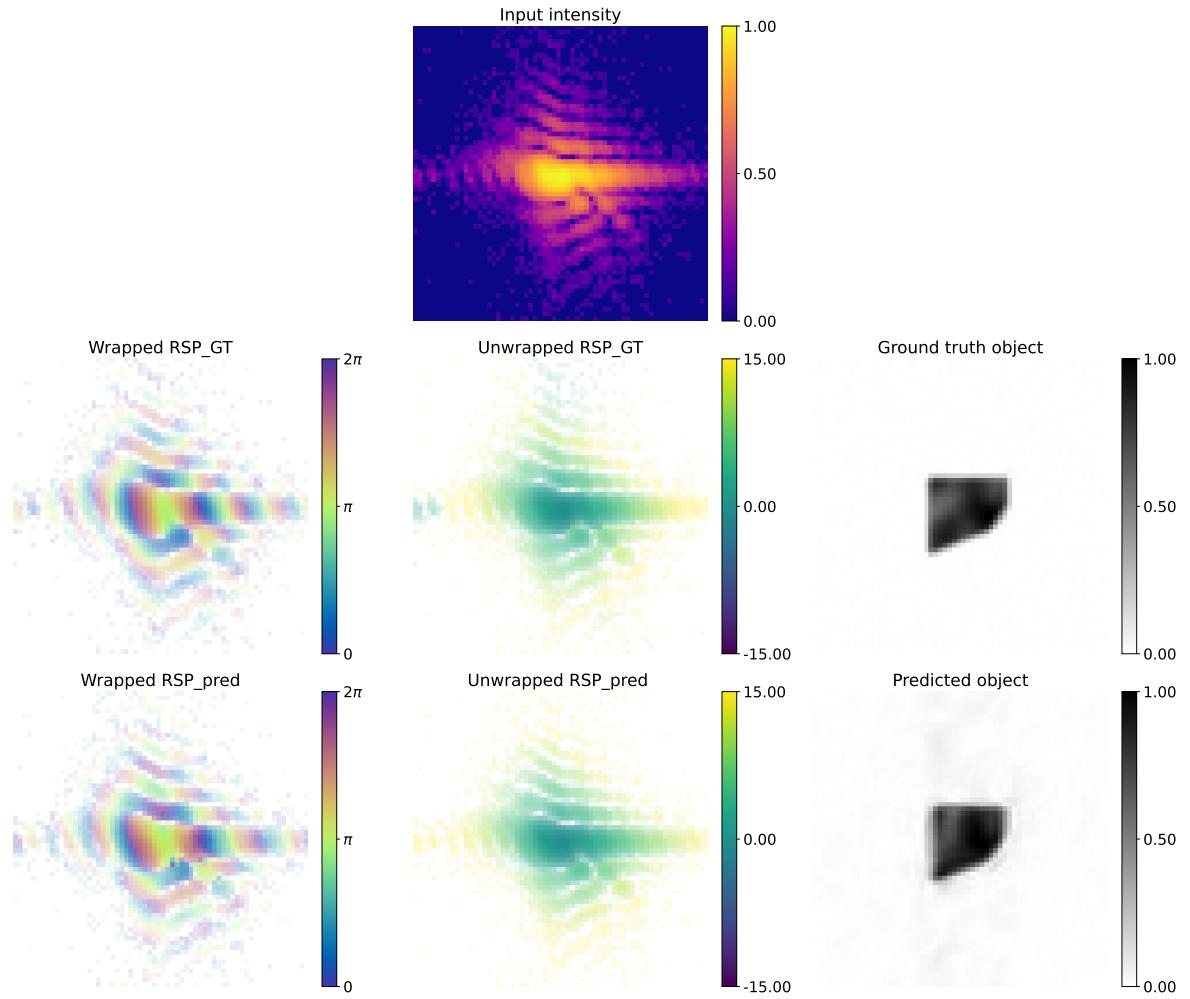


Figure 5.51: A stronger phase has been applied to the same object in Fig. 5.50. The strain field produces a more symmetric RSP that is correctly retrieved by the DL model, thus a better object is retrieved despite the higher strain.

5.11 Other model test

In this last section I would like to assess the real advantage of predicting the RSP rather than the complex real space object. In order to conduct this experiment, the most recent model in the literature of Deep Learning for BCDI Phase Retrieval was considered. In particular, the model presented by Yu and coauthors in [129], that makes use of complex convolutional layers seem to be a good candidate for the testing of our DL model. At this scope, the 2D model that was presented in the paper was transformed into a similar one for 3D data, for a total amount of 36 million trainable parameters.

The model has been trained for the same amount of epochs (60) on the same training dataset used for our DL model. Model structure and loss function were adopted like presented in the paper and the tests have been performed on the same data shown in Fig. 5.32.

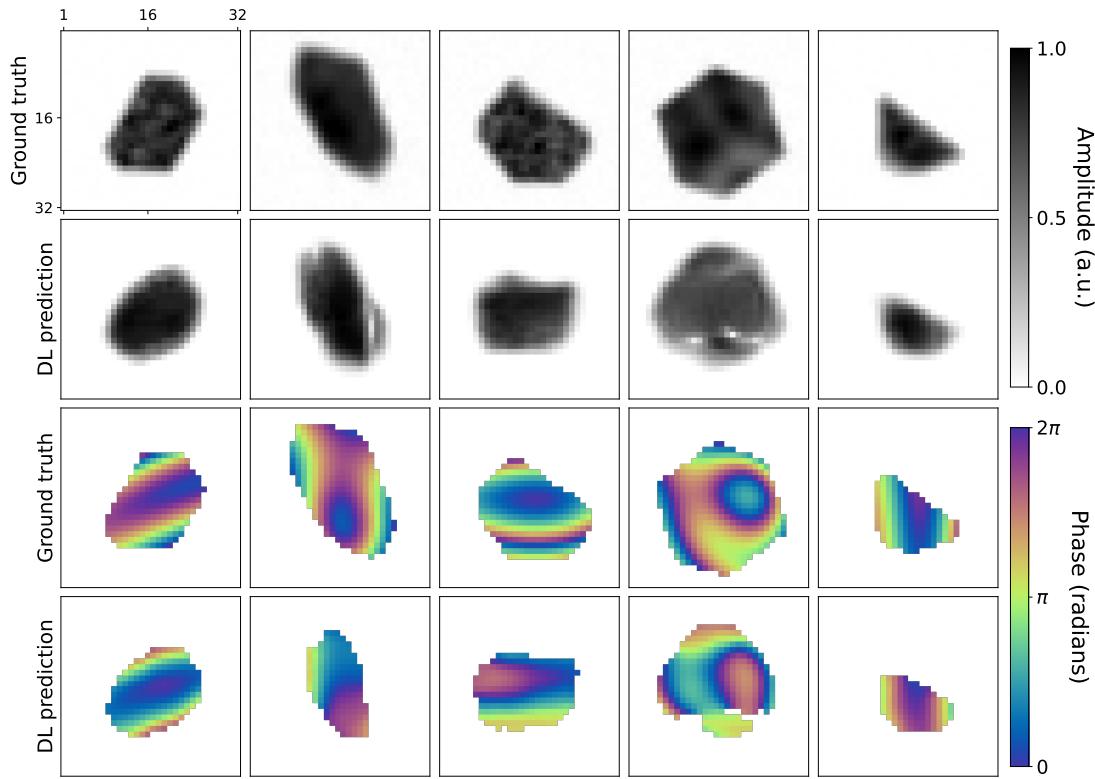


Figure 5.52: Predicted objects obtained by the 3D adaptation of the 2D Complex U-Net presented in [129]. The model returns a complex tensor that represents the real space solution. Although much less noisy than our predictions, the reconstructed objects possess incorrect shapes and phases.

The model returns objects that have a better resolution and less noise levels because of the loss function calculated in real space. However, the shape and phase of the results are not correct, thus unusable for ER refinement because misleading initial guesses. This simple example shows that the prediction of the RSP suites better the PR of BCDI patterns with convolutional neural network, especially for highly strained particles.

5.12 Conclusion

To conclude this journey through the developments, results and interpretations of the DL model for the BCDI RSP prediction the main points can be summarized as follows.

- A novel approach for the DL-based PR of BCDI patterns, focused on the prediction of the RSP has been investigated. The method proved to be advantageous since (i) it entails the inference of a single array rather than two coupled ones, (ii) it exploits the exact calculation of the IFT, directly using the diffraction pattern in the transform, rather than a learned map from the diffraction pattern and the solution in real space. (iii) it takes

advantage of the similarity between the input and the output variables, making the best use of the skip connections of the U-Net architecture. The superiority of this approach over the more conventional one found in literature has been demonstrated as discussed in 5.11.

- The training of the DL model for the prediction of the RSP can be optimized with the use of the custom WCA loss function, designed specifically for the handling of complex phases. Adopting the WCA loss not only improves the results but is also speeding up the training as it takes shorter time than a loss computed in real space. Moreover, the use of the WCA avoids any reference to the real space object during the training, opening the door for a patching approach, where the RSP is predicted from a sub-volume of diffracted intensity.
- Such designed model is trained in supervised fashion on simulated data only. This can be at first a limiting factor as it requires the simulation of big and diverse datasets. However, being the BCDI technique restricted to single crystals, the population of shapes, though being potentially infinite, is physically confined. It is thus easier for the DL model to generalize for new particle's shapes.
- The DL model trained as presented is able to perform on highly-strained particles as well, showing promising results in 2D and 3D. It has proven to be successful inverting experimental data that happened to be extremely challenging with conventional iterative methods only. This achievement marks a milestone for the BCDI community, upgrading the DL studies on BCDI PR from a “*proof of concept*” level to an actual *practical use* in the analysis of experimental data.
- The DL model proved to be compatible with iterative refinement using conventional algorithms, contributing to a faster and more robust pipeline for PR.
- The DL model exhibits certain shortcomings attributable to the intrinsic symmetry of RSP. It in fact “prefers” RSPs with a spherically symmetric distribution and the major hypothesis for this fact is that such symmetry is typical of the intensity distribution of the average diffraction pattern. Hence, the difficulty of the model to escape this bias for 3D data more than 2D ones. Additionally, it is curious to notice that this symmetry seems to be the strongest limitation for the DL model, lying above the sign symmetry and the high strain, which are both resolved when in many cases. Future investigations that elucidate the root causes of this phenomenon may yield improved performance on more heterogeneous data sets.
- At last, the substrate-induced strain imposed on Winterbottom particles often manifests a symmetry that is faithfully reproduced by the pair of Gaussian functions used to generate the phase of the training ensembles. This symmetry gives rise to a spherically symmetric RSP, which the DL model, by design, generally predicts with high accuracy.

AUTOMATIC DIFFERENTIATION FOR BCDI PHASE RETRIEVAL

“An approach that would be superior to the ones considered here would be one that minimizes the Fourier-domain error while inherently satisfying the object-domain constraints, or one that minimizes an error metric that combines the Fourier- and object-domain constraints [...]. Something along these lines would be very useful for the problem of a single intensity measurement; clearly, more could be done in this area”

J.R.Fienup [2]

In this chapter a different approach to the BCDI phase retrieval will be presented. It originated from the need to resolve those cases in which neither standard alternating algorithms, nor the DL assisted PR can succeed to converge to a satisfactory reconstruction. The developed approach differs from the alternating projections algorithms classically used for the Fourier PR, as it is formulated as minimization problem solved with gradient descent (GD). The gradients however are computed through the efficient automatic differentiation (AD) enabled by graph-based differentiable programming packages like Tensorflow and PyTorch, accelerated on GPU. For this reason one could see the AD approach as unsupervised machine learning on a single training dataset.

The GD - based optimization is fundamentally different from fixed point alternating projections. Here one could qualitatively say that if the latter switches between real and reciprocal space applying constraints in both, the former initializes a complex object and updates at each cycle its modulus and phase using the gradients, with respect to them, of the differences between the observed and calculated diffracted intensities. In this way, the knowledge on the particle can be

implemented by initializing the object with some physical constraints or adding regularization terms that will drive the updates towards more reasonable solutions.

6.1 State of the Art

AD methods for PR have been investigated already in 2014 by Jurling and Fienup [135] who first considered the use of AD for GD-based PR. In this theoretical work the authors proposed a pedagogical “manual automatic differentiation” approach for the phase problem, extended to complex-valued variables. The authors also denounced the lack of suitable softwares as major limitation to the use of AD-based PR. The advent of high-level, GPU oriented, libraries such as Tensorflow, PyTorch, JAX and Autograd has opened the opportunity to efficiently exploit AD algorithms for the phase problem. The first implementations in the CDI field have considered mostly ptychography in forward and Bragg geometries [136, 137] and multi-Bragg CDI [138]. Chronologically, it was firstly Nashed and coauthors in 2017 [136] who opened the field using a Tensorflow AD model for ptychography using the ADAM optimizer. From the same group, Kandel *et al.* in 2019, showed the competitive performance of the AD model when compared to conventional algorithms and extended the model to multi-angle Bragg ptychography on simulated data. In 2023 Maddali and coauthors [138] explored the use of AD methods for multi reflection Bragg CDI. The authors leverage the flexibility of the GD approach by designing a global optimization function that simultaneously accounts for the geometrical and physical constraints related to multi reflection BCDI. More recently Zhou in 2024 [139] and Wu in 2025 [140] developed AD-based PR algorithms that are able to reconstruct large particles, for which a dynamical description of the scattering processes is required. These works again exploit the flexibility of AD-based models for the implementation of a forward model tailored to the specific problem of mixed kinematic and dynamic x-ray scattering taking place in large crystals.

At the moment of writing, there aren’t any published works that aim at solving the phase problem for hardly invertible BCDI datasets.

6.2 Model implementation

In an AD-driven optimization problem some trainable parameters are initialized. In the first basic formulation these trainable parameters can be the values of the voxels corresponding to the modulus m and the phase φ of the complex objects that represents the solution of the PR problem. All of these voxels contribute to the creation of a simulated diffracted intensity pattern via the forward model $I_{calc} = |\mathcal{F}\{me^{i\varphi}\}|^2$. Subsequently, the gradients of a metric (loss function) that estimates the distance between the observed BCDI pattern I_{obs} and I_{calc} are calculated with respect to each of the trainable variables with automatic differentiation. At this point the value of each of these voxels is updated using a chosen optimizer (SGD, ADAM, etc.) and a given learning rate. The Tensorflow library allows for an easy implementation of the trainable variables and loss function and handles gradient operations with predefined methods. It is therefore straightforward to run the optimization as it follows the same structure of a deep learning model, with less trainable parameters and for a single data.

However, such simple formulation of the complex object as mere real-valued variables is

not optimal for a non-linear and non-convex inverse problem such the Fourier phase retrieval. In fact, many non-physical modulus-phase configuration could yield a I_{calc} that is close to I_{obs} . The presence of these local minima is the reason why, in conventional PR, algorithms like hybrid input-output, capable of escaping them, are employed. Moreover, it was shown by Marchesini in [79] that steepest GD and even more sophisticated conjugate GD are more prone to get stuck in local minima, reason why they are not commonly utilized for Fourier PR. However, the active research field of machine learning has brought important advancements in the formulation of efficient and robust optimizers based on stochastic gradient descent with powerful features like Nesterov or adaptive momentum (ADAM [110]). These GD techniques are more robust to local minima, since the gradient is computed on mini-batches of trainable variables rather all of them (stochastic rather than classical steepest GD), and converge faster thanks to the “memory” of previous steps. Additionally, they are often wrapped into handy classes, ready to use, in Tensorflow and Pytorch libraries.

However, to facilitate the convergence the formulation of the complex object to be optimized has embedded some physical considerations that helped to restrict the solution space. First of all, both support and phase built on a 3D grid occupying half the volume of the input BCDI data to account for the oversampling ratio which has to be at least 2 in all directions to ensure invertibility. Additionally, other constraints specifically designed for the object shape and phase were considered.

6.2.1 Object’s shape

The formulation of the object’s shape has started considering the typical crystalline samples that are studied with the BCDI technique and the requirements the modulus of the reconstructed object need to fulfill to be considered a “good solution”. Usually, successful reconstruction show a *homogeneous* modulus, sometimes quantitatively assessed through the mean-to-max metric [141], [142], as in standard BCDI the form factor is approximated uniform across all the scattering sites. Enforcing a homogeneous modulus by construction limits the search space and helps the convergence to the solution¹. It follows that parametrizing the *surface* of the support, and setting to 1 the inside, is much more advantageous than optimizing the full 3D volume. This approach, already proposed by Scheinker and Pokharel in [121], also significantly reduces the number of variables to optimize.

An additional consideration is that the probed samples are crystalline, thus often *faceted* and *convex*. Therefore, one could simplify even more the construction of the object shape by building a certain amount of planes in the 3D space and obtain the support from the volume that lies inside the intersections of all them. This would remove the possibility to have spikes or rough surfaces that might satisfy some local minimum but wouldn’t represent a crystal. Moreover, with this representation the number of trainable variables would be further reduced.

According to this scheme the relevant parameters to be optimized are the angles θ and φ of the spherical coordinates and the length d of a given number N of the so-called *half-spaces*. More formally, the normals n_i for each of the N half-spaces are defined with a pair of (θ, φ) that its orientation in space (Eq. 6.1). Subsequently, only the intersection of those (x, y, z) coordinates for which the dot product with each n_i is smaller than the length d_i is considered

¹This approach of constraining the modulus of the object to be homogeneous was already considered in the literature (see [143])

as support (Eq. 6.2).

$$\mathbf{n}_i = \begin{pmatrix} \sin \varphi_i \cos \theta_i \\ \sin \varphi_i \sin \theta_i \\ \cos \varphi_i \end{pmatrix}, \quad (6.1)$$

$$\mathcal{S} = \bigcap_{i=1}^N \left\{ \mathbf{x} = (x, y, z) \in \mathbb{R}^3 : \mathbf{n}_i \cdot \mathbf{x} \leq d_i \right\}, \quad (6.2)$$

A schematic representation of this construction is provided by Fig. 6.1.

With this approach the user needs to provide a number of half-spaces as hyperparameter meaning that a sort of prior knowledge on the sample can be leveraged in these regards as well. However, this number doesn't have to be precisely the number of facets expected. In fact, a large N is often advised for unknown sample shape such that even roundish objects can be retrieved. In case of well faceted samples the large N is a minor problem as many n_i will be automatically aligned to the same $(\theta_i, \varphi_i, d_i)$ at the cost of some more trainable parameters.

The first drawback of this convex-hull parametrization is that concave objects can't be retrieved. However, these cases are much less frequent in typical BCDI experiment. The second limitation is that this formulation is incapable of modeling defects that would zero the contribution of the object's modulus to the diffraction pattern [144]. A correct BCDI reconstruction of particles affected by this type of defects presents "holes" inside the hull in correspondence of the defect. However, the current model cannot address this type of features as the support is by construction fully homogeneous inside the borders. Further developments of the algorithm could indeed aim at a more complete formulation of the construction of the object modulus.

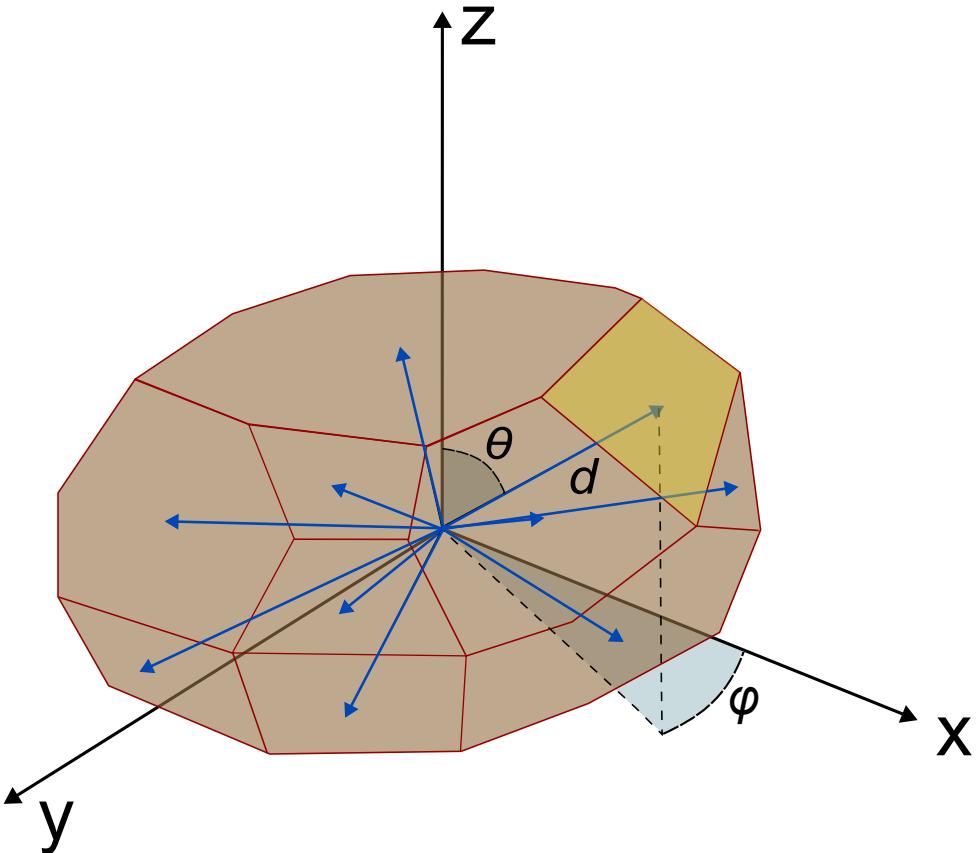


Figure 6.1: Construction of the convex hull with half-spaces expressed with spherical coordinates

The last important consideration of this parametrization is that the support \mathcal{S} is sharply divided into a binary variable (1 inside and 0 outside) thus leading to differentiability problems. In fact, in such a way the gradients, essential for the support update, are not defined. For this reason \mathcal{S} is first passed through a sigmoid function controlled by a hyperparameter ϵ responsible for the smoothening of the support borders. This measure can also be seen as a control of the resolution of the object. Additionally, a mildly steep sigmoid in the early stage of the optimization can function help retrieving a low resolution estimate of the support, that can be further refined adjusting the ϵ parameter.

6.2.2 Object's phase

The parametrization of the object's phase is more challenging. From a qualitative point of view, the prior knowledge that can be exploited for a tailored implementation, is limited to the awareness that a physically meaningful atomic displacement field cannot have “too many” sharp variations. This observation is translated into code by smooth functions parametrization or total variation (TV) regularization of the object's phase. While the former would enforce smoothness by construction the latter would operate adding a penalty to the data-fidelity term of the loss function for non-smooth solutions. Both approaches have been explored and are here reported.

Forcing a scalar field defined on an $L \times H \times W$ grid to exhibit smooth behavior is equivalent to seeking a sparse representation of that field—that is, to concentrating its essential information into far fewer degrees of freedom than the original $L \times H \times W$ samples. Concretely, one looks for a change of basis in which the field can be written as a linear combination of a hierarchy of modes or atoms, ordered by “importance.” In a Fourier or wavelet expansion, for instance, the expansion coefficients are naturally sorted from largest (low-frequency or coarse-scale modes) to smallest (high-frequency or fine-scale modes). Retaining only the largest coefficients both compresses the data and removes rapid oscillations, yielding an inherently smoother reconstruction. Equivalently, in the matrix case a Singular Value Decomposition (SVD) identifies an orthonormal basis in which only a few singular values are nonzero; by truncating to the top singular values one obtains a low-rank—and thus smoother—approximation [145]. For higher dimensional data, this same principle underlies higher-order generalizations of the SVD—Tucker/HOSVD, CP, Tensor-Train, and T-SVD—each of which orders multilinear “modes” by their singular-value (or eigenvalue) strength, and truncating to a small subset produces both compression and smoothness [146].

In this case the Tucker decomposition was chosen, among the several possible methods, for its simplicity of implementation with the Tensorflow library and for the suitability for moderately low dimensions [147]. For a 3D tensor the Tucker decomposition is done as follows:

Considering $\varphi \in \mathbb{R}^{L \times H \times W}$ the 3D object’s phase. The Tucker decomposition expresses φ as:

$$\varphi = \mathcal{G} \times_1 U^{(1)} \times_2 U^{(2)} \times_3 U^{(3)},$$

where:

- $\mathcal{G} \in \mathbb{R}^{R_1 \times R_2 \times R_3}$ is the **core tensor**,
- $U^{(1)} \in \mathbb{R}^{I \times R_1}$, $U^{(2)} \in \mathbb{R}^{J \times R_2}$, and $U^{(3)} \in \mathbb{R}^{K \times R_3}$ are the **factor matrices**,
- \times_n denotes the mode- n tensor-matrix product.

In index notation, this becomes:

$$\varphi_{i,j,k} = \sum_{\alpha=1}^{R_1} \sum_{\beta=1}^{R_2} \sum_{\gamma=1}^{R_3} \mathcal{G}_{\alpha,\beta,\gamma} \cdot U_{i,\alpha}^{(1)} \cdot U_{j,\beta}^{(2)} \cdot U_{k,\gamma}^{(3)}.$$

With this formulation the parameters R_1, R_2, R_3 are set by the user and define the “storage space” in which the information required to represent φ has to be condensed. It is proven that for $R_i = L, H, W$ respectively, the tensor φ is exactly represented. However, being the goal a spare representation of the object’s phase these numbers are chosen significantly smaller than any of the sizes of the array. The Tensorflow implementation of the Tucker decomposition is rather straightforward as the function `tf.einsum()` takes care of the tensor contraction.

A different approach that has been considered, leverages the TV regularization to push the algorithm towards a smooth object’s phase. The full $L \times H \times W$ tensor is therefore optimized and a penalty on the sum of the absolute value of the gradients of the phase is added to the loss function. Precisely, the formula that has been used calculates the sum of the *squared*

gradients, since the square root operation, necessary to obtain the correct formula, creates problem around zero because of the infinite gradient. The final equation is therefore:

$$TV = \alpha \sum_{i=1}^L \sum_{j=1}^H \sum_{k=1}^W \mathcal{S}[(\varphi_i - \varphi_{i-1})^2 + (\varphi_j - \varphi_{j-1})^2 + (\varphi_k - \varphi_{k-1})^2] \quad (6.3)$$

where α is a hyperparameter that acts as a scaling factor, (i, j, k) are the indices running over the coordinates of the $L \times H \times W$ grid and \mathcal{S} is the object support. The parameter α in this case was chosen to be assigned as a fraction, imposed by the user, of the value of the data fidelity loss. Further developments could aim at finding adaptive formulations for the magnitude of α .

6.2.3 Loss function

Another important aspect of the model is the loss function. Typically, for inverse problems there is a *data fidelity* term that in this case measures the distance between I_{obs} and I_{calc} according to some metric, and other additional *regularization* terms that guide the optimization process with physical constraints.

Data fidelity: The most common and intuitive metrics are the Mean Squared Error (MSE) and the Mean Absolute Error (MAE) that evaluate the Euclidean distance between the observed and calculated intensities. Practically, because of the large dynamic range of typical BCDI data, the MAE performs better as it doesn't focus on bright pixels only, but manages to correct for lower intensity tails as well. A more faithful metric for BCDI experimental data is the Poisson Negative Log-Likelihood (P-NLLK). This metric assumes indeed the handling of count data, like the type obtained by photon counting detectors, and that the stochasticity of physical process is Poisson distributed. When summed over the full dataset, the discrepancies between calculated and observed intensities are not intended as Euclidean distances but like divergences between two probability distributions. In other words, the P-NLLK estimates the likelihood that I_{calc} belongs to the same Poisson distribution of I_{obs} [148]. Derived from the equation for the probability for Poissonian events, the formula of the averaged P-NLLK, in the form of a Kullback-Liebler divergence is:

$$\langle P - LLK \rangle = \frac{2}{N_{obs}} \left[\sum_{I_{obs} > 0} \left(I_{calc} - I_{obs} + I_{obs} \ln \frac{I_{obs}}{I_{calc}} \right) + \sum_{I_{obs} = 0} I_{calc} \right]. \quad (6.4)$$

Both the MAE and the P-NLLK have been tested on several simulated and experimental datasets and the MAE has always shown better convergence. An explanation for this unexpected result is yet to be found, but the main suspect is that the gradients calculated during the backpropagation can have instabilities because of the logarithm.

Regularizations: Beside the TV on the object's phase to ensure smoothness, another term that was considered concerns the size of the support. For a given data fidelity value, it is known that the object with the smallest support represents the optimal solution [134]. Intuitively this could be explained with the fact that there are many more object phase configuration that would combine constructive and destructive interferences to match the observed intensity in

reciprocal space. The analogous measure is the shrinkwrap algorithm [83] utilized in alternating projections algorithms. For this reason a penalty P on the size of the support can be added to the loss function with the formula:

$$P = \beta \sum_{i,j,k} \mathcal{S} \quad (6.5)$$

where β is a hyperparameter that similarly to α is chosen by the user with respect to the data fidelity loss. Both the hyperparameters can be tuned manually during the optimization, to adjust in case of need, the strength of the regularization terms. The final formula for the loss function can be ultimately expressed as:

$$L = \frac{1}{N_{\text{obs}}} \sum_{i=1}^{N_{\text{obs}}} |I_{\text{calc},i} - I_{\text{obs},i}| + \alpha TV(\varphi) + \beta P(\mathcal{S}) \quad (6.6)$$

For the optimization, a tolerance on the MAE value or a fixed number of steps can be set to stop the algorithm. Empirical observations have shown that a MAE value around 0.2 is sufficiently low for the result to be considered good. However, an additional refinement with a few iterations (~ 300) is recommended for cross-validation.

Before concluding the paragraph, it is worth highlighting that this AD implementation offers the possibility to simultaneously run multiple reconstructions in parallel, efficiently on the GPU. One can create a 4D tensor by stacking several copies of the 3D intensity data, creating therefore a batch. For each element in the batch a different initial support and phase configuration can be chosen, hence increasing the likelihood to converge to the solution.

6.3 Results

In this section two relevant results will be presented. The first examples is a highly strained Palladium particle on measured at the ID01 beamline of the ESRF [149]. The large strain inside the particle distorts the BCDI pattern and makes the reconstruction with conventional iterative algorithms overly challenging.

6.3.1 Hardly-invertible BCDI patterns with high strain

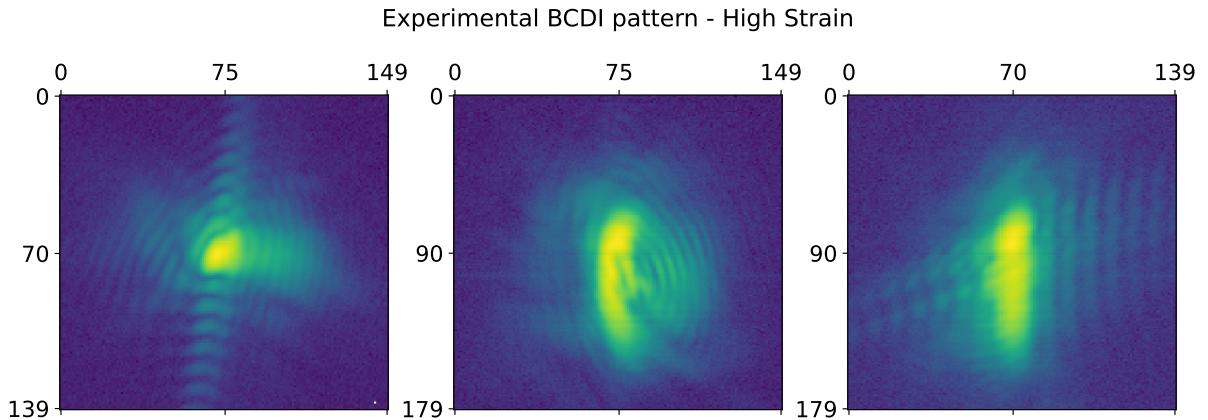


Figure 6.2: Projections along the three axes of a BCDI pattern of the highly strained Nickel particle.

In the following lines a comparison between different reconstruction methods will follow. Namely, (i) the results obtained from 40 independent runs of standard PR using PyNX software. Each run consisted of 400 HIO iterations followed by 1000 RAAR and 300 ER ones. The parameters of are displayed in Table 6.1. At the end of the process, the two best reconstructions among the 40 runs according to the “mean to max” metric [141, 142] are combined using the mode decomposition technique proposed in [134]. This method is referred in the text to as “PyNX”.

Parameter	Value
recipe	400 HIO + 1000 RAAR + 300 ER
nb_runs	40
support_threshold	(0.15, 0.4)
smooth_width	(2, 0.5, 600)
post_expand	None
support_update_period	50
update_border_n	2
smooth_width_begin	2
smooth_width_end	0.5
support_autocorrelation_threshold	(0.09, 0.11)
update_psf	100
psf	'pseudo-voigt, 0.5, 0.1, 10'
support_update_border_n	2
support_post_expand	'1, -2'

Table 6.1: PyNX parameter settings for standard PR.

The second method makes use of the DL model presented in the previous chapter. The large original data is firstly binned to a (196, 140, 140) shape and then cropped in a (80,90,110) shaped ROI. The DL predicted object is then interpolated back to the original size and refined with PyNX using a single run of 300 ER, with the parameters listed in Table 6.2. This combined method is referred in the text to as “DL + PyNX”.

Parameter	Value
recipe	300 ER
nb_runs	1
support_threshold	0.3
smooth_width	(2, 0.5, 600)
post_expand	None
obj	DL_obj
support_update_period	30
update_border_n	1
smooth_width_begin	2
smooth_width_end	0.5
update_psf	100
psf	'pseudo-voigt, 0.5, 0.1, 10'
support_update_border_n	1
support_post_expand	'1, -1'

Table 6.2: PyNX parameter settings for the refinement after the DL prediction

The third method employs the AD model presented above with the parameters listed in Table 6.3. Additionally, as last step, the final object is obtained computing the IFFT of the complex diffracted amplitude built using the experimental diffracted measurement as modulus and the RSP extracted from the FFT of the object itself. This last step that can also be seen as a “*half ER step*”.

The overall here described method is referred in the text to as “AD”. Furthermore, similarly to the DL case, the AD retrieved object can be refined with 300 cycles of ER using PyNX. In this case, this last passage can be used to verify the credibility of the found solution. In fact, such defined AD model can always yield a faceted crystal with homogeneous amplitude that is however far from the actual solution. This method, that runs PyNX with the parameters shown in Table 6.2 using as initial guess the object found with the AD, is in the text referred to as “AD + PyNX”.

Parameter	Value
batch_size	20
nb_half_spaces	128
sigmoid_eps	0.5
kernel_size	(8, 8, 8)
alpha_TV	0.0
beta_small	0.01
initial_lr	0.05
nb_opt_steps	5000

Table 6.3: Parameters initialization for the AD model. In order they represent: (i) the number of copies optimized in parallel, (ii) the number of half-spaces used to build each of the optimized objects' shape, (iii) the ϵ parameter controlling the “spatial resolution” of the support, (iv) the size of the 3D core tensor used to represent the object phase in the Tucker decomposition, (v) the coefficient multiplying the TV loss on the phase (vi) the coefficient multiplying the penalty on the support size, (vii) the initial learning rate for the ADAM optimizer and (viii) the number of iterations.

The following Figures 6.3 - 6.4 - 6.5 - 6.6 show the results of the reconstructions obtained with the “PyNX”, “DL + PyNX”, “AD” and “AD + PyNX” methods respectively.

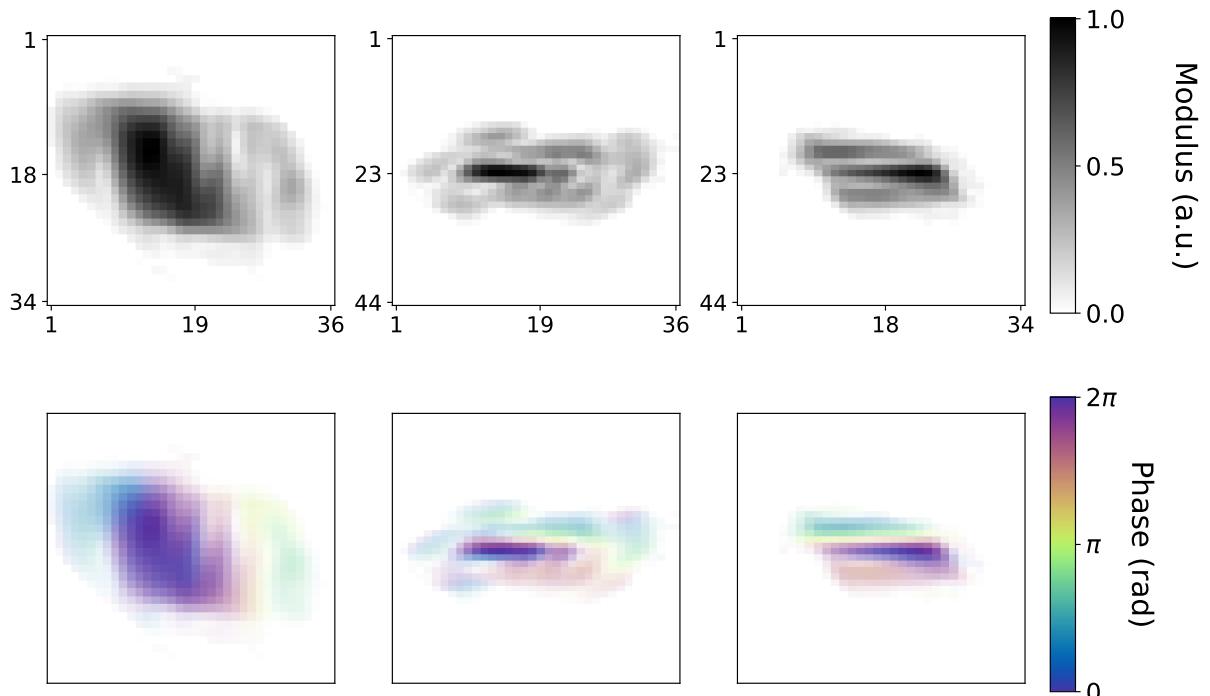


Figure 6.3: Central slices for modulus (first row) and phase (second row) of the reconstruction obtained with the PyNX method. The presence of holes and inhomogeneous object's electron density suggests a poor quality reconstruction.

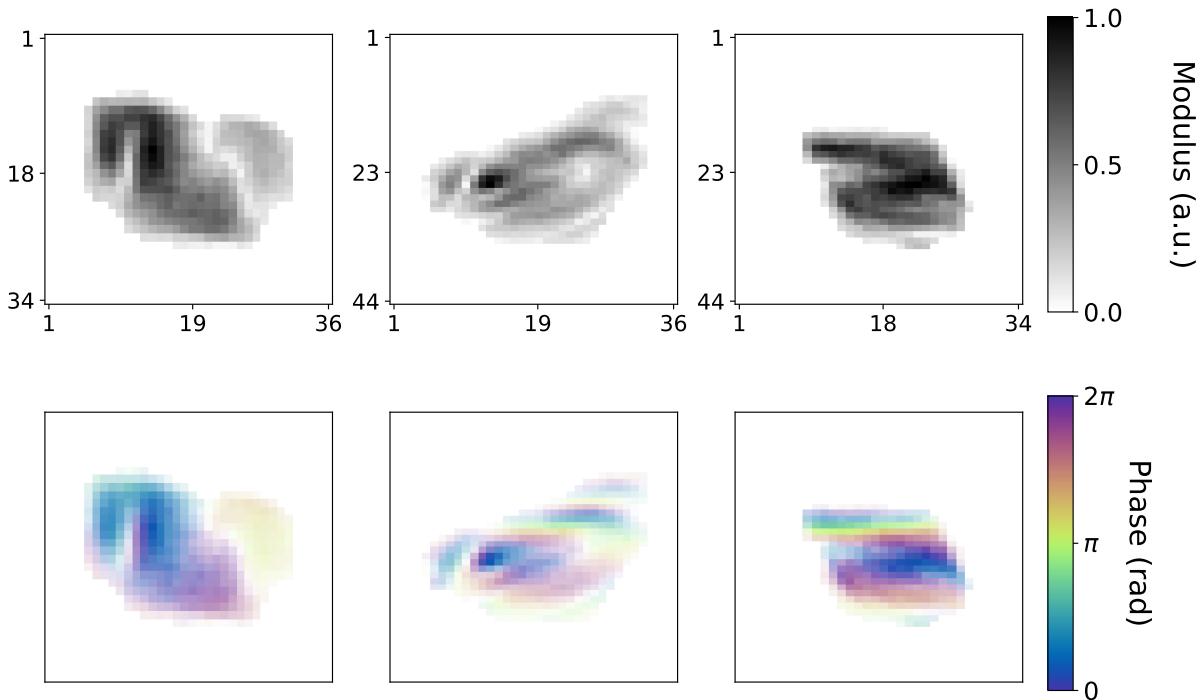


Figure 6.4: Central slices for modulus (first row) and phase (second row) of the reconstruction obtained with the DL + PyNX method. Although the increased quality, the result cannot be considered a good reconstruction.

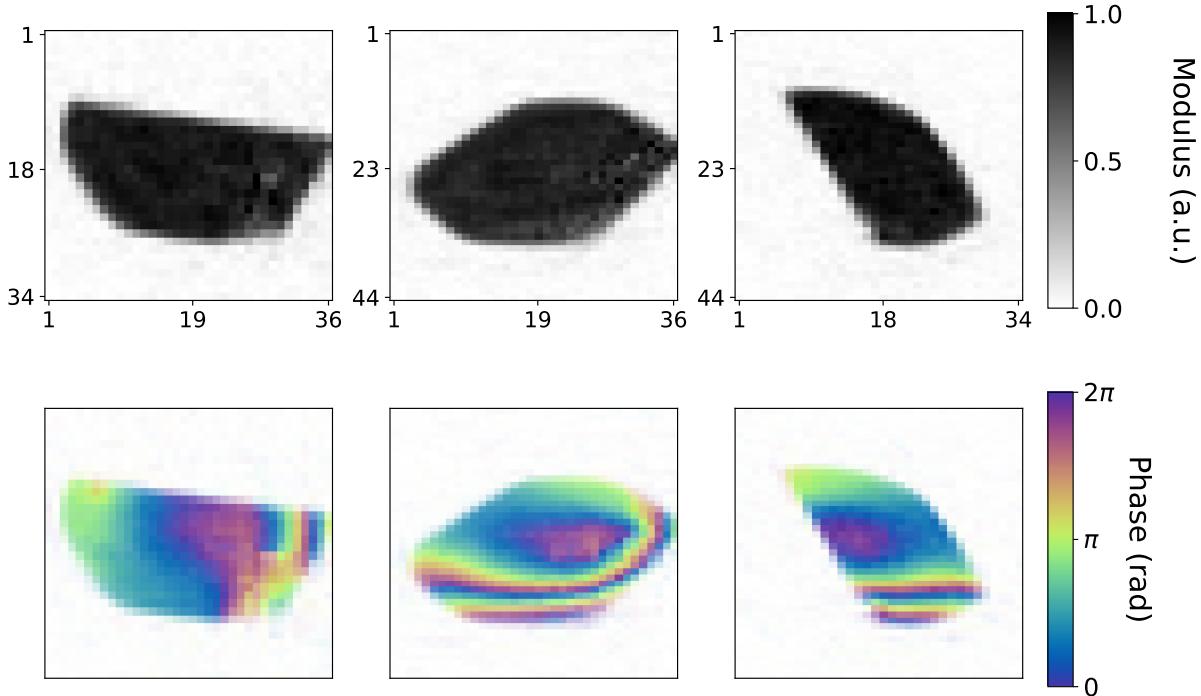


Figure 6.5: Central slices for modulus (first row) and phase (second row) of the reconstruction obtained with the AD method. The model converges to a reasonable result for Winterbottom shaped particle with visible high-strain given by the large phase ramp wrapped multiple times.

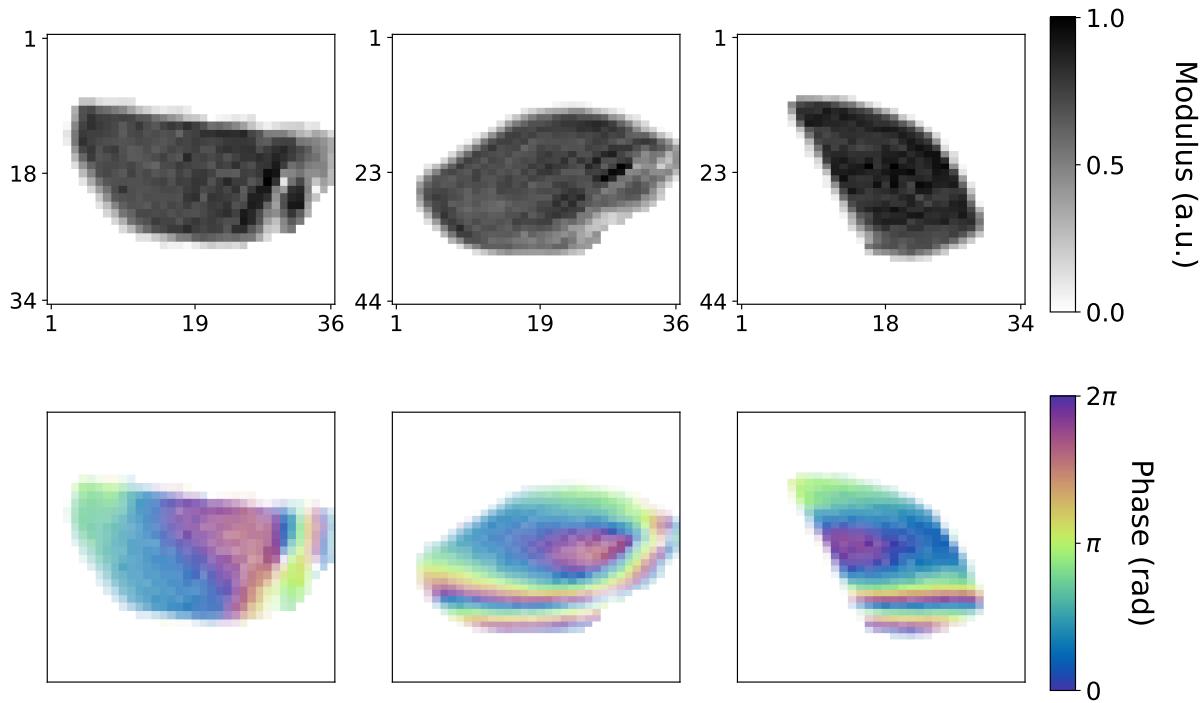


Figure 6.6: Central slices for modulus (first row) and phase (second row) of the reconstruction obtained with the AD + PyNX method. The 300 iterations of ER refinement do not alter the shape nor the phase found by the AD model, therefore validating the solution.

6.3.2 Hardly-invertible BCDI patterns with multiple dislocations

In this paragraph another illustrative example is presented. This time a ... particle. The 4 different methods presented above are repeated here for this dataset in the same way and the results are shown in the following Figures 6.8 - 6.9 - 6.10 - 6.11.

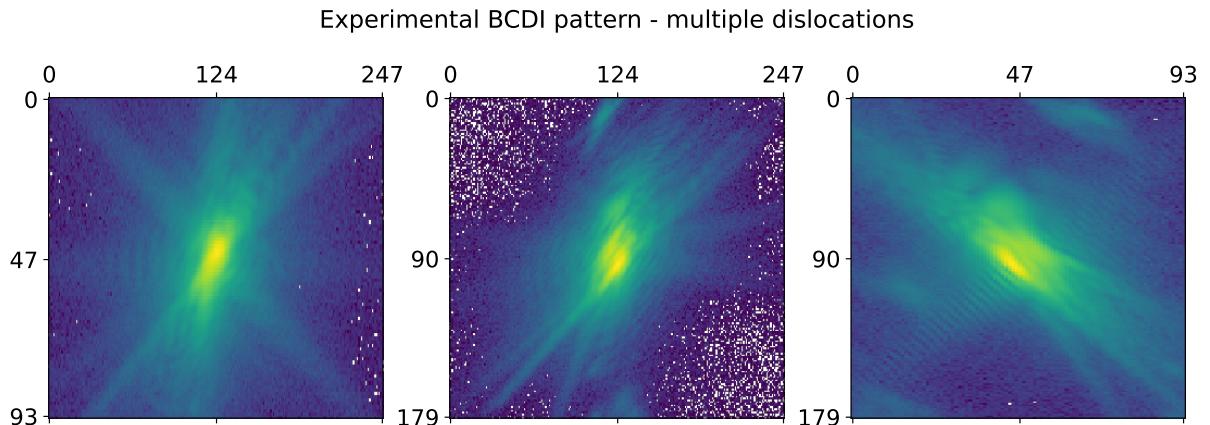


Figure 6.7: Projections along the three axes of a dataset with multiple dislocations.

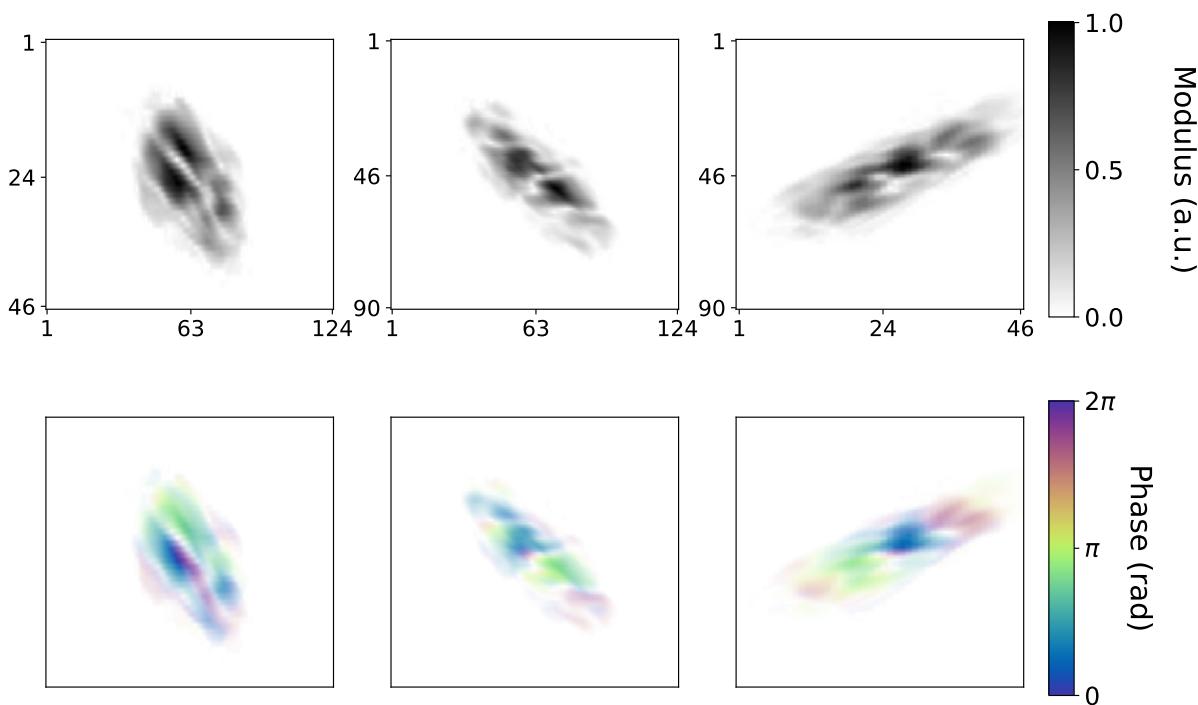


Figure 6.8: Central slices for modulus (first row) and phase (second row) of the reconstruction obtained with the PyNX method. The presence of holes and inhomogeneous object's electron density suggests a poor quality reconstruction.

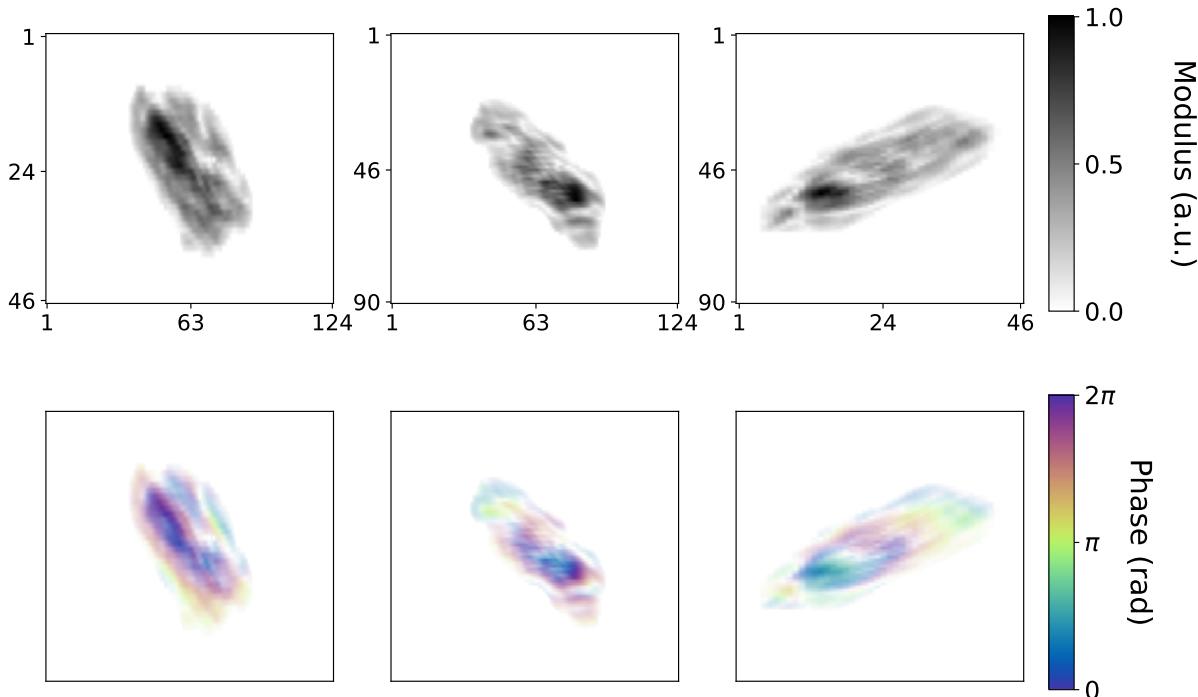


Figure 6.9: Central slices for modulus (first row) and phase (second row) of the reconstruction obtained with the DL + PyNX method. Here, a failure of the DL prediction is expected also because it hadn't been trained on datasets with dislocations.

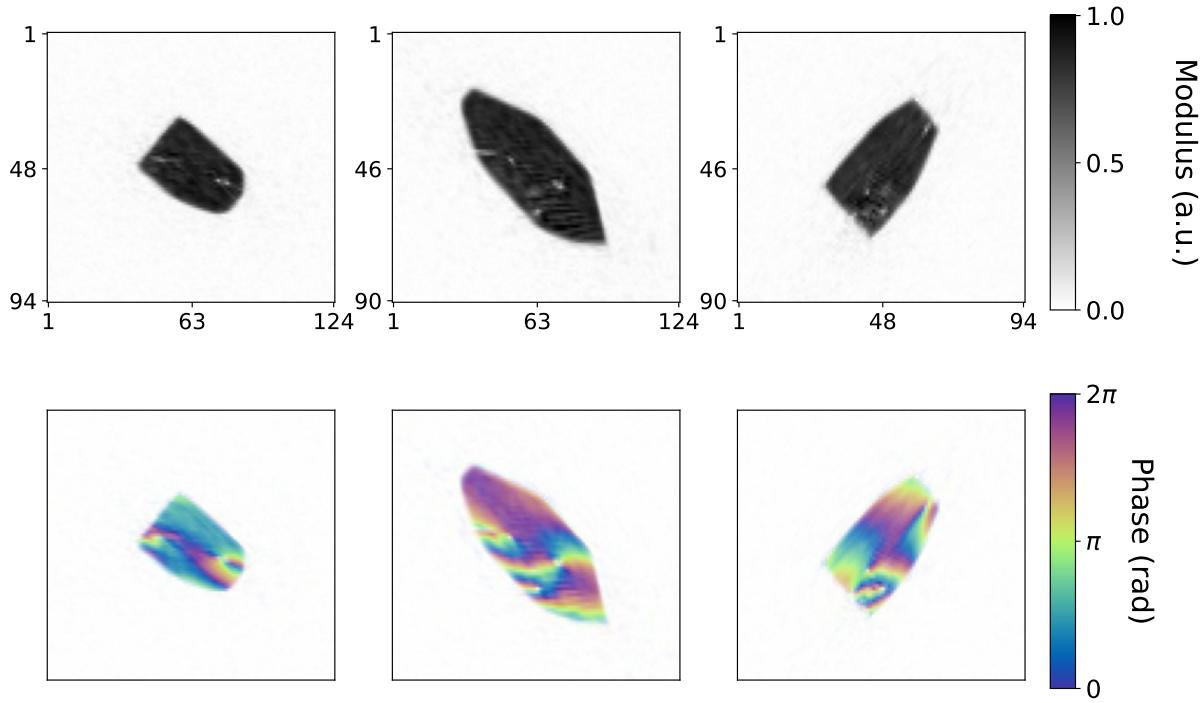


Figure 6.10: Central slices for modulus (first row) and phase (second row) of the reconstruction obtained with the AD method. The AD model converges to a reasonably faceted crystal with several dislocations.

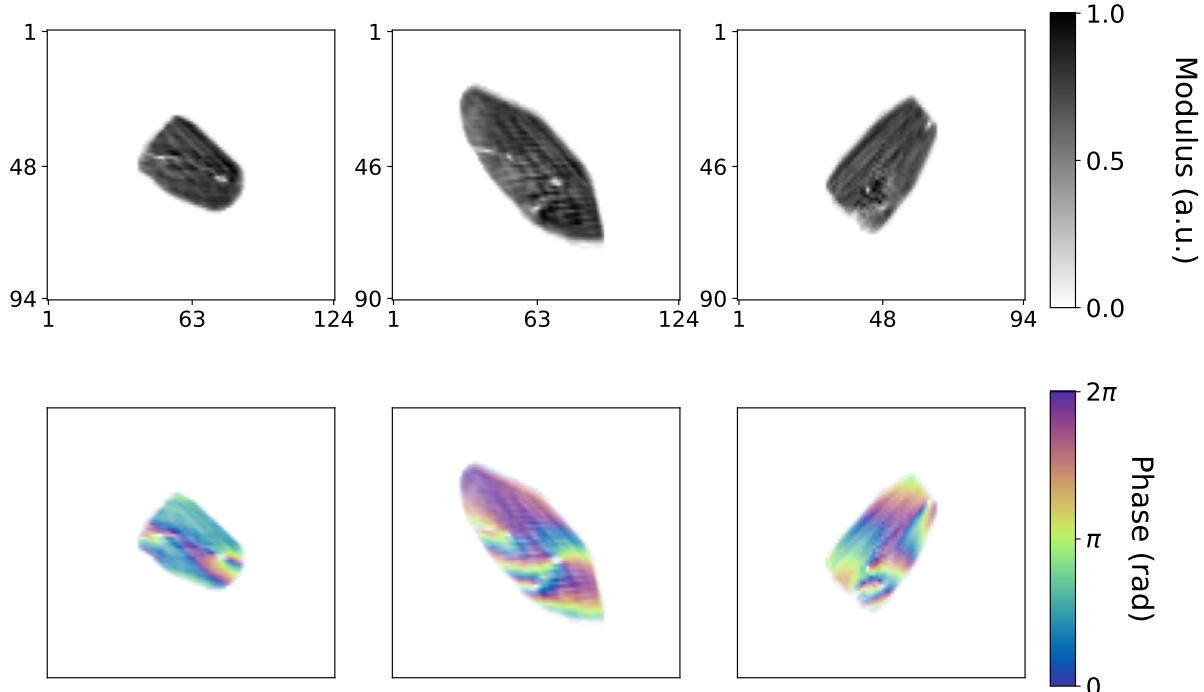


Figure 6.11: Central slices for modulus (first row) and phase (second row) of the reconstruction obtained with the AD + PyNX method. The 300 iterations of ER refinement do not alter the shape nor the phase found by the AD model, therefore validating the solution. Some ripples appear instead in the objects' modulus and phase, probably caused by the presence of some neighbor crystals illuminated by the x-ray beam, scattering on the same detector ROI (*aliens*).

6.4 Conclusions

In this chapter a novel PR method for BCDI based on a physics-informed AD model has been presented. The goal of this project was to explore GD-based methods for PR, aiming at resolving those difficult cases for which both conventional iterative algorithms and DL assisted methods struggle. To sum up, the major factors that made the model successful are here listed briefly:

- The efficient exact gradient calculation offered by the automatic differentiation, already identified by Jurling and Fienup as potential alternative to alternating projections for PR, is today easily accessible and GPU accelerated by common machine learning programming libraries like Tensorflow and PyTorch. This ingredient is fundamental for PR of 3D datasets in competitive computational times, comparable to standard PR algorithms optimized for GPUs.
- Being Fourier PR known to be sensitive to local minima, it is crucial to employ stochastic gradient descent strategies. In particular, the ADAM optimizer combines the stochasticity of mini-batch optimization with adaptive step-size tuning (eliminating the need to set a single learning rate for all parameters) and first and second order moment estimation (thus providing both direction smoothing and per-parameter scaling), leading to more stable and faster convergence. Once again this optimizer is already wrapped into a handy Tensorflow method.
- The possibility to easily embed physical constraints in the forward model. In this case the prior knowledge on the homogeneous and compactly supported nature of the crystal electronic density can be easily implemented with the half-spaces method, thus restricting the solution space without need for additional regularization. In the same way, the Tucker decomposition of the object's phase tensor facilitates the finding of physical solutions.
- The multidimensional tensor-based computations typical of modern machine learning libraries make it straightforward to extend the optimization to multiple parallel instances of the same phase retrieval problem, with only a modest increase in computational time. The main limitation, however, lies in the substantial GPU memory requirements, as this approach can be highly memory-intensive. To give some useful numbers, with the parameters shown in Table 6.3 a GPU with 32GB of RAM can sustain the optimization of maximum 30 copies of a $64 \times 64 \times 64$ diffraction pattern.
- The loss function definition gives the user high flexibility of implementing the most suitable metric for the specific problem, allowing for parameter tuning during the optimization as well. In this case the MAE metric was found to be the best one for the BCDI problem.

This study has therefore shown that AD-based PR for BCDI is a valuable alternative to conventional or data-driven PR algorithms. It offers an additional tool that can extend the range of applicability of the BCDI technique to highly defective or strained crystals. Furthermore, it can be improved to include non-convex objects or separated ones (e.g. in case of twin boundaries) as well as different forward models beyond the kinematic approximation.

CONCLUSIONS

APPENDIX A

ADDITIONAL DATA AND METHODS

APPENDIX B

APPENDIX

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Annexes

APPENDIX A

APPENDIX