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**Imagerie diffractive cohérente de Bragg pilotée par l'apprentissage automatique : de la collecte à la réduction des données**

**Bragg coherent diffractive imaging driven by machine learning: from data collection through reduction**

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## 51 MOTIVATION AND OUTLOOK

### 52 1.1 Preface

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

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

### 73 1.2 Context, PhD objectives and manuscript outline

74 Over the past two decades the field of Coherent X-ray Diffraction Imaging (CXDI or CDI) has  
75 combined the study of microscopic structures enabled by X-rays with the imaging world. This  
76 powerful connection has been enabled by the availability of high-brilliance and coherent X-ray  
77 sources and the developments of computer algorithms for Phase Retrieval (PR) [1–3]. In fact,  
78 unlike conventional imaging methods, instead of relying on high-quality optics to form an

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79 image, CDI records only the intensity of the diffracted waves on a detector and then reconstructs  
80 the object by iterative algorithms. This approach removes the limitations imposed by imperfect  
81 lenses, enabling spatial resolutions set primarily by the wavelength and the numerical aperture  
82 of the scattering geometry. Since the first work by Miao [4] the CDI technique has been  
83 widely employed in materials science for characterization at the nanometric scale of different  
84 functional materials. [5–8]

85 When performed in Bragg geometry (Bragg CDI or BCDI), the technique enables precise 3D  
86 visualization and investigation of the internal atomic structure of single-crystal nanoparticles.  
87 Since its first demonstration in 2001 by Robinson et al. [9] for imaging gold nanoparticles, BCDI  
88 has proven to be a powerful tool for studying strain distributions [10–12], defect populations  
89 [13–15], and particle morphologies under different physico-chemical conditions [16–19]. Over  
90 the years, it has been applied to a broad range of technologically relevant systems, including  
91 nanotechnology and electronics [13], Li-ion and Na-ion batteries [20, 21], catalysis [22], and,  
92 more recently, biological materials [23].

93 In typical experiments, the recorded diffraction patterns are processed by PR computer  
94 algorithms returning 3D complex arrays, the modulus of which represents the electron density  
95 of the sample, while the phase is associated with a component of the strain distribution inside  
96 the sample. Unlike other X-ray diffraction techniques, BCDI enables the study of isolated  
97 single particles, down to a few tens of nanometers in size [24, 25], with spatial resolution on  
98 the electron density of the order of 10 nm [26] and sensitivity to strain in the order of a few  
99 picometers [14].

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

105 **Chapter 2** introduces the fundamental theoretical background of this technique and de-  
106 scribes the practical conditions for a BCDI experiment at ID01.

107 The successful application of BCDI critically depends on computational algorithms that  
108 transform measured diffraction patterns into real-space reconstructions of particle shape and  
109 strain fields. From the early developments, significant efforts have been devoted to improv-  
110 ing the speed, robustness and reliability of these algorithms. These algorithms are usually  
111 available in Python or MATLAB based softwares, including Bonsu [30], the widely used PyNX  
112 [31] with related toolkits [32, 33] and the more recent SPRING [34]. This field has gained  
113 further momentum with the advent of machine learning (ML). In parallel, the recent upgrade  
114 of numerous synchrotron facilities to fourth-generation light sources – including MAX IV  
115 (2017), ESRF-EBS (2020), Sirius (2020), and APS (2025) – has dramatically increased coherent  
116 flux, boosting the potential of crystalline nano-imaging techniques such as BCDI and Bragg  
117 ptychography [27, 35–37]. These advances also pose new challenges, particularly the need for  
118 faster and more efficient data processing pipelines capable of handling the rapidly growing  
119 volume of experimental data.

This PhD project, as part of the ENGAGE doctoral program<sup>1</sup>, and in collaboration with ID01 beamline and the Algorithms and Data Analysis group of the ESRF, was conceived in this context, with the goal of exploring how ML algorithms can address specific computational challenges in BCDI. **Chapter 3** addresses the main aspects of the Phase Problem in BCDI and provides the necessary theoretical background on conventional phase retrieval algorithms and **Chapter 4** 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 are often made of arrays of chips, butted together in a way that leads to data gaps within the diffraction patterns. **Chapter 5** 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 wave-field, making its retrieval an inherently challenging computational task. **Chapters 6 and 7** present how ML-based approaches developed during this PhD can assist or complement conventional phase retrieval methods.

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## 136 BRAGG COHERENT DIFFRACTION IMAGING

137 In this chapter some basic theoretical insights about the BCDI technique are provided, with the  
138 aim to highlight the key concepts, assumptions and physical interpretations. More thorough  
139 descriptions can be found in papers, textbooks and PhD manuscripts. I will adopt the formalism  
140 of Als-Nielsen and McMorrow in [38] but similar derivations and complementing observations  
141 can be found in [39, 40] as well as some more recent papers [41], PhD theses [42–44] and  
142 lecture notes [45].

### 143 2.1 Foreword on typical assumptions and approximations 144 in BCDI

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

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

154 The word “Diffraction” refers to the type of mechanism describing the interaction between  
155 the X-rays and the samples. Paraphrasing [39] at page 4, this mechanism can be divided into  
156 two main phenomena, namely (i) the scattering of the radiation by each individual atom in the  
157 sample and (ii) the interference between the waves scattered by these atoms. The interference  
158 mechanism, in turn, is enabled because these scattered waves are coherent with the incident  
159 radiation and therefore between themselves. In other words, the information of each scatterer  
160 is shared with the other scatterers as the diffracted waves “talk to each other”. The complete  
161 mathematical description of these two phenomena without approximations is prohibitive,  
162 hence some simplifications are usually adopted:

- 163 • **No refraction, no absorption:** Scattering is the only mechanism considered. Because  
164 of their short wavelengths (0.5 - 2.5 Å), X-rays refraction is very small, hence for small

samples the deviation of the outgoing rays with respect to the incident ones is negligible. Moreover, we assume the absence of any absorption effect. While inherently present, it can be neglected for 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 instead bounces off 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 that 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 ( $> 1\mu m$ ) 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* [46–48]. However, in our case, the size of the typical samples studied with BCDI rarely exceeds  $1\mu m$  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 sample's 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 the 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.

## 2.2 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. 2.1). In this simple case we can imagine the X-ray electromagnetic field exerting a force (Lorentz 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. If the scattering is 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's 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 [38, 39, 49].

$$\mathbf{E}_{\text{dip}}(\mathbf{r}, t) = -r_e \frac{e^{ikr}}{r} e^{-i\omega t} E_0 \hat{\mathbf{z}} \quad (2.1)$$

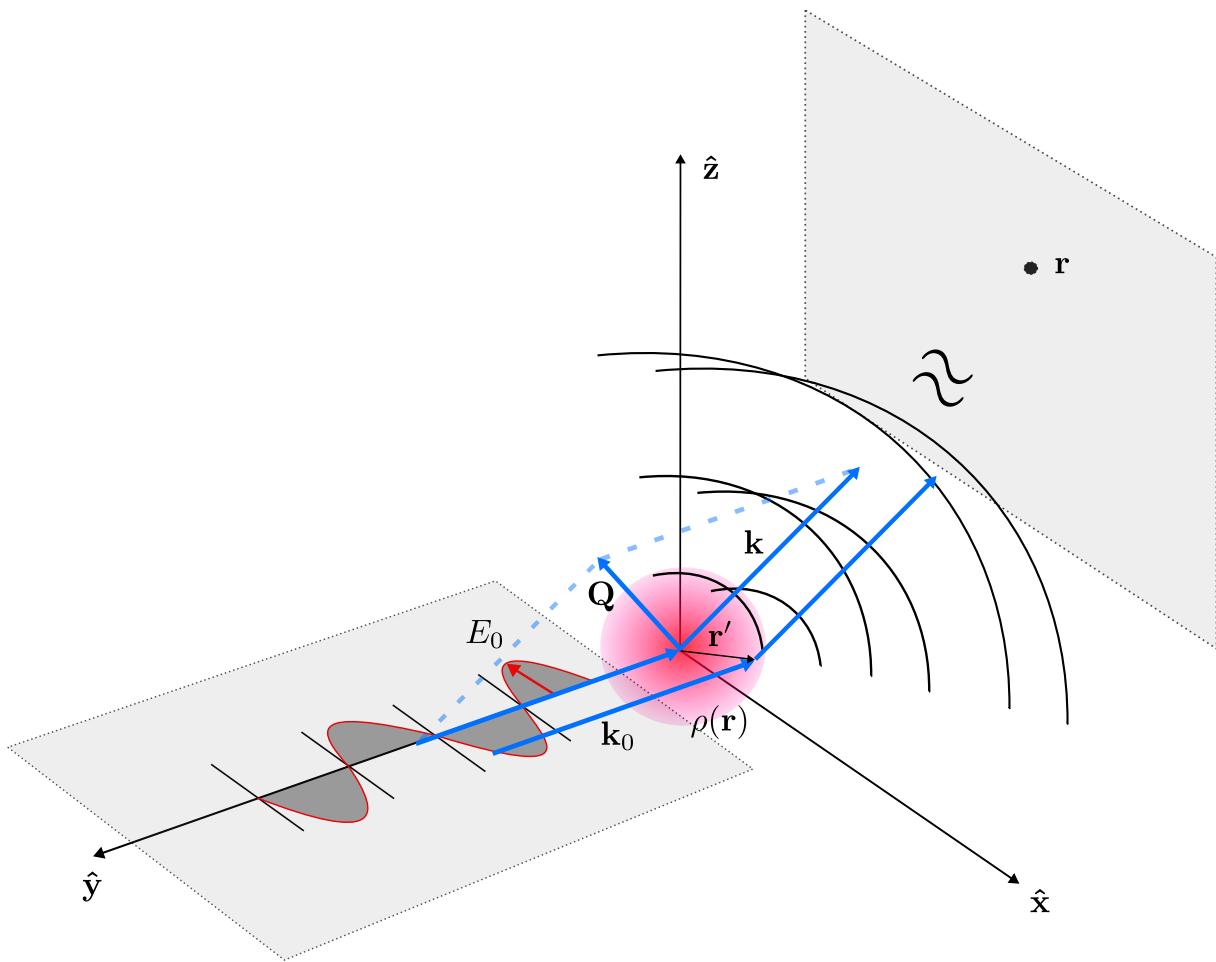
where  $r_e$  is the classical radius of the electron, or Thomson scattering length,  $k$  is the outgoing wave-vector,  $\omega$  is the pulsation of the X-ray beam (incoming and outgoing),  $E_0$  is the electric field of the incoming radiation.

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 incoming and outgoing wave vectors,  $\mathbf{k}_0$  and  $\mathbf{k}$  respectively, with  $\mathbf{Q}$  often called *scattering vector* (see Fig.2.1). The equation can thus be rewritten like:

$$\mathbf{E}_{\text{atom}}(\mathbf{r}, t) = r_e \frac{e^{ikr}}{r} e^{-i\omega t} E_0 \hat{\mathbf{z}} \sum_{n=1}^N e^{i\mathbf{Q} \cdot \mathbf{r}'_n} \quad (2.2)$$

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:

$$\mathbf{E}_{\text{atom}}(\mathbf{r}, t) = r_e \frac{e^{ikr}}{r} e^{-i\omega t} E_0 \hat{\mathbf{z}} \int_{\mathbb{R}^3} \rho_a(\mathbf{r}') e^{i\mathbf{Q} \cdot \mathbf{r}'} d^3 \mathbf{r}' \quad (2.3)$$



**Figure 2.1:** 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.

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

$$F(\mathbf{Q}) = \int_{\mathbb{R}^3} \rho(\mathbf{r}') e^{i\mathbf{Q} \cdot \mathbf{r}'} d^3 \mathbf{r}' \quad (2.4)$$

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

237 To continue, we should bear in mind that X-ray photon counting detectors are sensitive  
 238 to the time-averaged intensity of the signal as their time response is much slower than the  
 239 oscillating frequency of X-rays ( $\sim 10^9$  Hz for typical read-out limited frame rates of the Maxipix  
 240 [50] against the  $\sim 10^{18}$  Hz for X-rays at 10 keV). This limitation is also at the core of the “Phase  
 241 Problem” that we will see later on, for which the phase information of the complex-valued  
 242 wave-field is lost in the measurement. In order to do so, the time-averaged Poynting vector is  
 243 calculated.

$$\langle \mathbf{S}(\mathbf{r}) \rangle = r_e^2 \frac{1}{r^2} J_0 \left| \int \rho(\mathbf{r}') e^{i\mathbf{Q} \cdot \mathbf{r}'} d^3 r' \right|^2 \hat{\mathbf{r}} \quad (2.5)$$

244 where  $J_0 = |\mathbf{E}_0|^2 / 2\mu_0 c$  is the incident intensity. To conclude we consider the power  
 245 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:

$$P(\mathbf{Q}) = r_e^2 J_0 \left| \int \rho(\mathbf{r}') e^{i\mathbf{Q} \cdot \mathbf{r}'} d^3 r' \right|^2 d\Omega \quad (2.6)$$

246 Eq.2.6 shows that the signal captured by the detectors is now in  $\mathbf{Q}$  space, and it is propor-  
 247 tional to the square modulus of the Fourier transform of the electron density of the sample.  
 248 The square modulus operation also shows how the phase of the Fourier transformed scattering  
 249 complex-amplitude is lost. For this reason, in order to reconstruct the electron density, it is not  
 250 possible to simply use an inverse Fourier transform, but iterative methods are required.

### 251 2.2.1 One atom

252 If now we were to consider an atom, far from resonance, we could assume the electron density  
 253 being the main responsible for the scattering. It is known indeed that protons, because of the  
 254 larger mass, have a much smaller cross-section for the scattering with photons. Using Eq.2.4  
 255 we would therefore have the *atomic form factor* -  $f_l(\mathbf{Q})$  being defined as:

$$f_l(\mathbf{Q}) = \int_{\mathbb{R}^3} \rho_l(\mathbf{r}) e^{i\mathbf{Q} \cdot \mathbf{r}} d^3 \mathbf{r} \quad (2.7)$$

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

### 258 2.2.2 Ensemble of ordered atoms: a Crystal

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

$$\mathbf{R}_n = n_1 \mathbf{a}_1 + n_2 \mathbf{a}_2 + n_3 \mathbf{a}_3 \quad (2.8)$$

262 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  
 263 integer numbers. It follows that the information can be condensed in the unit cell, i.e. the  
 264 orientation in space of the basis vectors as any region of the lattice can be seen as the same  
 265 unit cell, translated from the origin by the amount given by  $\mathbf{R}_n$ .

266

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

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

$$\mathcal{C}(\mathbf{r}) = \mathcal{L}(\mathbf{r}) * \mathcal{B}(\mathbf{r}) \quad (2.9)$$

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

$$F_{\text{crystal}}(\mathbf{Q}) = \sum_{l=1}^{\text{All atoms}} f_l(\mathbf{Q}) e^{i\mathbf{Q} \cdot \mathbf{r}_l} \quad (2.10)$$

277 Secondly, observing that the position of each atom is given by the sum of the position of  
278 the atom inside the unit cell and the lattice vector  $\mathbf{r}_l = \mathbf{R}_n + \mathbf{r}_j$ , we can separate Eq.2.10 in  
279 two terms:

$$F_{\text{crystal}}(\mathbf{Q}) = \sum_{\mathbf{R}_n + \mathbf{r}_j}^{\text{All atoms}} f_l(\mathbf{Q}) e^{i\mathbf{Q} \cdot (\mathbf{R}_n + \mathbf{r}_j)} = \underbrace{\sum_n e^{i\mathbf{Q} \cdot \mathbf{R}_n}}_{\text{Lattice}} \underbrace{\sum_j f_j(\mathbf{Q}) e^{i\mathbf{Q} \cdot \mathbf{r}_j}}_{\text{Unit cell}} \quad (2.11)$$

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

$$F_{\text{crystal}}(\mathbf{Q}) = F_{\text{u.c.}}(\mathbf{Q}) \sum_n e^{i\mathbf{Q} \cdot \mathbf{R}_n} \quad (2.12)$$

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

### 286 2.2.3 Laue condition and Bragg's Law

287 The term we want to calculate is the sum of complex exponential, meaning that if the phases  
288  $\mathbf{Q} \cdot \mathbf{R}_n$  are misaligned the sum will mutually cancel out the contributions and the resultant  
289 will be very small, in the order of unity. On the contrary, when phase offsets are equal to an  
290 integer multiple of  $2\pi$  the *phasors*, complex vectors  $e^{i\mathbf{Q} \cdot \mathbf{R}_n}$  for each  $n$ , will add *in phase*. The  
291 problem is thus to find those  $\mathbf{Q}$  values for which

$$\mathbf{Q} \cdot \mathbf{R}_n = 2\pi \times \text{integer} \quad \forall n \quad (2.13)$$

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

$$\mathbf{a}_1 \cdot \mathbf{a}_1^* = 2\pi h \quad \mathbf{a}_2 \cdot \mathbf{a}_2^* = 2\pi k \quad \mathbf{a}_3 \cdot \mathbf{a}_3^* = 2\pi l \quad (2.14)$$

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

$$\mathbf{G}_{hkl} = h\mathbf{a}_1^* + k\mathbf{a}_2^* + l\mathbf{a}_3^* \quad (2.15)$$

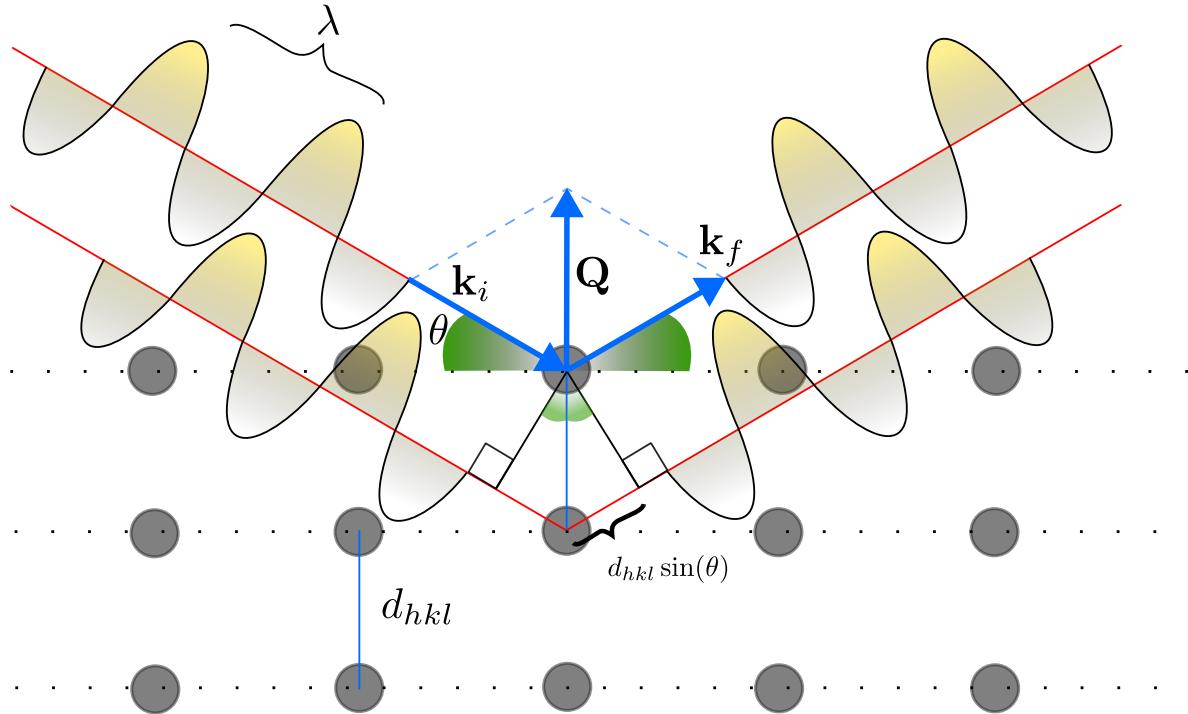
297 This result is telling us that the scattering amplitude of a diffracting crystal is detectable  
 298 only in those points in space for which the wave-vector  $\mathbf{Q}$  coincides with a point ( $hkl$  node) of  
 299 the reciprocal lattice, hence  $\mathbf{Q} = \mathbf{G}_{hkl}$ . These isolated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

300 This is known as the Laue condition for diffraction as it was discovered by Max von Laue  
 301 in 1912 [51].

302 A different but equivalent interpretation of the diffraction of a crystal was given by William  
 303 Lawrence Bragg in 1913 [52]. Here, the crystal lattice is seen as a stack of parallel planes  
 304 (see Fig.2.2) and the condition for constructive interference of the waves scattered by planes  
 305 of the same family is found as follows. Let us consider an X-ray beam of wavelength  $\lambda$  and  
 306 propagation vector  $\mathbf{k}_i$  impinging with an angle  $\theta$  on a crystal. We call  $d_{hkl}$  the distance between  
 307 the planes of the crystal, along a specific  $hkl$  direction. The scattered beam is leaving the crystal  
 308 with the same angle  $\theta$  and with a propagation vector  $\mathbf{k}_f$  equal in magnitude to the incident one  
 309 (*elastic scattering*). At this point one can find the relationship between  $\theta, d_{hkl}, \lambda$  that allows for  
 310 a constructive interference of the waves diffracted from the series of planes by evaluating the  
 311 optical path length difference induced by the spacing. Reminding that  $|k_i| = |k_f| = 2\pi/\lambda$  and  
 312 with the help of Fig. 2.2 we can observe that this difference is  $\Delta l = 2d_{hkl} \sin(\theta)$  and therefore  
 313 the phase offset between two waves is  $\Delta\phi = |k| \Delta l = 4d_{hkl}\pi \sin(\theta)/\lambda$ . We have seen above  
 314 that the condition for constructive interference requires the phase differences to be equal to a  
 315 multiple of  $2\pi$ , therefore:

$$2d_{hkl} \sin(\theta) = n\lambda \quad \text{where } n \in \mathbb{Z} \quad (2.16)$$

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



**Figure 2.2:** 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  $\theta$  given by Eq.2.16.

320 Equation 2.16 is known as Bragg's law, and it can be derived from the Laue condition.

321 We can now rewrite Eq.2.12 for the case of an infinitely extended perfect 3D crystal as:

$$F_{\text{crystal}^\infty}(\mathbf{Q}) = F_{\text{u.c.}}(\mathbf{Q}) \sum_h \sum_k \sum_l \delta(\mathbf{Q} - \mathbf{G}_{hkl}) \quad (2.17)$$

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

## 324 2.2.4 Finite size crystals

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

331 Mathematically we can derive the result for crystal of shape  $S(\mathbf{r})$  considering the function  
332  $S$  a window cropping a finite portion of the 3D infinite crystal. This function can also be  
333 referred to as an Ewald function [53] or *support*. In real space this corresponds to the product:

$$\rho_{\text{fin}}(\mathbf{r}) = \rho_\infty(\mathbf{r})S(\mathbf{r}) \quad (2.18)$$

334 where

$$S(\mathbf{r}) = \begin{cases} 1, & \text{inside the crystal region,} \\ 0, & \text{otherwise.} \end{cases} \quad (2.19)$$

335 From the convolution theorem we have now that the Fourier transform  $\mathcal{F}\{\rho_{\text{fin}}(\mathbf{r})\}$  is equal  
 336 to the convolution  $\mathcal{F}\{\rho_{\infty}(\mathbf{r})\} * \mathcal{F}\{S(\mathbf{r})\}$ . Using Eq.2.12 for the Fourier transform of the infinite  
 337 crystal and calling  $\widehat{S}(\mathbf{Q}) = \mathcal{F}\{S(\mathbf{r})\}$  we can therefore write:

$$F_{\text{fin. crystal}}(\mathbf{Q}) = \left[ F_{\text{u.c.}}(\mathbf{Q}) \sum_h \sum_k \sum_l \delta(\mathbf{Q} - \mathbf{G}_{hkl}) \right] * \widehat{S}(\mathbf{Q}) \quad (2.20)$$

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

$$\begin{aligned} F_{\text{fin. crystal}}(\mathbf{Q}) &= \widehat{S}(\mathbf{Q}) * \left[ F_{\text{u.c.}}(\mathbf{Q}) \sum_h \sum_k \sum_l \delta(\mathbf{Q} - \mathbf{G}_{hkl}) \right] \\ &= \int \widehat{S}(\mathbf{Q} - \mathbf{Q}') F_{\text{u.c.}}(\mathbf{Q}') \sum_h \sum_k \sum_l \delta(\mathbf{Q}' - \mathbf{G}_{hkl}) d^3 \mathbf{Q}' \end{aligned} \quad (2.21)$$

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

$$F_{\text{fin. crystal}}(\mathbf{Q}) = \sum_h \sum_k \sum_l \int \widehat{S}(\mathbf{Q} - \mathbf{Q}') F_{\text{u.c.}}(\mathbf{Q}') \delta(\mathbf{Q}' - \mathbf{G}_{hkl}) d^3 \mathbf{Q}' \quad (2.22)$$

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

$$F_{\text{finite}}(\mathbf{Q}) = \sum_h \sum_k \sum_l \widehat{S}(\mathbf{Q} - \mathbf{G}_{hkl}) F_{\text{u.c.}}(\mathbf{G}_{hkl}) \quad (2.23)$$

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

345

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

$$S(\mathbf{r}) = \prod_{j=1}^3 \mathcal{R}\left(\frac{\mathbf{r} \cdot \hat{\mathbf{a}}_j}{L_j}\right), \quad \mathcal{R}(x) = \begin{cases} 1, & |x| \leq 1/2, \\ 0, & \text{else.} \end{cases} \quad (2.24)$$

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

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

354 It follows that:

$$\widehat{S}(\mathbf{Q}) = \prod_{j=1}^3 L_j \operatorname{sinc}\left(\frac{\mathbf{Q} \cdot \hat{\mathbf{a}}_j L_j}{2}\right), \quad (2.25)$$

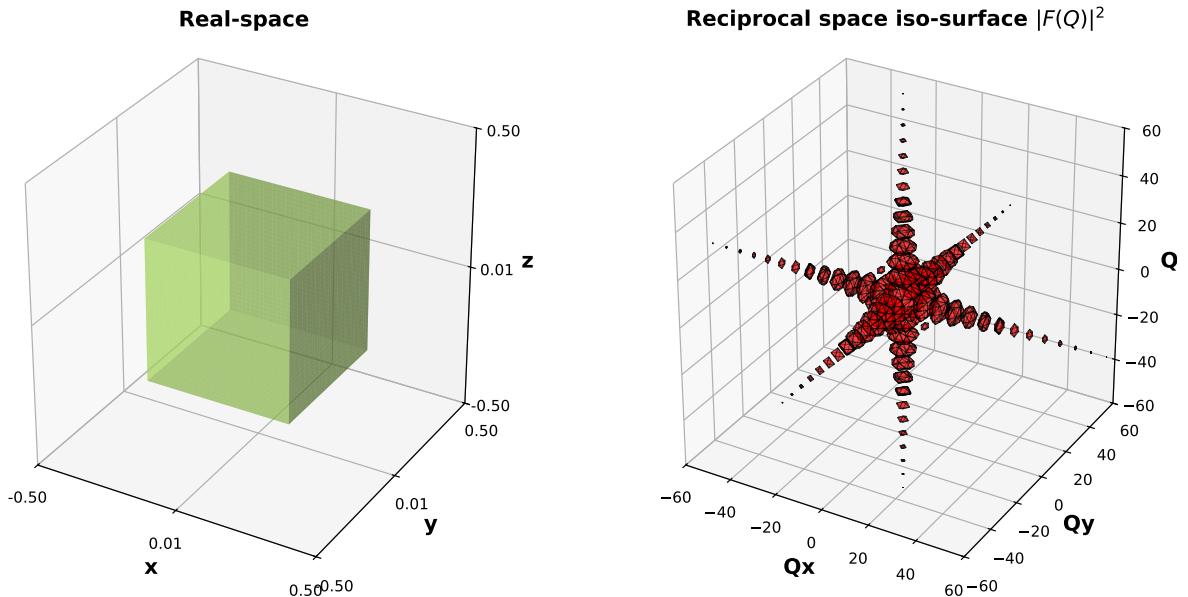
355 where we made use of the cardinal sine function defined as  $\operatorname{sinc}(x) = \frac{\sin x}{x}$ . Putting this  
 356 result in Eq. 2.23 we obtain:

$$F_{\text{cube}}(\mathbf{Q}) = \sum_h \sum_k \sum_l F_{\text{u.c.}}(\mathbf{G}_{hkl}) \prod_{j=1}^3 L_j \operatorname{sinc}\left(\frac{(\mathbf{Q} - \mathbf{G}_{hkl}) \cdot \hat{\mathbf{a}}_j L_j}{2}\right), \quad (2.26)$$

357 At this point of the derivation we can introduce another simplification, directly linked to  
 358 the BCDI technique. Up to now we have been considering the scattering amplitude in the full  
 359 reciprocal space. This means that Eq. 2.26 is encompassing all the  $hkl$  nodes in which we could  
 360 find the diffracted signal. However, in typical BCDI experiments only a small region around a  
 361 single Bragg peak is measured. This means that we can from now on focus on one  $\mathbf{G}_{hkl}$  vector  
 362 and drop the summation. Moreover, defining  $\mathbf{q} = \mathbf{Q} - \mathbf{G}_{hkl}$  and recalling the result obtained  
 363 in Eq. 2.6 one can find that the intensity of the diffraction pattern of the  $hkl$  Bragg peak is :

$$I_{\text{cube}}(\mathbf{q}) \propto F_{\text{u.c.}}(\mathbf{G}_{hkl}) \left| \prod_{j=1}^3 L_j \operatorname{sinc}\left(\frac{\mathbf{q} \cdot \hat{\mathbf{a}}_j L_j}{2}\right) \right|^2, \quad (2.27)$$

364 The diffracted signal for a cubic crystal in the vicinity of the  $hkl$  Bragg peak has therefore  
 365 the shape of a *squared* 3D cardinal sine function as represented in Fig. 2.3

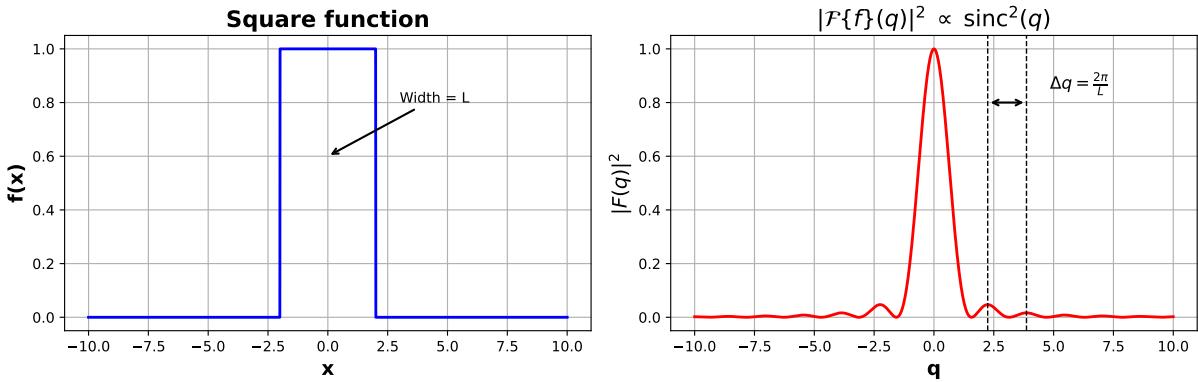


**Figure 2.3:** Illustration of the squared modulus of the Fourier transform of cube as representation of the diffracted intensity around a Bragg peak for a cubic crystal.

366 An interesting observation concerns the spacing between the fringes of the interference  
 367 pattern. Studying Eq. 2.27 we can infer that:

- 368 • The maximum of the peak is concentrated at  $\mathbf{q} = 0 \Rightarrow \mathbf{Q} = \mathbf{G}_{hkl}$  as expected.
- 369 • The intensity decays as  $\sim 1/q^2$  along the intensity streaks (see Fig. 2.4), also known as  
 370 crystal truncation rods (CTR)<sup>1</sup>. This decay depends on the shape. For a spherical crystal  
 371 it follows a  $\sim 1/q^4$  law.
- 372 • The intensity drops to zero every time the argument  $(\mathbf{q} \cdot \hat{\mathbf{a}}_j L)/2 = n\pi$  with  $n$  integer.  
 373 This results in the presence of fringes that have a thickness as a function of the  $\mathbf{q}$  direction  
 374 given by:  $\Delta q_j = 2\pi/L$  (see Fig. 2.4).
- 375 • Given the above relationship between the thickness of the fringes and the size of the  
 376 crystal, one can intuitively imagine extending  $L$  to infinity, thus narrowing the fringes  
 377 to zero width and the central lobe to a Dirac delta in  $\mathbf{q} = 0$  as in the infinite lattice case.

<sup>1</sup>More in general, CTRs appear as lines of more intense diffraction, perpendicular to facets of the diffracting nano-crystal [54].



**Figure 2.4:** 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.

378 For the understanding and visualization of Fourier transforms in crystallography I recom-  
379 mend the pedagogical work of Aubert and Lecomte [55].

### 380 2.2.5 Real crystals

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

$$F_\infty^{\text{strain}}(\mathbf{Q}) = \sum_n F_n(\mathbf{Q}) e^{i\mathbf{Q} \cdot (\mathbf{R}_n + \mathbf{u}(\mathbf{R}_n))} \quad (2.28)$$

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

$$F_{\text{fin}}^{\text{strain}}(\mathbf{Q}) = \sum_n F_n(\mathbf{Q}) e^{i\mathbf{Q} \cdot \mathbf{R}'_n} * \hat{S}(\mathbf{Q}) \quad (2.29)$$

395 Developing the convolution integral we obtain:

$$\begin{aligned}
F_{\text{fin}}^{\text{strain}}(\mathbf{Q}) &= \int \sum_n F_n(\mathbf{Q} - \mathbf{Q}') e^{i(\mathbf{Q}-\mathbf{Q}') \cdot \mathbf{R}'_n} \widehat{S}(\mathbf{Q}') d^3 \mathbf{Q}' \\
&= \sum_n \int F_n(\mathbf{Q} - \mathbf{Q}') e^{i(\mathbf{Q}-\mathbf{Q}') \cdot \mathbf{R}'_n} \widehat{S}(\mathbf{Q}') d^3 \mathbf{Q}' 
\end{aligned} \tag{2.30}$$

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:

$$\begin{aligned}
F_{\text{fin}}^{\text{strain}}(\mathbf{Q}) &= \sum_n F_n(\mathbf{G}_{hkl}) \int e^{i(\mathbf{Q}-\mathbf{Q}') \cdot \mathbf{R}'_n} \widehat{S}(\mathbf{Q}') d^3 \mathbf{Q}' \\
&= \sum_n F_n(\mathbf{G}_{hkl}) \int e^{i\mathbf{Q} \cdot \mathbf{R}'_n} e^{-i\mathbf{Q}' \cdot \mathbf{R}'_n} \widehat{S}(\mathbf{Q}') d^3 \mathbf{Q}' \\
&= \sum_n F_n(\mathbf{G}_{hkl}) e^{i\mathbf{Q} \cdot \mathbf{R}'_n} \int e^{-i\mathbf{Q}' \cdot \mathbf{R}'_n} \widehat{S}(\mathbf{Q}') d^3 \mathbf{Q}' \\
&= \sum_n F_n(\mathbf{G}_{hkl}) e^{i\mathbf{Q} \cdot \mathbf{R}'_n} S(\mathbf{R}'_n)
\end{aligned} \tag{2.31}$$

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:

$$\begin{aligned}
F_{\text{fin}}^{\text{strain}}(\mathbf{Q}) &= \sum_n F_n(\mathbf{G}_{hkl}) e^{i\mathbf{Q} \cdot \mathbf{R}'_n} S(\mathbf{R}'_n) \\
&= F_{\text{u.c.}}(\mathbf{G}_{hkl}) \sum_n e^{i(\mathbf{G}_{hkl} + \mathbf{q}) \cdot (\mathbf{R}_n + \mathbf{u}(\mathbf{R}_n))} S(\mathbf{R}'_n) \\
&= F_{\text{u.c.}}(\mathbf{G}_{hkl}) \sum_n e^{i\mathbf{G}_{hkl} \cdot \mathbf{R}_n} e^{i\mathbf{G}_{hkl} \cdot \mathbf{u}(\mathbf{R}_n)} e^{i\mathbf{q} \cdot (\mathbf{R}_n + \mathbf{u}(\mathbf{R}_n))} S(\mathbf{R}'_n) \\
&= F_{\text{u.c.}}(\mathbf{G}_{hkl}) \sum_n S(\mathbf{R}'_n) e^{i\mathbf{G}_{hkl} \cdot \mathbf{u}(\mathbf{R}_n)} e^{i\mathbf{q} \cdot \mathbf{R}'_n}
\end{aligned} \tag{2.32}$$

The formula we obtained tells us that the 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 multiplied by 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

412 second complex exponential evaluates the phase delay associated to each displaced unit cell  
413 with respect to the center of Bragg peak.

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

- 418 •  $\mathbf{q} \cdot \mathbf{u}(\mathbf{R}_n) = 0$  which means that the projection of the displacements on the vector  $\mathbf{q}$   
419 originating in the  $hkl$  and exploring the vicinity of the Bragg peak, is small. Given the  
420 relatively reduced volume of  $\mathbf{q}$ -space around the Bragg peak that is usually probed in BCDI  
421 this approximation is also reasonable. This is also known as the Takagi approximation  
422 [46]. When this approximation is applied Eq. 2.33 takes the form of the so-called *kinematic*  
423 *sum* and it is implemented in software for simulation of diffraction patterns starting from  
424 atomic positions like, for instance, the scattering module of PyNX [56].
- 425 •  $S(\mathbf{R}'_n) = S(\mathbf{R}_n)$ , which means that the shape function is not altered by the internal  
426 or superficial displacements. This is the case for displacements much smaller than the  
427 lattice parameter, which is easily fulfilled in typical BCDI samples. In practice, this  
428 approximation allows to describe the electron density on a periodic grid.

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

$$F_{\text{fin}}^{\text{strain}}(\mathbf{q}) = F_{\text{u.c.}}(\mathbf{G}_{hkl}) \sum_n \tilde{S}(\mathbf{R}_n) e^{i\mathbf{q} \cdot \mathbf{R}_n} \quad (2.33)$$

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

433 Eq. 2.33 is now a discrete Fourier transform (DFT) of  $\tilde{S}(\mathbf{R}_n)$  calculated over the  $n$  points in  
434 coordinates  $R_n$  of the perfect crystal lattice and is the most important result of the chapter.  
435 Lastly, it is important to mention that when dealing with numerically computed DFT, in 3D,  
436 instead of the point-like  $\mathbf{R}_n$  coordinate, a general coordinate  $\mathbf{r}$  pointing at voxels of size  
437  $\delta r_i = 2\pi/q_i^{\max}$  (where  $q_i^{\max}$  is the extent of the Fourier window along the  $i$ -th dimension) is  
438 used. Moreover, later in the manuscript we will always refer to this complex-valued shape  
439 function as the “*complex object*” or “*reconstructed particle*” and will often deal with its complex  
440 phase.

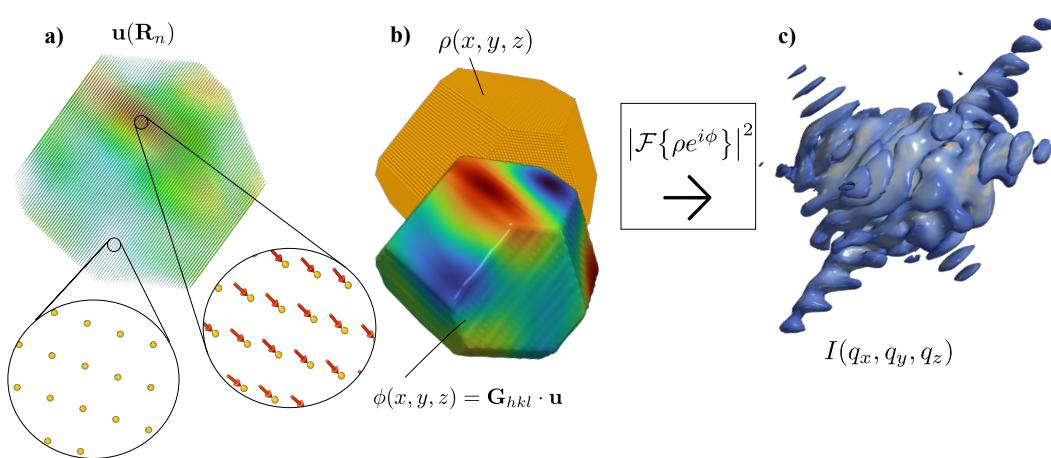
441 Specifically it will be used for simplicity the following relationship, which is derived like  
442 2.6 and drops all the constant terms:

$$I(\mathbf{q}) = |\mathcal{F}\{\rho(\mathbf{r})e^{i\phi(\mathbf{r})}\}|^2 \quad (2.34)$$

443 Eq. 2.34 has the advantage of expressing the diffraction pattern with a DFT, which allows for  
444 fast computations via efficient algorithms, rather than a sum evaluated in each individual unit  
445 cell position. This represents a key ingredient for the most used phase retrieval algorithms in  
446 BCDI (as we will see in the next Chapter), due to the large number of DFT operations required.

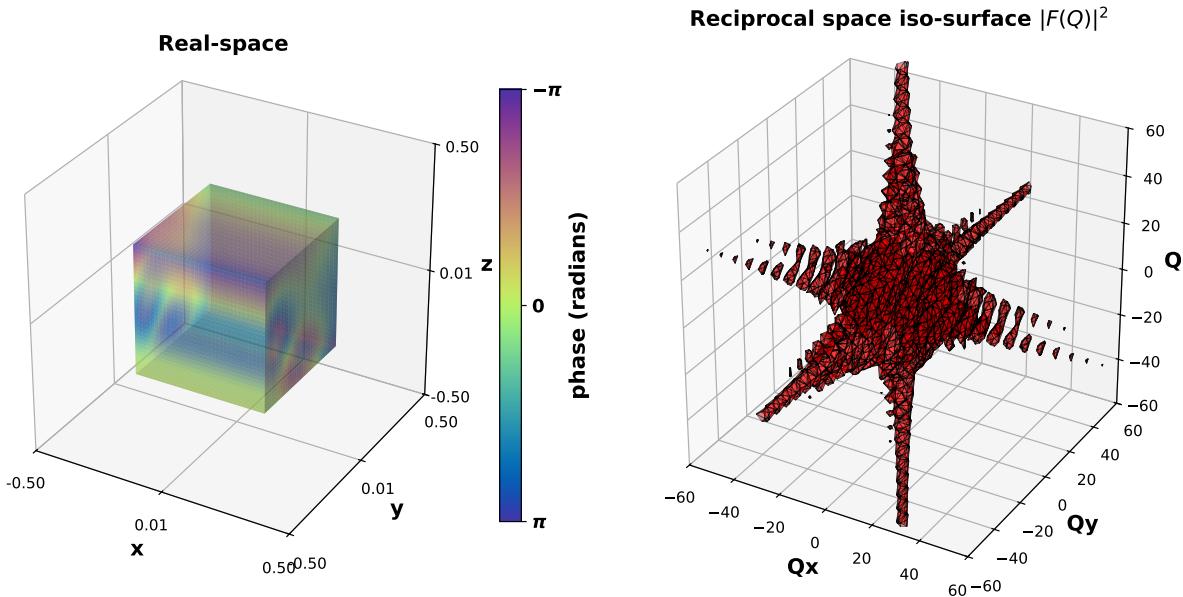
447 Interesting works have evaluated the validity of these approximations [57] and the discrepancy  
 448 between diffraction patterns calculated with the DFT and kinematic sum [58, 59].

449 A schematic representation of the link between the lattice displacements, the complex  
 450 object function and the corresponding diffraction pattern is provided in Fig. 2.5.



**Figure 2.5:** 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.

451 It is interesting here to analyze what the effect of the displacement is on the shape of  
 452 the diffraction pattern. Intuitively we can think that the complex phase term is altering the  
 453 interferences of the scattered waves. If, for a perfect lattice, we had Dirac deltas broadened  
 454 and modulated by the Fourier transform of the shape function, here we have to consider an  
 455 additional broadening due to the strain. Moreover, we know from Friedel's law [60] that the  
 456 diffraction pattern of a real valued function (shape function for perfect crystals, i.e. without  
 457 strain) is always centro-symmetric while here, having introduced a complex term, we expect  
 458 this symmetry to be broken. Fig.2.6 shows the effect of the displacement field on the diffraction  
 459 pattern.



**Figure 2.6:** 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.2.3 for the perfect lattice case.

460 From Eqs. 2.33-2.34 we can intuitively imagine that the distortion of the diffraction pattern is  
 461 linked to (i) the *magnitude* of the displacements, (ii) the *number* of lattice sites involved, (iii) the  
 462 *coherence* of each local contribution  $S(\mathbf{R}_i)e^{i\phi(\mathbf{R}_i)}$  with respect to its neighbors. The interplay  
 463 of these parameters gives rise to many possible distortions of the ideal crystal diffraction.

464 Lattice sites that are displaced with respect to the perfect lattice scatter at slightly different  
 465 Bragg angles. This produces intensity shifted to different  $\mathbf{q}$  values around the Bragg peak. The  
 466 displacements are proportional to the complex phases in the kinematic sum. If the phases are  
 467 the same across many sites, the sum is constructive, and the resulting diffraction signal remains  
 468 localized and peaked. On the other hand, if the phases vary from site to site, the constructive  
 469 interference is lost, and the diffraction pattern becomes more irregular.

470 It follows that the more voxels are involved in incoherent phase variations, the more  
 471 speckled the diffraction pattern becomes, since each contribution departs differently from the  
 472 original Bragg condition. This is often called *heterogeneous* strain. Conversely, in the case  
 473 of equal displacements across many sites, the contributions remain coherent, leading to the  
 474 so-called *homogeneous* strain. The diffraction signal in this case tends to split into two peaks:  
 475 one corresponding to the pristine lattice and one to the uniformly strained lattice.

476 An important case, discussed later in the text, is the high-strain regime, in which a significant  
 477 fraction of lattice sites undergo large heterogeneous displacements. This should be contrasted  
 478 with localized defects such as dislocations or grain boundaries (not treated here), where  
 479 relatively large displacements are confined to only a few lattice sites.

---

### 480 2.2.6 Noise

481 We should now introduce the last ingredient to our derivation, the presence of noise. When  
 482 dealing with real world experiments we have to consider that photons are quantum particles,  
 483 thus their arrivals on the detector surface are random independent events. Here, because of the  
 484 discrete nature of photons, a statistical approach based on the average arrival rate is required.  
 485 It follows that an uncertainty is intrinsically associated to the estimate, hence the emergence  
 486 of a source of noise. The statistical model describing these phenomena is known as Poisson  
 487 statistics [61]. In particular, given a constant *average* rate of arrival  $\eta$  over a unit of time, the  
 488 *expected* number of photons after an exposure time  $\tau$  is  $\mu = \eta\tau$ , and the probability of detecting  
 489  $N$  photons over an observing time  $\tau$  is given by the formula:

$$p(N) = \frac{(\eta\tau)^N e^{-\eta\tau}}{N!} \quad (2.35)$$

490 also called *Poisson probability distribution*. It can be proven that for the Poisson distribution  
 491 the variance equals the mean  $\mu = \sigma^2 = \eta\tau$ . Moreover, we can intuitively think the expected  
 492 number of photons collected during a time  $\tau$  by a pixel in position  $\mathbf{q}_i$  to be proportional  
 493 to the intensity in Eq. 2.34 evaluated in  $\mathbf{q}_i$  and integrated over a time  $\tau$ . At this point, if  
 494 we were to make a measurement, in absence of background, we would have at position  $\mathbf{q}_i$   
 495 an expected integrated intensity  $\mu = \bar{I}(\mathbf{q}_i)$  with an uncertainty of  $\sigma = \sqrt{\bar{I}(\mathbf{q}_i)}$ . Being the  
 496 Signal-to-Noise-Ratio (SNR) defined as  $SNR = \mu/\sigma$  we obtain in our case:

$$SNR = \frac{1}{\sqrt{\bar{I}(\mathbf{q}_i)}} \quad (2.36)$$

497 Though very simple, this equation is important and deserves some comments. First, we  
 498 can observe that in the limit of infinitely long integration times or infinite intensity (infinite  
 499 flux), the  $SNR \rightarrow 1$ , meaning that we would in principle able to deterministically measure  
 500 the diffraction pattern, in this case noiseless. Secondly, we know that most of the intensity  
 501 of a diffraction pattern is concentrated in the center of the Bragg peak (Fig. 2.4), thus we can  
 502 deduce from Eq. 2.36 that Poisson noise is affecting regions at high  $\mathbf{q}$  much more than at the  
 503 center. During PR, this uncertainty in high  $\mathbf{q}$  ranges reflects, in real space, to a lower accuracy  
 504 on fine resolution features of the reconstructed objects. The SNR, by mostly affecting lower  
 505 photon counts regions, typically located at high  $\mathbf{q}$  values, sets a limit to the actual attainable  
 506 direct-space resolution.

## 507 2.3 Practical BCDI at ID01 - ESRF

508 We have so far discussed the theoretical foundations for the understanding of the BCDI tech-  
 509 nique. It is now time to see the practical aspects.  
 510 Starting from the source of the probing radiation downstream to the detectors we will focus on  
 511 the most important stages, necessary to envision the experimental conditions as well as typical  
 512 numbers (resolution, energies, sizes).

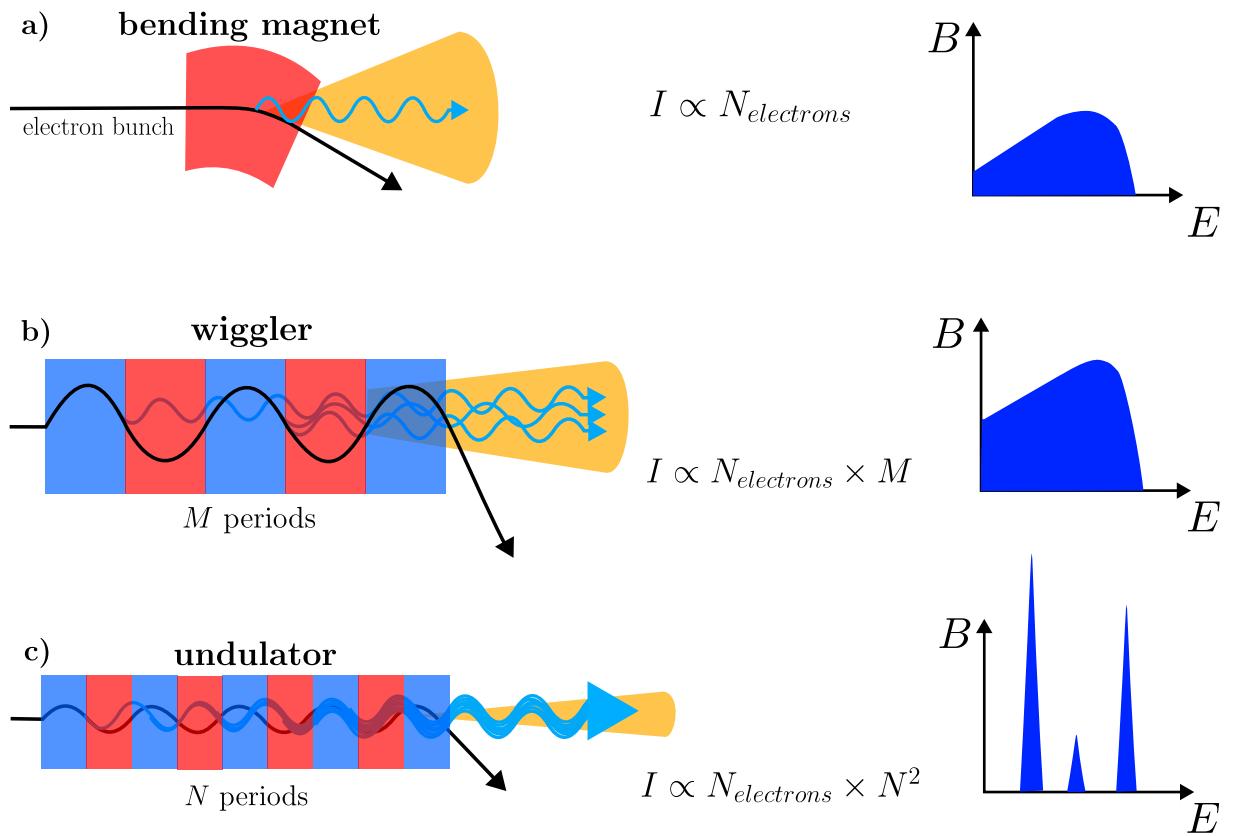
513 BCDI experiments require a *coherent* X-ray beam focussed onto a volume of a few cubic  
 514 microns or less. Moreover, such beams also need to deliver a sufficient amount of photons to

515 the sample, which, being in the micrometer range as well, possesses a limited scattering power.  
516 Such properties today can only be achieved in large scale facilities like synchrotrons and free  
517 electron lasers (FELs).

518 We know that electromagnetic waves can be generated by accelerating charges [62]. In syn-  
519 chrotron facilities electrons travel in a large circular ring (*storage ring*) at relativistic speeds  
520 and X-rays are produced deflecting their trajectories with the help of magnets. Depending on  
521 the magnet size, magnetic field and spatial configuration the X-ray beam is generated with  
522 different properties. Typically, these configurations are grouped into two main categories,  
523 namely: *bending magnets* and the more modern Insertion Devices (ID) like *w wigglers* and *undu-*  
524 *lators*. In synthesis, while bending magnets produce X-rays in a broad energy spectrum with  
525 relatively low intensity, wigglers exploit a series of smaller magnetic dipoles to increase the  
526 intensity of the radiation. Undulators on the contrary leverage the periodicity of the dipoles to  
527 stimulate coherent emission, hence sharpening the energy bandwidth around tunable peaks  
528 and increasing the intensity as well. The radiation generated by the latter is the most suited  
529 for coherent diffraction experiments because of its inherent higher coherence. An illustrative  
530 explanation is provided by Fig. 2.7 while an exhaustive description of synchrotron radiation  
531 can be found in the notable book from Prof. Giorgio Margaritondo [63].

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

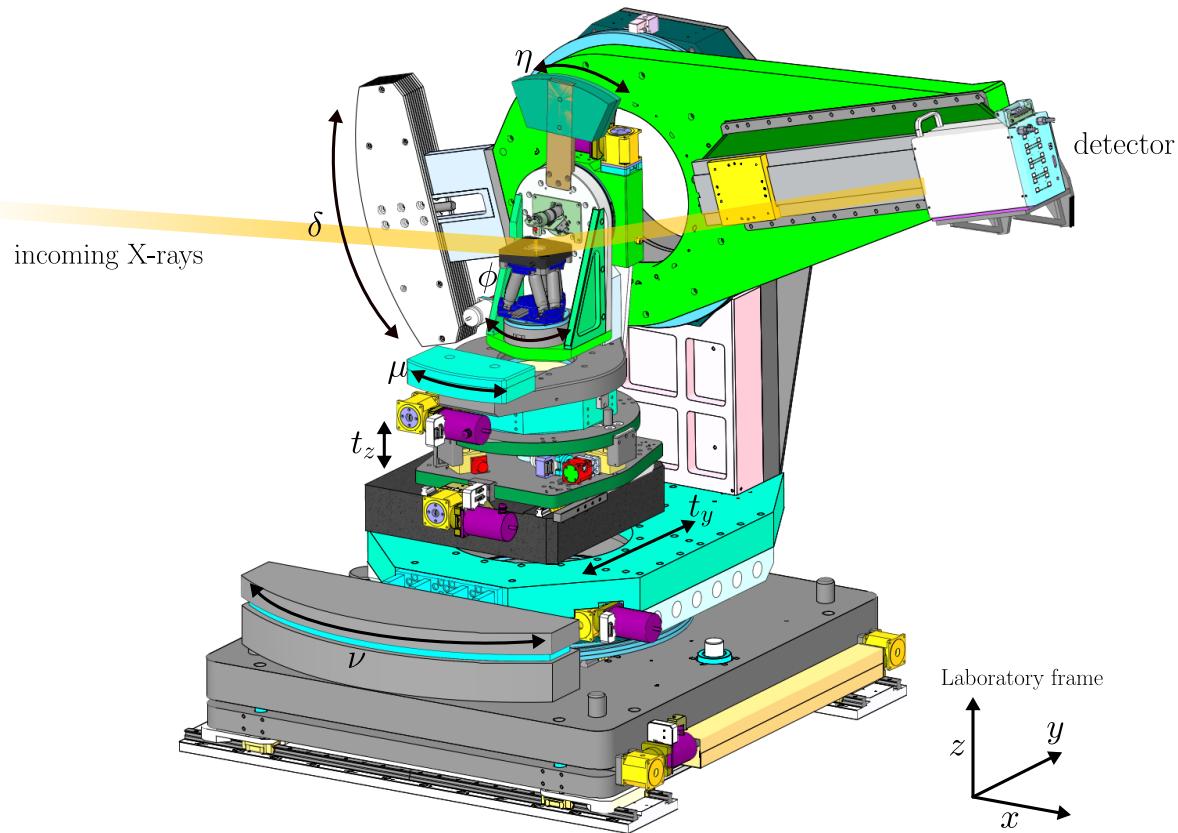
537 At the European Synchrotron Radiation Facility (ESRF) the ID01 beamline was conceived to  
538 combine the lattice parameter resolution provided by X-ray diffraction with spatial resolution  
539 of imaging techniques [27] and is able to offer various techniques for the investigation of strain  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



**Figure 2.7: 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 transverse coherence) as well as higher intensity and narrower bandwidths. Adapted from [64].

### 541 2.3.1 Beam size

542 Typical BCDI experiments are focused on single nanoparticles. The diffraction pattern is the  
 543 result of the scattering of a single crystal in Bragg condition. It is therefore necessary to have  
 544 a beam size of the order of the size of the crystal. At ID01 the beam size can scale down to  
 545  $35 \times 35\text{nm}$  [65], but typical sizes range between  $0.5 - 1\mu\text{m}^2$ . A too large beam could shine on  
 546 some neighbor particles which, if in Bragg condition, could contaminate the diffraction pattern.  
 547 These often appear as isolated bright spots, called *aliens* [66]. On the contrary, regions of the  
 548 same particle that are not illuminated do not contribute to the diffraction pattern, therefore are  
 549 not imaged in the reconstructed objects.



**Figure 2.8:** Computer Aided Design of the diffractometer at the ID01 beamline with the corresponding degrees of freedom. Adapted from [44]

### 550 2.3.2 Energy and flux

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

$$B = \frac{\text{nb photons}}{dt \cdot dA \cdot d\Omega \cdot 0.1\% dE/E} \quad (2.37)$$

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

---

<sup>1</sup>This is related to the concept of the *oversampling condition* discussed in the next chapter

561    **2.3.3 Coherence**

562    This fundamental property of electromagnetic radiations is probably the most important for  
 563    CDI. In the derivation of the scattering equation Eq. 2.1 and following, we have always assumed  
 564    the incident radiation to be described by a plane wave, perfectly monochromatic ( $\Delta\lambda \neq 0$ ) and  
 565    with no angular spreading ( $\Delta k = 0$ ). In the ideal case, the wave-field is perfectly coherent,  
 566    meaning that the relative phase between any two points in space is completely well-defined. In  
 567    reality, electromagnetic waves have finite temporal duration and therefore, by Fourier transform  
 568    relations, a finite spectral bandwidth. Likewise, they are produced by extended sources and  
 569    observed at finite distances, which implies a finite angular spread of the wave-vectors. These  
 570    limitations introduce phase decorrelation between fields evaluated at two different points, either  
 571    *along* the propagation direction (due to bandwidth) or *across* it (due to angular dispersion). For  
 572    this reason, it is useful to introduce quantities that characterize the “degree of coherence” of  
 573    the radiation. An intuitive approach is to define a *longitudinal coherence length*, associated with  
 574    phase delays from spectral bandwidth, and a *transverse coherence length*, associated with phase  
 575    delays from angular spread. A pictorial view of these characteristic lengths is provided by Fig.  
 576    2.9

577

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

$$N\lambda = (N - 1)(\lambda + \Delta\lambda) \quad (2.38)$$

thus,  $\Delta\lambda(N - 1) = \lambda$

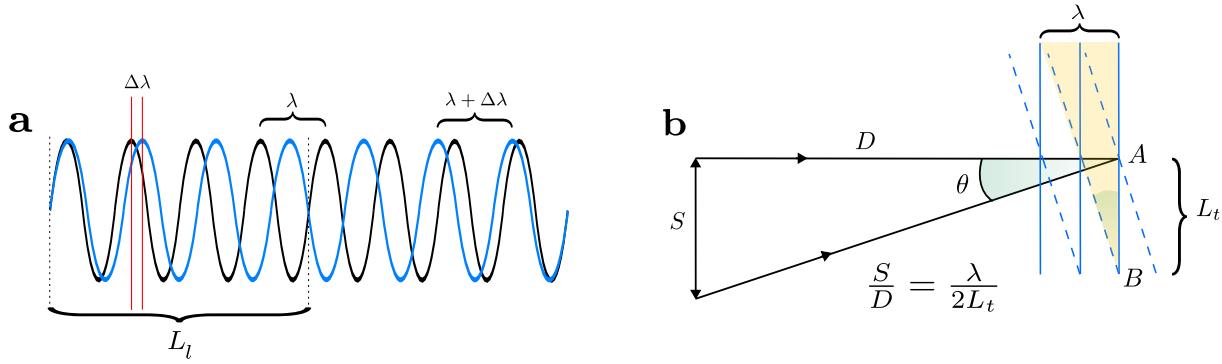
583    In case of small  $\Delta\lambda/\lambda$  we can approximate  $N - 1 \approx N$ , and obtain  $N \approx \lambda/\Delta\lambda$ . This leads  
 584    us to the expression for the longitudinal coherence length:

$$L_l \approx \frac{\lambda^2}{2\Delta\lambda} \quad (2.39)$$

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

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

$$L_t \approx \frac{\lambda D}{2S} \quad (2.40)$$



**Figure 2.9:** **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. Adapted from [43]

591     Also in this case we see the coherence decreasing for high energies. Moreover, the dependence  
 592     on  $S$  and  $D$  explains the challenges to build long beamlines (118 meters for ID01) and  
 593     reduce the electron beam size and angular divergence. The transverse coherence length can be  
 594     easily lifted to a coherence surface for a source extended in 2D ( $L_t^{hor} \times L_t^{ver}$ ).  
 595

596     Given these premises we can now calculate the coherence volume ( $L_l \times L_t^{hor} \times L_t^{ver}$ )  
 597     available ID01 for a typical X-ray energy used in BCDI. Given the source sizes of  $60\mu m$  horizontally  
 598     and  $15\mu m$  vertically, at 8 keV ( $\lambda = 1.55 \text{ \AA}$ ) we obtain a transverse coherent surface  
 599     of  $152\mu m \times 610\mu m$ . The longitudinal coherence length is found to be approximately  $0.8\mu m$   
 600     considering the  $\Delta\lambda/\lambda \sim 10^{-4}$  typical of the double Si (111) crystal monochromator. It follows  
 601     that often, in BCDI experiments, the limitation on the particle size is given by the longitudinal  
 602     coherence [67].  
 603

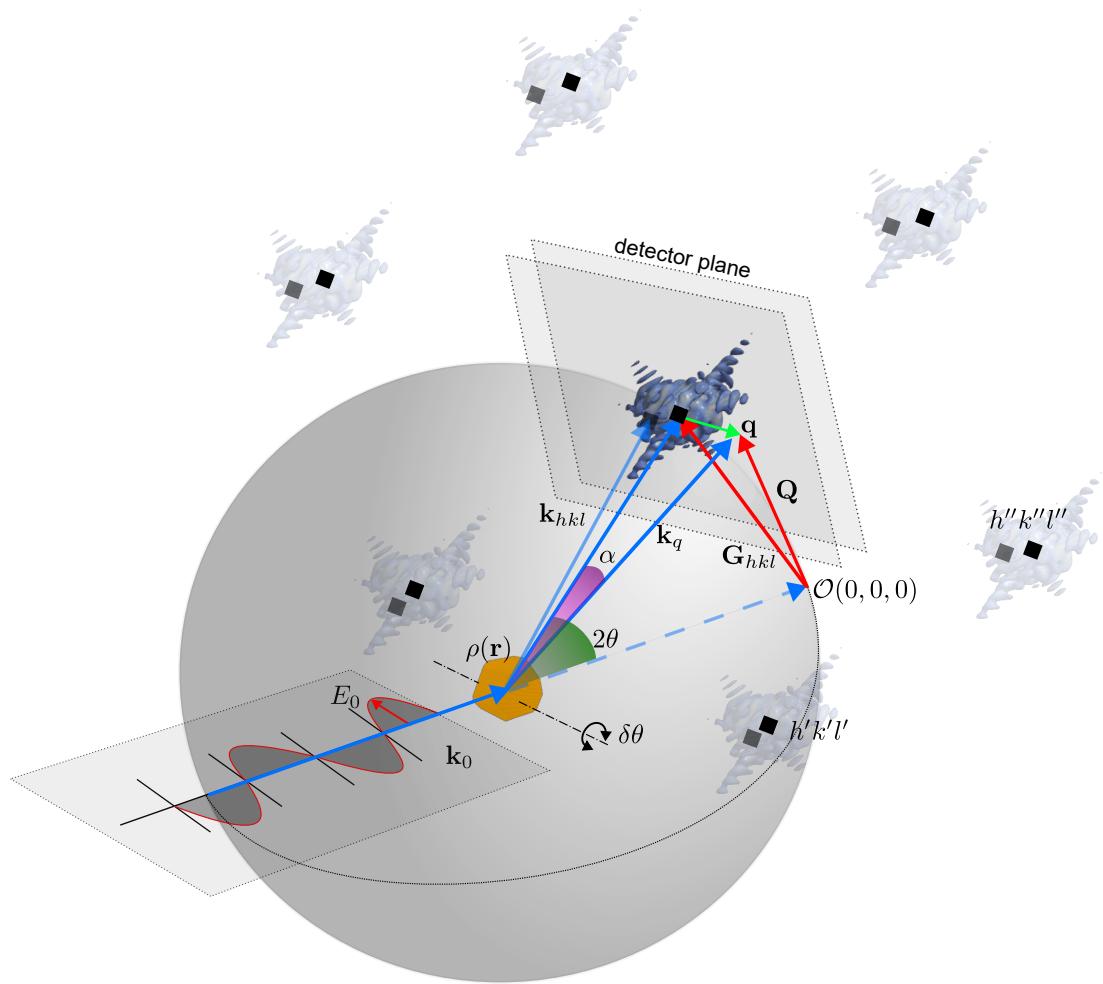
604     The coherence volume specifies the maximum spatial separation between two scatterers for  
 605     which their scattered waves remain mutually coherent and can interfere. For this reason, while  
 606     classical X-ray diffraction is limited to the interference of a few lattice points, in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

### 611     2.3.4 Ewald sphere and Rocking curves

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

615     We have said that in BCDI we focus on a single  $hkl$  node of the reciprocal lattice, and we  
 616     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 a sphere is called the *Ewald sphere* and it is illustrated in Fig. 2.10. 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 2.10:** Illustration of the typical geometry for a BCDI experiment and the Ewald sphere (not to scale). A monochromatic plane wave approximates a coherent X-ray beam that fully illuminates a sample with a wave-vector  $\mathbf{k}_i$ . A diffraction pattern given by Eq. 2.33 is formed around each  $hkl$  node. Of the different reciprocal lattice nodes a single one is selected for the measurement, depending on the detector. The Ewald sphere, of radius  $\mathbf{k}_{hkl}$  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  $\eta$  angle in Fig. 2.8, 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 the 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 one can consider that: geometrically the contact angle is equivalent to the angle  $\alpha$  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

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



## 644 THE PHASE PROBLEM IN BCDI

645 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  
646 the field of X-ray crystallography since the first measured diffraction patterns, but similarly  
647 affects other domains like astronomical and seismic imaging as well as the coherent diffraction  
648 imaging of our interest. As anticipated briefly in the preface, the phase problem arises from  
649 a technical limitation. In X-rays, the fast oscillations of the electromagnetic fields induce  
650 detectors to only measure a time-averaged intensity (Eq.2.5) with the consequent loss of the  
651 phase information in the measurement.

653 The Fourier phase problem is therefore the impossibility to compute the complex-valued  
654 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  
655 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  
656 thus necessary to find alternative strategies, often based on iterative algorithms, to perform  
657 the Phase Retrieval (PR) and recover the signal. However, as one can suspect, the modulus  
658 operation applied to the Fourier transform allows in principle an infinite variety of complex  
659 functions to be solution. For this reason the problem is said to be *ill-posed*. Consequently,  
660 the modulus operation renders the optimization landscape *non-convex*, so that phase retrieval  
661 algorithms must search for the desired solution (the *global minimum*) within a space populated  
662 by multiple “valleys” (*local minima*), where they may become trapped. In this context, the  
663 search for the solution to the problem has fascinated (and still does) scientists for decades,  
664 contributing to an extensive production of works in literature.

665 The first published studies date back to 1951 when Sayre, in a comment [68] to the paper  
666 by Shannon *Communication in the presence of Noise* [69], proposed a condition on the sampling  
667 of the diffraction pattern for the restoration of the unit cell extent. Later in 1972 Gerchberg  
668 and Saxton [70] developed an algorithm capable of inverting the diffraction pattern that is  
669 nowadays at the basis of currently used standard PR algorithms. However, a theoretical analysis  
670 of the uniqueness conditions was provided only later by Bruck and Sodin (1979) [71]. The  
671 authors showed that, for 2D and 3D problems, the phase retrieval has unique a solution except  
672 for rare cases, therefore conferring the mathematical solidity to the algorithm’s results. Later in  
673 1982 Bates draws the link between uniqueness and the Sayre sampling intuition, as necessary  
674 condition for 2D case [72]. A refined version of the Gerchberg - Saxton algorithm was proposed  
675 by J.R. Fienup in 1978 [1] who named it Error Reduction (ER). In [2], published in 1982, the  
676 same author developed the Hybrid-Input Output (HIO) algorithm, able to outperform ER, and

677 compared gradient-descent methods as well. In 1987 again Fienup showed the possibility  
678 of reconstructing *complex-valued* objects if the constraints on the object support are “tight”,  
679 i.e. the shape of the object is known [73]. This result is particularly interesting for BCID  
680 since, as we have seen in Eq. 2.34, the object to be retrieved is complex-valued. Based on  
681 the suggestion of Sayre in 1991 [74] the works of Miao and coauthors from 1998 opened the  
682 X-ray coherent diffraction imaging field by exploiting phase retrieval, combining the sampling  
683 proposed by Sayre and iterative algorithms developed by Fienup [4, 75, 76]. Among the  
684 many works published later it is worth mentioning the Difference Map algorithm [77], which  
685 generalized Fienup’s HIO introducing an additional non-linear term and the (Relaxed) Averaged  
686 Alternating Reflections algorithms developed by Luke [3, 78] with better convergence properties  
687 guaranteed by reflection operations. Later in 2004 the work of Rodenburg and Faulkner [79]  
688 showed improved PR for diffraction patterns obtained illuminating the sample from multiple and  
689 partially overlapping regions. It was the birth of ptychography, later successfully demonstrated  
690 in Bragg condition as well [80]. Though not usually employed in CDI because of the large  
691 computational costs, it is worth mentioning the more recent works of Candès and coauthors  
692 who have developed, the PhaseLift algorithm [81] and the “Wirtinger flow” [82]. While the  
693 former turns the PR into a *convex* problem, thus improving the stability and convergence  
694 guarantees, in the latter the PR is solved as least square problem with gradient descent using  
695 the Wirtinger derivatives for complex functions and a *spectral initialization* method that enables  
696 the start of the PR near the global minimum.

697 Further details on the applications of PR algorithms can be found in the review by Shechtman  
698 [83] while recent developments and theoretical insights can be found in the work of Fannjiang  
699 and Strohmer [84]. Of great help for a unified overview of the main PR algorithms with  
700 comparisons is the work of Marchesini [85].

701 Here we will present first the sampling condition, the phase ambiguities and the main  
702 alternating projections algorithms that will be encountered later in the chapters dedicated to  
703 the results.

## 704 3.1 Oversampling

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

$$O(x) = \rho(x)e^{i\phi(x)} \quad \tilde{O}(q) = A(q)e^{i\varphi(q)} \quad (3.1)$$

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

711 If  $O(x)$  is a square of size  $R$ , Nyquist theorem states that each point sampling  $\tilde{O}(q)$  should  
712 have a spacing  $\Delta q = 1/R$ . In our case we measure  $|\tilde{O}(q)|^2$  which, corresponds to the Fourier  
713 transform or the so-called *autocorrelation function* (or *Patterson map* [86]) of the object  $(O(-x)*$   
714  $O(x))$ , which extends over a size  $2R$ . Hence, the sampling should happen every  $\Delta q = 1/2R$   
715 to recover the autocorrelation of  $O(x)$  without aliasing. This should in principle contains the  
716 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 [4] 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  $\delta q$  is the extent of a single pixel. The  $\mathbf{q}$  vector of Fig.2.10 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.

$$\tilde{O}(q_k) = \sum_{n=0}^{N-1} O(x_n) e^{i \frac{q_k r_n}{N}} = \sum_{n=0}^{N-1} \rho(x_n) e^{i\phi(x_n)} e^{i \frac{q_k r_n}{N}} \quad (3.2)$$

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. Now, by using the 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:

$$\sigma = \frac{\text{total pixel number}}{\text{unknown-valued pixel number}} \quad (3.3)$$

In the 2D or 3D case the same factor 2 needs to be fulfilled in order to have an (over)determined system of equations. However, one can calculate an oversampling ratio along each dimension  $d$ , resulting to be  $\sigma \geq 2^{1/d}$  [87]. Nevertheless, it is preferable to ensure a larger value for  $\sigma$  along each dimension for better reconstructions [88, 89].

736

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.2.10), meaning that the same  $\Delta q$  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.2.10. We can therefore write:

$$\delta q = |k_q| \frac{p_{ix}}{D} \quad (3.4)$$

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.

$$\delta q \leq \frac{\delta q_{\text{fringe}}}{2} \quad (3.5)$$

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. In practice a spacing of at least three pixels is always aimed during experiments to ensure easier invertibility.

## 3.2 Phase Symmetries

We have seen how the oversampling condition is fundamental for the uniqueness of the solution of the Phase Problem. However, it is important to point out that this uniqueness is valid except for some trivial reciprocal space phase ambiguities [2, 90], or symmetries, that correspond to equivalently good solutions in direct space. They are here listed:

- **Object translation - Phase ramp:** A translation of  $\mathbf{x}$  from the center  $\mathbf{x}_0$  of the object in direct space will correspond in Fourier space to the addition of a linear phase ramp to the reciprocal space phase  $\varphi(\mathbf{q}) = \varphi(\mathbf{q}_0) + \mathbf{q} \cdot \mathbf{r}$ . The physical meaning is unaltered since the object itself has not changed but its absolute position in space has. Equivalently, a translation of the Bragg peak in reciprocal space yields a phase ramp applied to the object in direct space. Practically, if during PR the diffraction pattern was not centered, the reconstructed object presents a non-physical phase ramp artifact that if not removed can induce a misinterpretation of the strain field.
- **Global phase shift - Constant offset:** The addition of a constant offset to the reciprocal space phase  $\varphi(\mathbf{q}) = \varphi(\mathbf{q}) + \psi$  corresponds to a global phase shift applied to the object  $e^{i\psi} O(\mathbf{r})$ , which is physically equivalent to  $O(\mathbf{r})$ . In fact, the global phase shift does not affect the strain, as this is calculated from the gradient of the phase. This equivalence is expressed in Fourier space by the unaltered squared modulus  $|A(\mathbf{q})e^{i\varphi(\mathbf{q})}|^2 = |A(\mathbf{q})e^{i(\varphi(\mathbf{q})+\psi)}|^2 = |A(\mathbf{q})|^2$ .
- **Object inversion - Sign symmetry:** The geometric inversion of the complex conjugate of the object  $O = \rho(\mathbf{r})e^{i\phi(\mathbf{r})}$  given by  $O' = \rho(-\mathbf{r})e^{-i\phi(-\mathbf{r})}$  corresponds to a sign flip of the reciprocal space phase  $\varphi(\mathbf{q}) = -\varphi(\mathbf{q})$ . The overall diffracted intensity (information is lost in the modulus squared operation), the object  $O$  and its *twin image*  $O'$  are equivalently good solutions as the information about their absolute orientation in space is lost. It can however happen, especially for nearly centro-symmetric objects that this symmetry is not broken automatically during PR and that the algorithm stagnates on the superposition of, the object and its twin. This combination of  $O$  and  $O'$  is not a solution to the phase problem [91] and therefore de-twinning strategies have to be employed [92].

It follows that, during multiple PR runs of the same BCDI data, equivalent reconstructed objects can differ between each other by additional linear phases, as well as spatial translations, shifted phase ranges and inverted complex conjugation.

### 787 3.3 Alternating projections algorithms

788 In this section the class of algorithms known as “alternating projections” (AP) mentioned above  
 789 is presented, and the three most used algorithms in BCDI are described in more detail. We  
 790 invite the reader to refer to the more exhaustive lecture notes by Cegielski [93] or the review  
 791 written by Marchesini, from which the following paragraphs take inspiration [85].

792

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

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

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

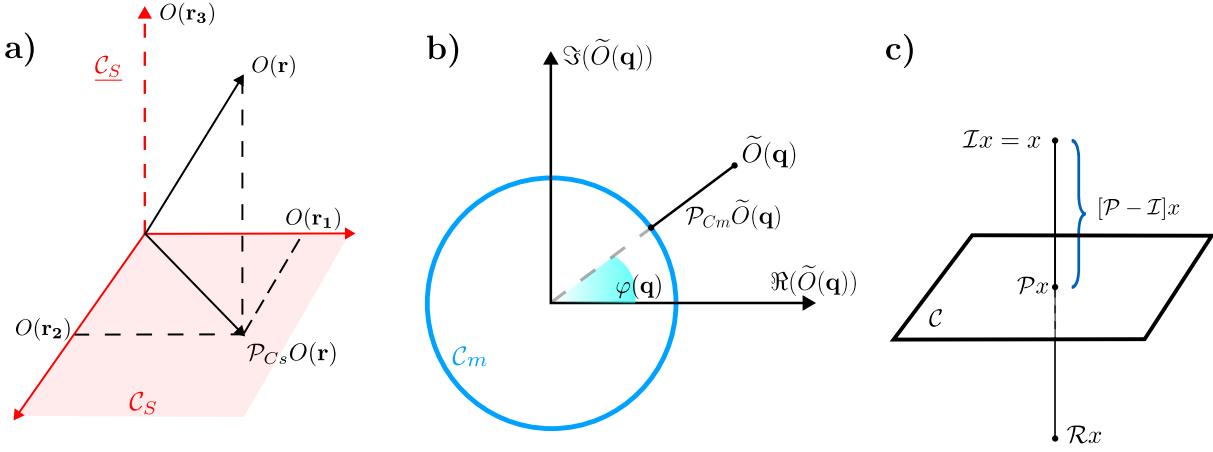
- 799 •  $\mathcal{C}_s = \{O(\mathbf{r}) \in \mathcal{H} : O(\mathbf{r}) = 0 \quad \forall \mathbf{r} \notin \mathcal{S}\}$  Often called “support constraint” is the set  
 800 containing all objects with zero amplitude outside the *support*  $\mathcal{S}$ . This last, in BCDI,  
 801 coincides with the shape function encountered in Eq.2.18 and it is in principle unknown.
- 802 •  $\mathcal{C}_m = \{O(\mathbf{r}) \in \mathcal{H} : |\mathcal{F}\{O(\mathbf{r})\}| = m\}$  Often called “modulus constraint” is the set  
 803 containing all objects with Fourier transform of modulus  $m$ . This set is however “non-  
 804 convex” as  $|\mathcal{F}\{O(\mathbf{r})\}| = m$  is fulfilled for any reciprocal space phase. This poses  
 805 challenges for deriving the convergence criterion of AP operating on this set [94].

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

$$\text{find the object } O(\mathbf{r})^* \in \mathcal{C}_s \cap \mathcal{C}_m \quad (3.6)$$

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

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



**Figure 3.1:** Geometrical representation of (a) the projection of the object onto the support constraint set, (b) the projection of the object onto the Fourier modulus constraint set, (c) the identity, projector and reflector operators. Inspired by [85]

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

$$\mathcal{P}_{C_s}(O(\mathbf{r})) = \begin{cases} O(\mathbf{r}), & \mathbf{r} \in \mathcal{S} \\ 0, & \mathbf{r} \notin \mathcal{S} \end{cases} \quad (3.7)$$

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

$$\mathcal{P}_{C_m}(\tilde{O}(\mathbf{q})) = \mathcal{P}_{C_m}(A(q)e^{i\varphi(q)}) = \sqrt{I(\mathbf{q})}e^{i\varphi(q)} \quad (3.8)$$

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

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

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

### 828 3.3.1 Error Reduction (ER)

829 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  
830 inverse Fourier transform of the squared root of the measured intensity with a random complex  
831 phase array  $\varphi^0(\mathbf{q})$ , we can express the object at the  $k$ -th iteration of the *Error Reduction* (ER)  
832 algorithm as:

$$O^{k+1}(\mathbf{r}) = \mathcal{P}_{C_m}\mathcal{P}_{C_s}(O^k(\mathbf{r})) \quad (3.9)$$

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

$$\varepsilon_m(O) = \|\mathcal{P}_{Cm}(O) - O\|_{L2} \quad (3.10)$$

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

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

$$O^{k+1}(\mathbf{r}) = \mathcal{P}_{Cs}(O^k(\mathbf{r})) - \frac{1}{2} \nabla_s \varepsilon_m^2(O^k(\mathbf{r})) \quad (3.11)$$

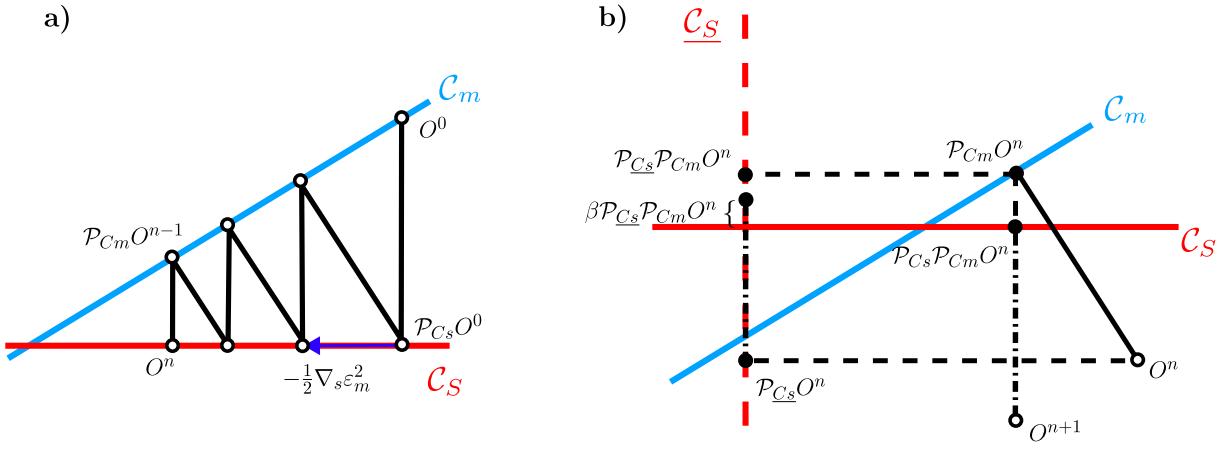
845 which makes explicit the equivalence between ER and steepest descent projected onto the  
 846 support constraint set, with a fixed step size of  $1/2$ .

### 847 3.3.2 Hybrid Input-Output (HIO)

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

$$O^{k+1}(\mathbf{r}) = \begin{cases} \mathcal{P}_{Cm}(O^k(\mathbf{r})), & \mathbf{r} \in \mathcal{S} \\ (\mathcal{I} - \beta \mathcal{P}_{Cm})(O^k(\mathbf{r})), & \mathbf{r} \notin \mathcal{S} \end{cases} \quad (3.12)$$

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



**Figure 3.2:** Geometrical representation of (a) 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 the estimate  $O^0$  is near the solution (b) geometrical representation of the HIO algorithm. Inspired by [85]

### 860 3.3.3 Relaxed Averaged Alternating Reflections (RAAR)

861 Another AP algorithm typically used in CDI is the Relaxed Averaged Alternating Reflections  
 862 (RAAR) developed by Luke in 2004 [3]. According to this algorithm the object at the  $k$ -th  
 863 iteration is:

$$O^{k+1}(\mathbf{r}) = \beta \frac{1}{2} (\mathcal{R}_{Cs} \mathcal{R}_{Cm} + \mathcal{I})(O^k(\mathbf{r})) + (1 - \beta) \mathcal{P}_{Cm}(O^k(\mathbf{r})) \quad (3.13)$$

864 where  $\beta \in [0, 1]$ . Eq.3.13 can be split into two terms. The first term, known as Average  
 865 Alternating Reflection (ARR) [78] acts like an average of the current estimate and its reflection  
 866 across both constraints sets. The alternated reflections search for the intersection of the two  
 867 sets, which is a fixed point of the problem, while exploring more broadly the solution space as  
 868 they do not project directly on the constraint sets. The second term is a relaxation term that  
 869 projects the current estimate on the modulus set, like in the HIO and ER.

870 In short, one could see the RAAR as a controlled (through  $\beta$ ) balance of exploration and  
 871 stability.

### 872 3.3.4 Support update

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

$$A(\mathbf{r}) = \mathcal{F}^{-1}\{I(\mathbf{q})\} \quad (3.14)$$

878     However, the autocorrelation extends over a region roughly twice the linear size of the true  
 879     object (see Fig. 3.4).

880     An important step forward was introduced by Marchesini in 2003 [95]: the *shrinkwrap*  
 881     algorithm, which adaptively refines the support during reconstruction. After a given number  
 882     of phase retrieval iterations, the modulus of the current object estimate is convolved with  
 883     a Gaussian kernel and subsequently thresholded (typically at  $\sim 20\%$  of the maximum value),  
 884     yielding a binary mask that defines the updated support.

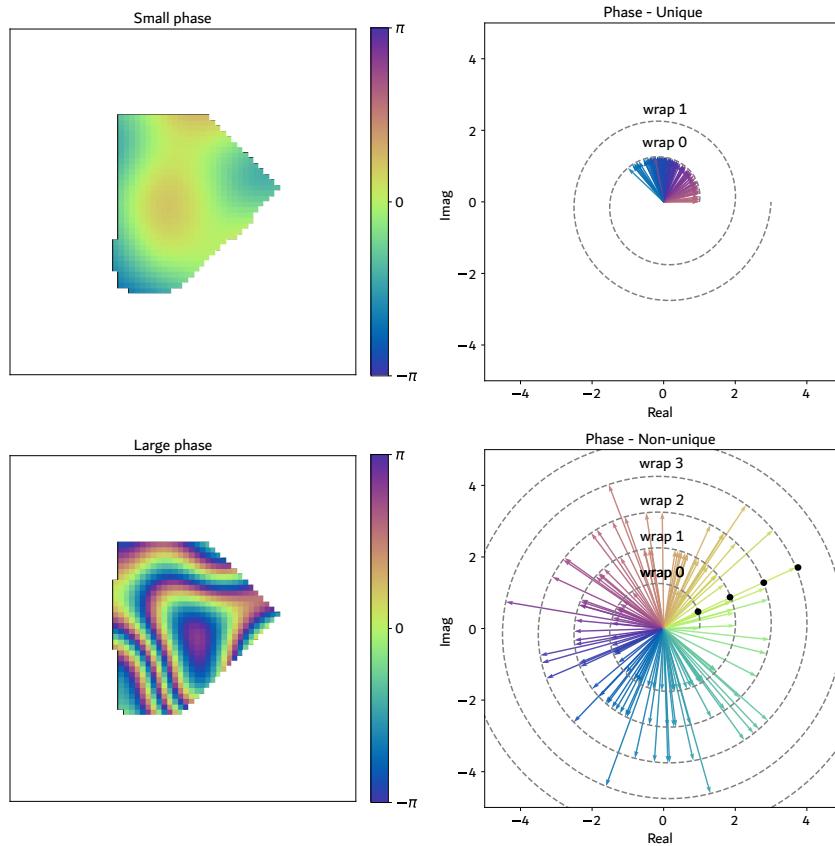
885     This adaptive support refinement was shown to significantly improve convergence and  
 886     reconstruction quality in many experimental cases (depending on the signal-to-noise ratio and  
 887     choice of threshold), and it is now a standard component of BCDI phase retrieval pipelines.  
 888

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

### 891     3.4 BCDI Phase Retrieval in presence of high-strain

892     At this point it is interesting to understand how and why the presence of high-strain complicates  
 893     the PR. and therefore postulate on the possible strategies that can be adopted to regularize the  
 894     problem and facilitate convergence.

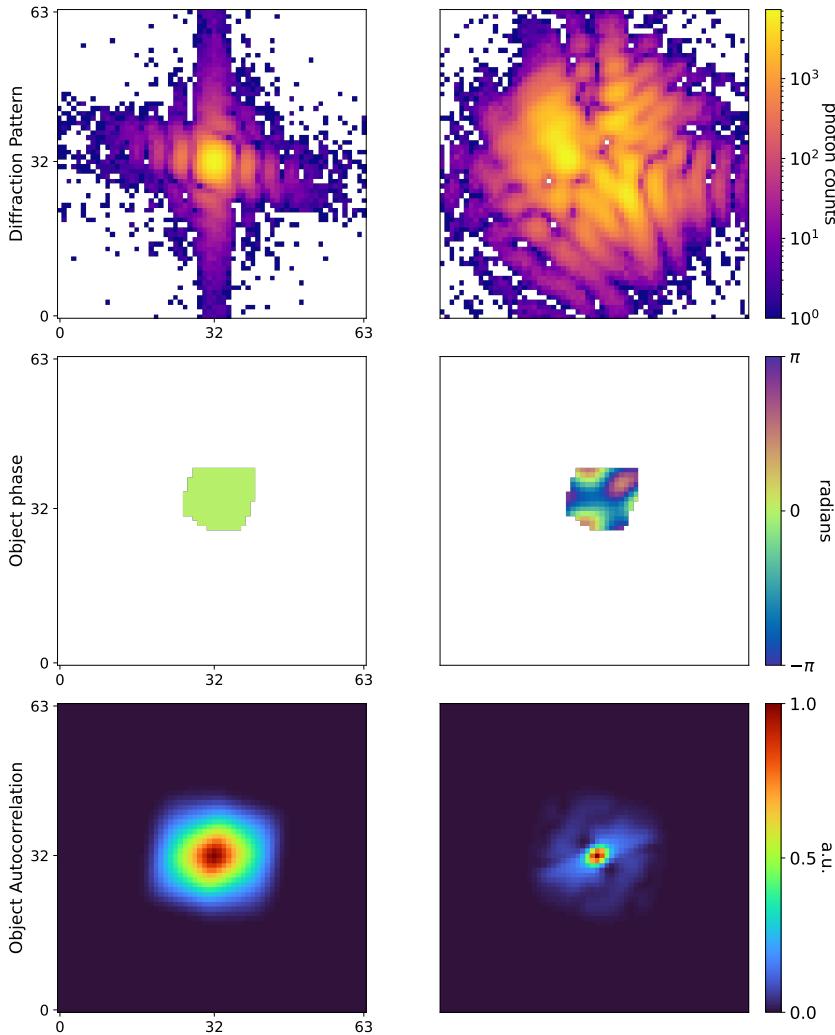
895     From a pictorial point of view it could help to see the problem as follows. If we consider  
 896     each voxel constituting the object as a complex vector made of modulus (normalized between 0  
 897     and 1) and phase, we can plot the full object as the ensemble of these complex vectors lying on  
 898     the Argand plane, where modulus and phase are also seen on the Cartesian real and imaginary  
 899     axes (see Fig.3.3). For small phases we would see the vectors spreading over a limited range of  
 900     the unitary circle. On the contrary large phases will cause the vectors to spread even more to  
 901     the full circle and eventually extend to multiple phase wraps. At this point, if the phases are  
 902     bounded within the  $2\pi$  range, each vector’s angle is uniquely determined by its coordinates  
 903     on real and imaginary axis. On the contrary, if the range exceeds  $2\pi$ , some of these vectors  
 904     with phase  $\phi$  may overlap with some other vectors whose phase is  $2\pi + \phi$ , thus making each  
 905     point on the unitary circle no longer uniquely assigned to a single phase. This degeneracy is  
 906     responsible for the presence of additional local minima that can hinder the PR.



**Figure 3.3: Pictorial visualization of phases on the Argand plane.** The figure provides an intuitive illustration of the origin of additional local minima that arise for large phase ranges. Each voxel is represented by an arrow vector directed according to its phase, unwrapped along a spiral line. This representation highlights that objects with small phase variations (within  $2\pi$ ) correspond to solutions of the (PR) problem that lie on the 0-th wrap, where each phase value is uniquely determined. In contrast, objects with large phase ranges ( $> 2\pi$ ) extend the solution space to higher-order wraps, creating degeneracies with lower-order wraps ( $\phi = \phi + 2\pi \times \text{wrap}$ ). This degeneracy in the solution space increases the ambiguity of the phase problem.

907 In the literature it is known that particles with phase range beyond  $2\pi$  are more challenging  
 908 to reconstruct and these occurrences are often referred to as “strong-phase” or similarly “high-  
 909 strain” cases [96–98]. Although strictly related, these two concepts are different, since the strain  
 910 is ultimately given by the *gradient* of the phase. From the PR perspective, it is the strong-phase  
 911 complicating the PR. Practically, given the finite number of voxels over which the phase can  
 912 extend, the strong-phase case coincides to the high-strain case. Later in the text the two terms  
 913 will be used interchangeably.

914 Another reason that can practically hamper the PR is difficult initialization of the algorithms.  
 915 In fact, it is interesting to notice how the autocorrelation changes for highly strained particles  
 916 and how this can affect the PR algorithms initialization and consequent convergence. Fig. 3.4  
 917 depicts two simulated diffraction patterns from a particle with the same support but different  
 918 applied strain fields. While in the first case (Fig. 3.4a) the low-strain does not distort the  
 919 diffraction pattern and therefore the autocorrelation function, beside the sharp central peak, is  
 920 uniform and its support well-defined. On the contrary, in Fig. 3.4b, the high-strain distorts the  
 921 diffraction pattern and the calculated autocorrelation presents a poorly homogeneous support.



**Figure 3.4: Object autocorrelation** **a)** In case of low strain the object autocorrelation function has uniform amplitude outside the central peak, with well-defined support. One can notice that it clearly extends over a domain that is double the size of the object. In this case the sharp autocorrelation support edges facilitate the shrink-wrap algorithm to extract the support. To the same particle a large phase was applied **b)** and the calculated autocorrelation shows a sharper central peak and a much less intense and inhomogeneous surrounding region. It follows that in this case a lower shrink-wrap threshold needs to be adopted in the initialization. However, this can include holes in the support which might be an effect of the large phase and not physical.

922 The result of the local minima and the hard estimate of the object support make such  
 923 that conventional PR algorithms have to be launched multiple times (in the order of tens or  
 924 hundreds normally), with different initializations. In this way the probability of landing near  
 925 the solution is increased, and the convergence is more likely. This is however computationally  
 926 expensive and time-consuming.

927 In the years, and during this PhD as well, different approaches aiming at improving the PR  
 928 of highly-strained crystals have been developed, as it still represents a major limitation of  
 929 the BCDI technique. Solutions that employ the use of structured 2D modulators placed before  
 930 or after the sample have been studied by respectively Calvo *et al.* [99] and Zhao *et al.* [100].  
 931 These methods allow to introduce an additional constraint, given by the modulator, during

the PR. From the algorithmic perspective one often seeks regularization of the problem with some prior knowledge. Typically, the assumption of homogeneous electron density inside the support is leveraged with uniformity constraints during PR [59, 101, 102] or object's modulus minimization in  $\mathcal{L}_1$  norm [97], or guiding the algorithm by promoting objects with uniform and sharp modulus [103]. Recent studies have also explored the combination of genetic algorithms with standard PR [104] for improved exploration of the solution space. In other cases the additional knowledge is provided by multiple and different measurements of the same sample. For instance multiple Bragg reflections of the same particle can be phased simultaneously [105] to constrain the solution to a unique reconstruction fulfilling each measurement. Alternatively, if the sample undergoes phase transitions inducing high-strain, the support retrieved at low-strain level can be fixed across the transition during the PR of high-strain states [106].

The regularization of the strongly ill-posed PR for highly strained crystals with prior knowledge is the key to successful reconstructions. Another type of strategy concerns the use of *learned* priors, i.e. that emerge from the properties of the learned statistical distributions of the particular class of inverse problems. It is in this framework that Convolutional Neural Networks models presented in Chapter 6 were conceived.

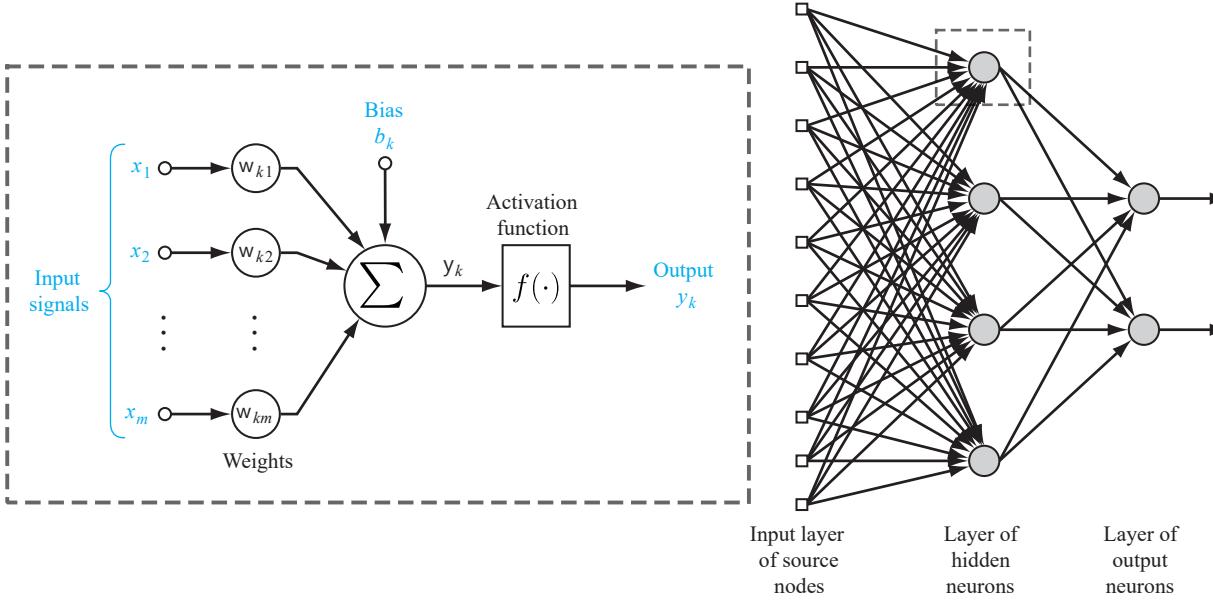
## 949 KEYNOTES ON CONVOLUTIONAL NEURAL NET- 950 WORKS

951 In this chapter a short overview on the basic concepts of Convolutional Neural Networks  
952 (CNNs) will be given. The scope is to give the necessary background for the understanding of  
953 the structure and the motivations behind the Deep Learning (DL) models employed for the  
954 analysis of BCDI data, presented in the next chapters.

955 For more comprehensive and exhaustive dissertations about Machine Learning (ML) and  
956 Artificial Neural Networks (ANNs) the book of Goodfellow [107] and the more recent from  
957 Prince [108] are suggested to the reader.

### 958 4.1 Artificial Neural Networks (ANNs)

959 ANNs are a type of machine learning algorithm inspired by the biological neuron structure.  
960 ANNs are generally composed of interconnected nodes where the signal is processed through  
961 operations with tunable parameters named weights and biases for multiplications and addition  
962 respectively. An important feature of each node is the *activation function*, that introduces a non-  
963 linear operation and returns the node's output [109]. Several kinds of these activation functions  
964 exist and their use depends on the properties of each (bounds, derivatives, positivity, etc.) [110].  
965 In the following chapters the modified rectifying linear unit, known as LeakyReLU [111], and  
966 the sigmoid, also known as logistic, function will be used. Neurons are generally organized  
967 into *layers* and are connected to neurons of other layers. In *feed-forward* neural networks the  
968 information flows from the input layer to the output layer with forward connections only. In  
969 other neural networks, like *recurrent* ones, the connections are also designed backwards.



**Figure 4.1: Schematic of an artificial neuron and a neural network.** On the left, a series of input signals  $x_i$  are multiplied by the tunable weight  $w_k$  and summed together with the tunable bias  $b_k$  relative to the  $k$ -th neuron. The output is then passed through the activation function  $f$  which produce an output  $y_k$  which is a non-linear combination of the inputs. On the right, the feed-forward network composed of two *hidden* layers of artificial neurons processing the ten units long vector  $x$  and returning a binary output. Networks of this type are said to be *fully connected* as each input component, or node's output is processed by all neurons. Adapted from [112]

#### 970 4.1.1 Neural networks as universal approximators

971 The use of non-linear functions is found to be fundamental for the powerful analytical and  
 972 statistical properties of ANNs, which have been progressively established over the years. It was  
 973 shown [113–115] that ANNs with appropriate activation functions and sufficient parameters  
 974 can approximate *any* continuous function on a compact domain to arbitrary accuracy (*universal*  
 975 *approximation theorems*). Later the proof has been extended to deep convolutional neural  
 976 networks as well [116].

977 Formally, we can consider  $\mathcal{X}$  as the set of all events belonging to the same statistical  
 978 distribution and  $\mathcal{Y}$  similarly. We also assume that an unknown mapping  $\mathcal{M}$  between an input  
 979  $x \in \mathcal{X}$  and the output  $y = \mathcal{M}(x)$  exists. At this point, the goal of the ANNs is to be the  
 980 closest approximation of  $\mathcal{M}$ . Typically, this implies seeking the combination of parameters  
 981  $\theta_k = (w_k, b_k)$  of the neural network  $\mathcal{N}_\theta$  such that  $\mathcal{N}_\theta \approx \mathcal{M}$

982 **How is this mapping found?** The core idea is known as Empirical Risk Minimization  
 983 and states that when the  $\theta_k$  are adjusted to fit a sufficiently large dataset consisting of samples  
 984 drawn from an underlying distribution, the function being approximated reflects the statistical  
 985 relationships encoded in that distribution [117]. By the Law of Large Numbers, the empirical  
 986 distribution observed in the *training set* converges to the true distribution as the sample size  
 987 grows, and thus the empirical risk minimized during training approaches the true risk. This  
 988 statistical foundation explains why ANNs are able to generalize to new, unseen data drawn

989 from the same population.

990 It is therefore sufficient to have a large but finite number of samples representative of both  
 991 the sets  $\mathcal{X}$  and  $\mathcal{Y}$  to approximate  $\mathcal{M}$ . This powerful statistical property underlies the concept  
 992 of *supervised* training of ANNs.

993 More formally, one can consider having a limited set of  $N$  examples  $(x_1, y_1 = \mathcal{M}(x_1)), \dots, (x_N, y_N =$   
 994  $\mathcal{M}(x_N))$  which we call *training set*  $\mathcal{T}$ . Here, each  $x_i$  is an input instance and  $y_i$  the correspond-  
 995 ing ground truth transformation operated by  $\mathcal{M}$ . We can now introduce a non-negative scalar  
 996 real-valued *loss function*  $L(\hat{y}, y)$  which measures the difference between the ground truth  $y$   
 997 and the output of the neural network  $\hat{y} = \mathcal{N}_\theta(x)$  for each element of the training set.

998 It follows that the best approximation of the mapping  $\mathcal{N}_\theta^{\mathcal{T}} \approx \mathcal{M}^{\mathcal{T}}$  is obtained when the  
 999 score of the loss function averaged across the number of training samples is lowest. The  
 1000 problem of approximating the mapping between the input and ground truth in the training set  
 1001 is then formulated as a minimization problem of the type:

$$\hat{\theta}_{\mathcal{T}} = \arg \min_{\theta} \left\{ \frac{1}{N} \sum_{i=1}^N L(y_i, \mathcal{N}_{\theta}^{\mathcal{T}}(x_i)) \right\} \quad (4.1)$$

1002 For a sufficiently large  $N$  the  $\hat{\theta}_{\mathcal{T}} \approx \hat{\theta}$  valid for the whole statistical distribution. The size of  
 1003  $N$  needed to approach the true mapping depends obviously on the complexity of the mapping  
 1004 and on the complexity of the minimization task. While the first is inherent to the problem, the  
 1005 second can be engineered with the choice of a metric for which the loss function minimization  
 1006 is easier. This will be clear in Chapter 6 where different loss functions are tested. In the  
 1007 same way, the network design can impact the facility with which the ANN converges to the  
 1008 approximation of the desired mapping. In this regard, the use of multiple intermediate layers  
 1009 of neurons (*hidden layers*) was shown to strongly improve the ability of the NN to fit more  
 1010 complex functions (see [108] - Chapter 4.5). For this reason these network called *deep* have  
 1011 taken over *shallow* ones. Another example, briefly discussed in the next section, is given by  
 1012 Convolutional Neural Networks (CNNs), a type of ANN suited for natural image processing.

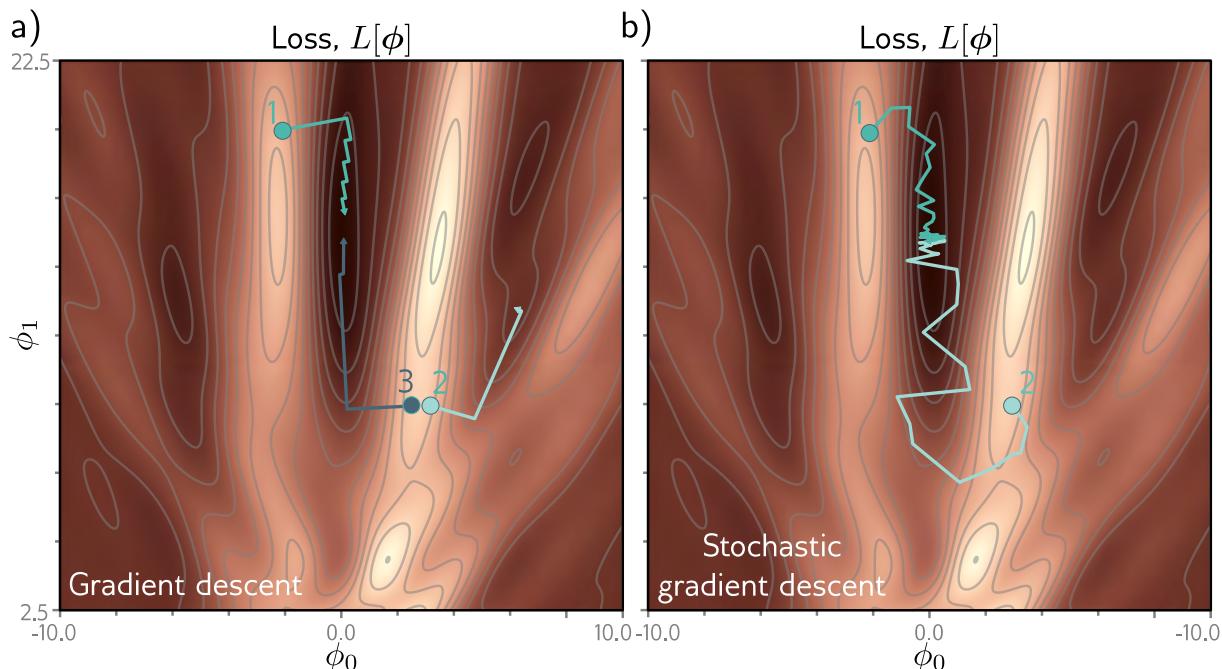
1013 Moreover, in cases in which one does not have a training set composed of input - ground  
 1014 truth pairs, but instead incorporates prior knowledge into the model architecture or the loss  
 1015 function, the training is said to be *unsupervised*. This approach has not been explored in the  
 1016 context of this PhD.

### 1017 4.1.2 Gradient Descent

1018 At this point another question may arise: **How do we solve the minimization problem?** The  
 1019 most straightforward manner to solve a minimization problem is with gradient descent. This  
 1020 implies an iterative process in which at each iteration the derivatives of the loss function with  
 1021 respect to each parameter  $\theta$  are calculated, and each parameter is updated correspondingly.

$$\theta^{t+1} = \theta^t - \eta \nabla_{\theta} \left[ \frac{1}{N} \sum_{i=1}^N L(y_i, \mathcal{N}_{\theta}(x_i)) \right] \quad (4.2)$$

1022 Where  $\eta$  is the *learning rate* that is given as an external parameter (*hyperparameter*) to the  
 1023 model. Eq.4.2 implements the steepest gradient descent. However, in most machine learning  
 1024 optimizers a variant of this algorithm is computed. Namely, the gradients are calculated for  
 1025 each sub-set, often called *mini-batch* of the whole training set. During the training, within each  
 1026 *epoch* as many mini-batches as are needed to make the full dataset, are minimized in series.  
 1027 This approach, called *stochastic* gradient descent (SGD), is less expensive in terms of memory  
 1028 and offers well established convergence properties that outperform classical steepest descent  
 1029 methods [118]. In fact, the “noise” affecting the updates induced by the minimization of a small  
 1030 sub-set can be beneficial for escaping saddle points which may trap the search. Over the years,  
 1031 different variants of SGD have been proposed. The “momentum” calculation [119], that keeps  
 1032 track of the magnitude and direction of the updates and determines the next update as a linear  
 1033 combination of the gradient and the previous update, was first applied to SGD [120]. Later,  
 1034 adaptive approaches have aimed at tuning the learning rate differently for each parameter  
 1035 (AdaGrad [121], ADAM [122]). Later in the text, the DL models employed have adopted the  
 1036 ADAM optimizer. It is worth mentioning that, though the most widely used, gradient descent  
 1037 approaches are not the only strategies that have been explored to minimize the loss function.  
 1038 Evolutionary algorithms inspired by natural selection mechanisms and tensor optimization  
 1039 techniques developed in the quantum-many body field have been employed as well [123, 124].



**Figure 4.2: Gradient descent and Stochastic gradient descent.** **a)** Depicts the trajectory of the error score in a 2D loss function in case of gradient descent with line search from three different initializations. The algorithm converges to the global minimum when it starts from points 1 and 3 while it fails when it starts from point 2, located outside the valley of the global minimum. **b)** The same experiment solved with stochastic gradient descent achieves good convergence also when initialized from point 2. Indeed, the “noise” introduced by the mini-batches drives the updates outside the wrong valley and allow the convergence to the global minimum. Adapted from [108]

### 1040 4.1.3 Backpropagation and automatic differentiation

1041 At this stage, a crucial question arises: **how can SGD be efficiently implemented in practice?**  
 1042 ANNs often involve millions or even billions of parameters  $\theta$ , thus computing exact derivatives  
 1043 with respect to such a large number of variables, for every batch and across multiple epochs,  
 1044 requires highly optimized algorithms and hardware acceleration. Indeed, the practical feasibility  
 1045 of training neural networks was significantly advanced by the seminal work of Rumelhart,  
 1046 Hinton, and Williams in 1986 [120], which introduced the efficient *back-propagation* technique  
 1047 that made large-scale training computationally tractable.

1048 Back-propagation bases its principles on the fundamental *chain rule* proposed by Leibniz  
 1049 in 1676 to calculate derivatives of function compositions. In fact, a ANNs can be seen as a  
 1050 composition of functions (the layers) in which at each stage the layer  $i$ -th processes the output  
 1051 of the layer  $(i - 1)$ -th. Formally, one can write:

$$\mathcal{N}_\theta = f_\theta^{(L)} \circ f_\theta^{(L-1)} \circ \dots \circ f_\theta^0 \quad (4.3)$$

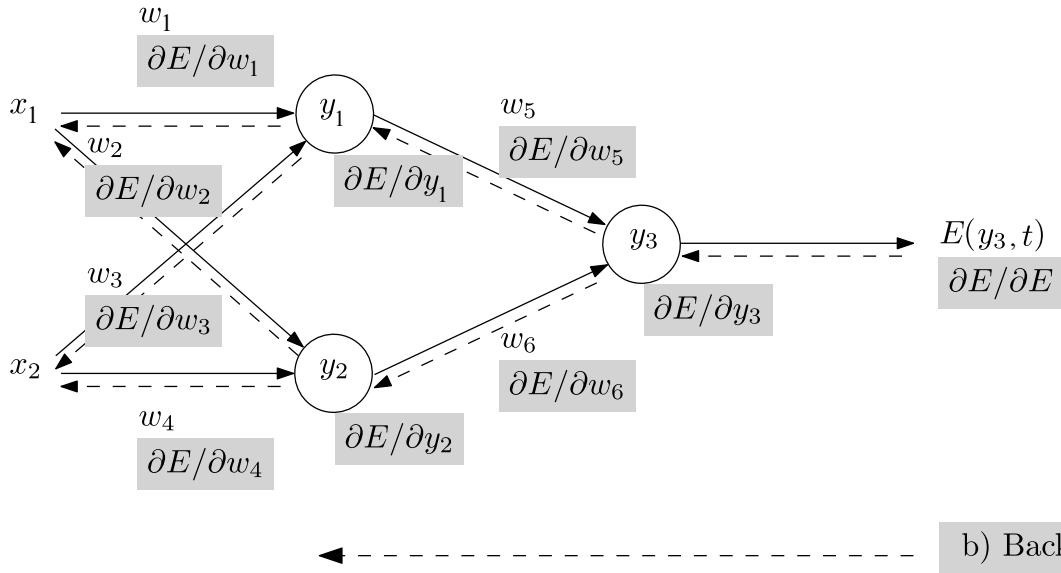
1052 Where  $f_\theta^{(L)}$  is the function representing the  $L$ -th layer of neurons of parameters  $\theta$ . The  
 1053 gradient of the loss function with respect to the parameters is therefore expressed, according  
 1054 to the chain rule as:

$$\nabla_{\theta^{(l)}} L = \frac{\partial(L)}{\partial f^{(L)}} \cdot \frac{\partial f^{(L)}}{\partial f^{(L-1)}} \cdot \dots \cdot \frac{\partial f^{(l)}}{\partial \theta^{(l)}} \quad (4.4)$$

1055 This method decomposes the calculation of the full gradient into a sequence of smaller,  
 1056 local derivatives, each associated with the intermediate states of the network. During the  
 1057 forward pass, the activations and local Jacobians are computed and stored in memory, a process  
 1058 sometimes referred to as *forward accumulation*. These stored quantities are then systematically  
 1059 reused during the backward pass to propagate gradients from the output layer back through  
 1060 the network parameters. This reuse of intermediate computations is what makes the algorithm  
 1061 highly efficient, and it is from this backward flow of gradients that the name *back-propagation*  
 1062 originates.

1063 In modern machine learning frameworks such as PyTorch [125] and TensorFlow [126],  
 1064 back-propagation is implemented through the construction of a *computational graph*. Each  
 1065 node in the graph represents an operation, while edges encode the dependencies between  
 1066 operations. Once the forward pass has been executed, this graph allows the backward pass  
 1067 to be automatically derived and executed, efficiently parallelized across GPUs or TPUs. In  
 1068 fact, back-propagation is a specific instance of a more general family of techniques known as  
 1069 *automatic differentiation* (or *algorithmic differentiation*) (AD) [127, 128], and more precisely  
 1070 corresponds to *reverse-mode automatic differentiation*. This connection highlights that back-  
 1071 propagation is not unique to neural networks, but rather belongs to the rigorous and general  
 1072 framework for computing exact derivatives of functions defined by computer programs.

a) Forward pass



b) Backward pass

**Figure 4.3: Schematic of Back-propagation on computational graph.** a) The training inputs  $x_i$  are processed in the forward pass by the network that produces intermediate states  $y_{1,2}$  using weights  $w_i$ , and the final output  $y_3$ . b) The error  $E$  computed between the output and the estimate  $t$  is propagated backward and the gradients with respect to the weights  $\nabla_{w_i} E = (\frac{\partial E}{\partial w_1}, \dots, \frac{\partial E}{\partial w_6})$  are obtained from the chain rule of derivatives. Adapted from [128]

## 1073 4.2 Convolutional Neural Networks (CNNs)

1074 It was anticipated earlier in the Chapter that the network design can strongly impact the  
 1075 learning of the mapping  $\mathcal{M}$ . In the specific case in which  $\mathcal{X}, \mathcal{Y}$  represent sets of natural images,  
 1076 neural networks with layers employing convolution operations have shown to outperform  
 1077 fully connected ones [129–131]. The reasons behind the success of convolutions are intuitively  
 1078 explained as follows:

- 1079 • Images are inherently high-dimensional, as they are defined on 2D or 3D grids. As a result,  
 1080 the number of parameters required to connect each pixel in a fully connected neural  
 1081 network grows rapidly and soon becomes intractable. Convolutional layers address this  
 1082 issue by employing a *kernel*: a small trainable filter that is convolved with the input  
 1083 image or intermediate feature maps. This mechanism enables weight reuse across spatial  
 1084 locations, drastically reducing the number of trainable parameters, a property commonly  
 1085 referred to as *parameter sharing*. Furthermore, the localized receptive field of the kernel  
 1086 induces a form of structured sparsity, effectively regularizing the model by constraining  
 1087 the mapping to be approximated using compact, spatially coherent information.
- 1088 • In images, spatially adjacent pixels are typically correlated, while uncorrelated variations  
 1089 are often attributable to noise. Fully connected layers process each pixel independently, re-  
 1090 quiring the network to learn spatial dependencies solely through training. Convolutional  
 1091 layers, in contrast, explicitly exploit local spatial correlations by aggregating information  
 1092 from neighboring pixels, thereby embedding structural priors into the architecture. (see  
 1093 Fig. 4.4)
- 1094 • The image of a tree and another image of the same tree, translated require a different  
 1095 tuning of the weights in a fully-connected layer, although the interpretation of the  
 1096 image has not physically changed. Learning a different parameter configuration for  
 1097 each different translation of the same object is highly inefficient. On the contrary, an  
 1098 additional benefit of the parameter sharing in convolutional layers is the equivariance  
 1099 to translation that inherently optimize the learning. This property allows the output to  
 1100 respond to the input change in the same way. Note that this equivariance is not valid for  
 1101 other geometrical transformations like rotation and magnification.

1102 We have seen in Chapter 2 that BCDI data is in the form of 3D images in which peculiar  
 1103 spatial structures made of peaks and fringes are clearly visible, hence the natural choice of  
 1104 convolutional neural networks. Moreover, the tasks addressed in this PhD thesis, namely the  
 1105 gap-inpainting and the phase retrieval, are classified as inverse problems where the goal is to  
 1106 reconstruct missing or unmeasured information from incomplete observations. As highlighted  
 1107 in recent surveys and related works [132, 133], CNNs are particularly well suited for these  
 1108 problems because they can learn powerful image priors from data, act as regularizers even  
 1109 without extensive training (as shown in the Deep Image Prior framework [134]), and can be  
 1110 combined with physics-based models to enforce data consistency.

1111 Let us present now the building blocks of a typical convolutional neural network.

**4.2.1 Convolutional layer**

It needs to be clarified here that to improve computational efficiency, machine learning libraries typically implement the *cross-correlation* operation rather than a convolution. The difference is that the convolution flips the kernel before computing the sum, whereas cross-correlation does not. This simplifies the implementation with no expressive power loss, since the weights are learned during training but in turn breaks the commutative property of the convolution. Given an input vector  $\mathbf{x}$  with  $N$  entries the output of a convolutional layer  $\mathbf{y}$  with  $M$  entries can therefore be written as a matrix - vector multiplication of the type:

$$\mathbf{y} = \mathcal{W}\mathbf{x} \quad (4.5)$$

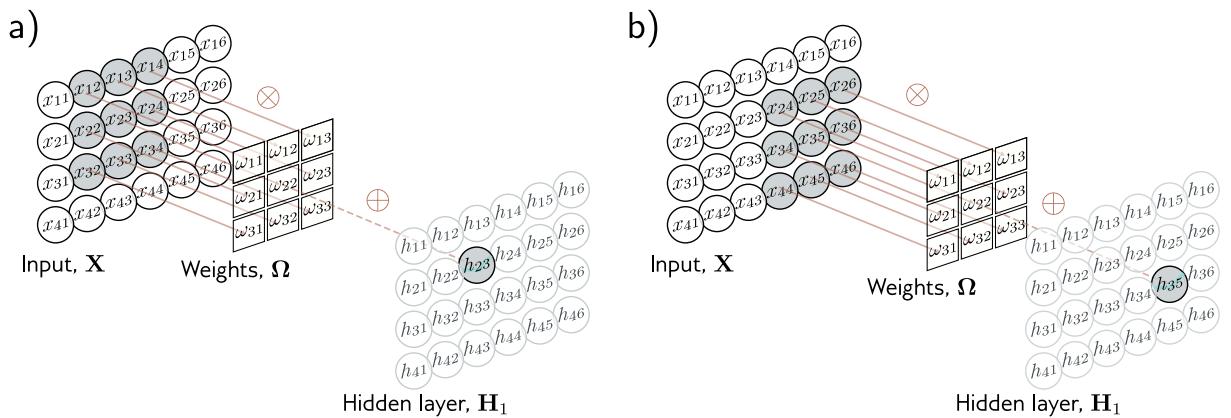
Where  $\mathcal{W} \in \mathbb{R}^{M \times N}$  is a Toeplitz matrix, where each descending diagonal contains a kernel entry, arranged such that the matrix performs the cross-correlation as a linear operation.

The initialization of a convolutional layer in typical machine learning libraries involve the definition of:

- An integer number of kernels. This number corresponds to number of filters through which the input is processed in a convolution operation. Each filter is normally associated to a different feature of the input image that is extracted by the corresponding kernel. The output of the convolutional layer will have an extra dimension (called *channel*) with size corresponding to the number of filters.
- The integer-valued size of the kernel. In 2D convolutions the kernel is a matrix and similarly extended to a 3D tensor for three-dimensional inputs. It controls the scale of the features that the convolutional layer can “see”. Small kernels highlight small features and vice-versa.
- The padding parameter which handles the convolution at the borders of the image, where the kernel would otherwise extend beyond the available data. It determines whether the output feature map maintains the same spatial dimensions as the input (“same” padding) or is reduced in size (“valid” padding).
- The stride parameter: This integer-valued parameter (matrix in 2D and tensor in 3D) indicates the lateral shift that is applied to the kernel across the image. If greater than 1 some pixels/voxels are skipped resulting in a sort of sub-sampling allowing to filter out some low-level features.
- The dilation rate, which is another integer-valued parameter (matrix in 2D and tensor in 3D) that allows to insert holes between consecutive elements of the kernel. In this way the actual size of the kernel is increased by the dilation value, but the overall number of weights is unvaried. This enables to cover a larger area of the input without increasing the number of trainable parameters.

Additional features of a typical convolutional layer include the addition of trainable biases and the initialization and regularization of the kernels and biases. Lastly, a non-linear activation is usually applied to the output of the convolutional layer.

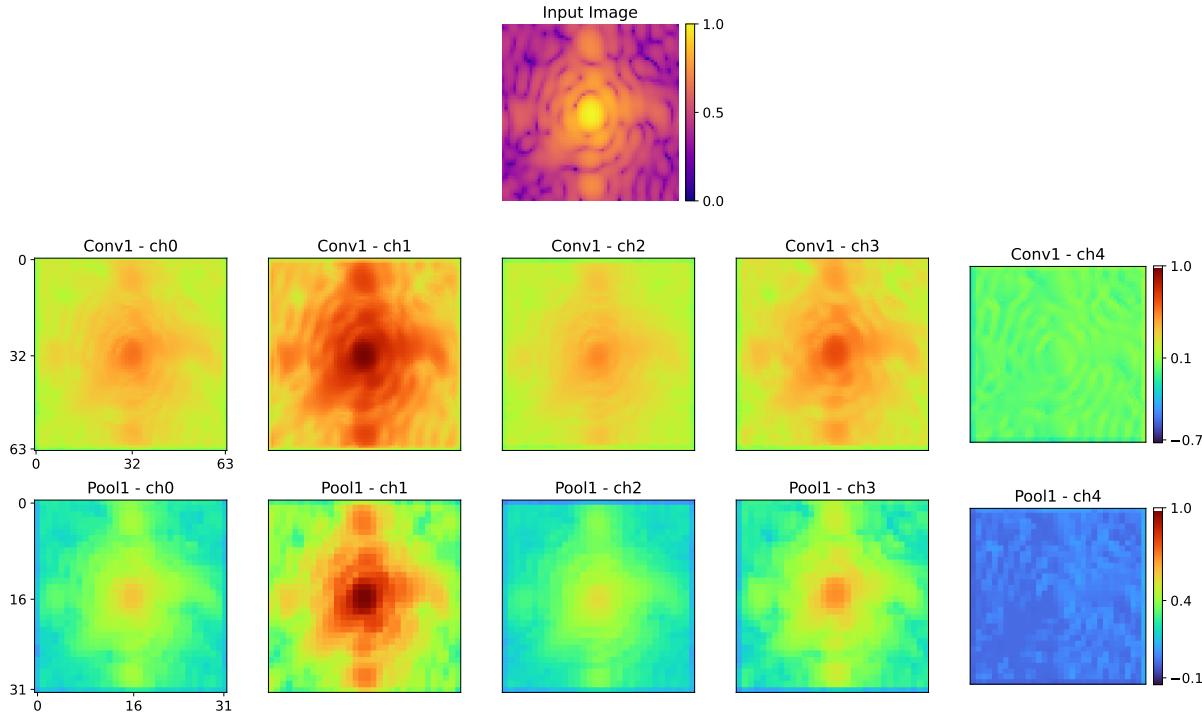
1149 We mention here the existence of so-called *transposed convolutions* which are sometimes  
 1150 used in CNNs. Here the conceptual difference is that Eq.4.5 is computed with the transposed  
 1151 Toeplitz matrix  $\mathcal{W}^T$ . Transposed convolutions naturally produce an output that is larger than  
 1152 the input, because they reverse the spatial reduction of a standard convolution. For this reason,  
 1153 they are often used to replace explicit *up-sampling* layers, which increase the spatial size  
 1154 through fixed interpolation (e.g. nearest- neighbor or bilinear). Unlike standard interpolation,  
 1155 transposed convolutions perform a *learned* up-sampling, where the network optimizes the  
 1156 kernel weights to reconstruct high-resolution features in a data-driven way.



**Figure 4.4: Schematic of 2D convolutional layer:** **a)** The convolution is operated over a region of the image with same size as the kernel ( $3 \times 3$ ). The result of the multiplication of each pixel value ( $x_{12}, \dots, x_{34}$ ) times the weight ( $w_{11}, \dots, w_{33}$ ) is then summed into a scalar  $h_{23}$ . **b)** The same operation with the same kernel is performed to a shifted window of the input image. Adapted from [108]

## 1157 4.2.2 Pooling Layer

1158 After the convolutional layer and the activation function, it is often the turn of a *pooling*  
 1159 layer. For the 2D case, the output of the layer at specific location  $i, j$  is the result of a filtering  
 1160 operation on the neighboring pixels of the input. For instance, the “Max Pooling” layer replaces  
 1161 a rectangular region of the input with its maximum value, thus retaining only the strongest  
 1162 activation within that area. In contrast, the “Average Pooling” layer assigns to the output pixel  
 1163 the mean value of all pixels in the corresponding region. The benefits deriving from the pooling  
 1164 layer are mostly twofold. First, the size of the output is reduced by a factor proportional to the  
 1165 area of the pooling window (or the volume in the 3D case), thus lowering the computational  
 1166 and memory cost of subsequent operations. Second, by condensing information into a smaller  
 1167 region, pooling encourages the network to learn representations that are more robust to small  
 1168 translations. For instance, with Max Pooling, a shift of the most activated pixel within the  
 1169 pooling window does not affect the output. This behavior can be interpreted as an implicit  
 1170 prior that biases the optimization towards translation-invariant approximations.



**Figure 4.5: Example of Convolutional and Max pooling layers.** A simulated 2D BCDI pattern is input image (first row) of the CNN model presented later in Chapter 6, after the training. The output of the first convolutional layer + LeakyReLU activation is displayed in the second row (first 5 channels) while the output of the subsequent Max Pooling layer is displayed in the third row. Notice the similarity between the two rows despite the halved size of Max Pooling output. This shows that it efficiently condenses the information into smaller sizes.

1171     The typical convolutional block is therefore composed of these three layers (convolutional,  
1172     activation, pooling). A sequential application of the convolutional blocks to a 2D or 3D input  
1173     image is often called *encoder* and can reduce the dimension of the feature map to as many 1D  
1174     vectors as the number of filters of the last convolutional layer. During the training, this reduced  
1175     representation of the input is more and more driven to capture the essential information in  
1176     a lower-dimensional space. At this point, for *classification* tasks the feature map is flattened  
1177     into a single 1D vector being the input of a fully connected layer. The output of this last layer  
1178     is then usually interpreted as the probability score for the input image to belong to a specific  
1179     class. Among the milestone CNN models for classification, it is worth highlighting LeNet-5,  
1180     introduced by LeCun in 1998 [135], and AlexNet, which revolutionized the field by leveraging  
1181     more efficient GPU-based training [136].

1182     In *image generation* tasks, the lower-dimensional space, also called *latent space* serves as  
1183     input to a sequence of *deconvolutional* blocks, where *transposed* convolutions combined with  
1184     activation functions progressively reconstruct the output image at the desired resolution. This  
1185     sequence of deconvolutional layers is referred to as the *decoder*, as it mirrors the encoder.  
1186     In some architectures, up-sampling layers followed by standard convolutions are employed  
1187     instead of transposed convolutions. Encoder-decoder architectures for image generation were  
1188     first introduced by Hinton in 2006 [137]. This class of models is employed in a variety of tasks  
1189     such as de-noising, image compression, inpainting, and segmentation. Although other types  
1190     of models are also used for image generation, including Generative Adversarial Networks

1191 (GANs) [138] and Vision Transformers (ViTs) [139], in this manuscript we restrict our focus to  
1192 encoder-decoder architectures.

### 1193 4.2.3 Skip connections

1194 Before concluding the chapter it is worth mentioning the concept of *residual connections*, or  
1195 more in general *skip connections* which has been introduced in deep convolutional neural  
1196 networks with the ResNet model [140]. In this type of architecture the output feature maps in  
1197 the encoder blocks is summed to the input of the decoder blocks with corresponding size. In  
1198 other cases, like the U-Net architecture [141] a concatenation of the two feature maps along  
1199 the channel dimension is performed instead of the sum. The goal of the skip connections is to  
1200 facilitate the training of deep generative networks by “reminding” the model about the features  
1201 extracted in the beginning of the forward process. In fact, the more difficult training of deep  
1202 networks as compared to shallow ones is attributable to the more unstable gradients [142].  
1203 Tiny changes in the input result in a completely different gradient. A study conducted by Li *et*  
1204 *al.* has shown the benefits of skip connections through a study of the loss function landscape,  
1205 which tends to have less saddle points and local minima [143].

1206

1207 This paragraph concludes the introductory background on neural networks. More broadly,  
1208 the chapter complements the presentation of the physics and conventional data analysis of the  
1209 BCDI technique from the first two chapters, by introducing the concepts, terminology, and  
1210 insights needed to approach the content of the following three chapters.



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1212 DEEP LEARNING FOR DETECTOR GAPS INPAINT-

1213 ING

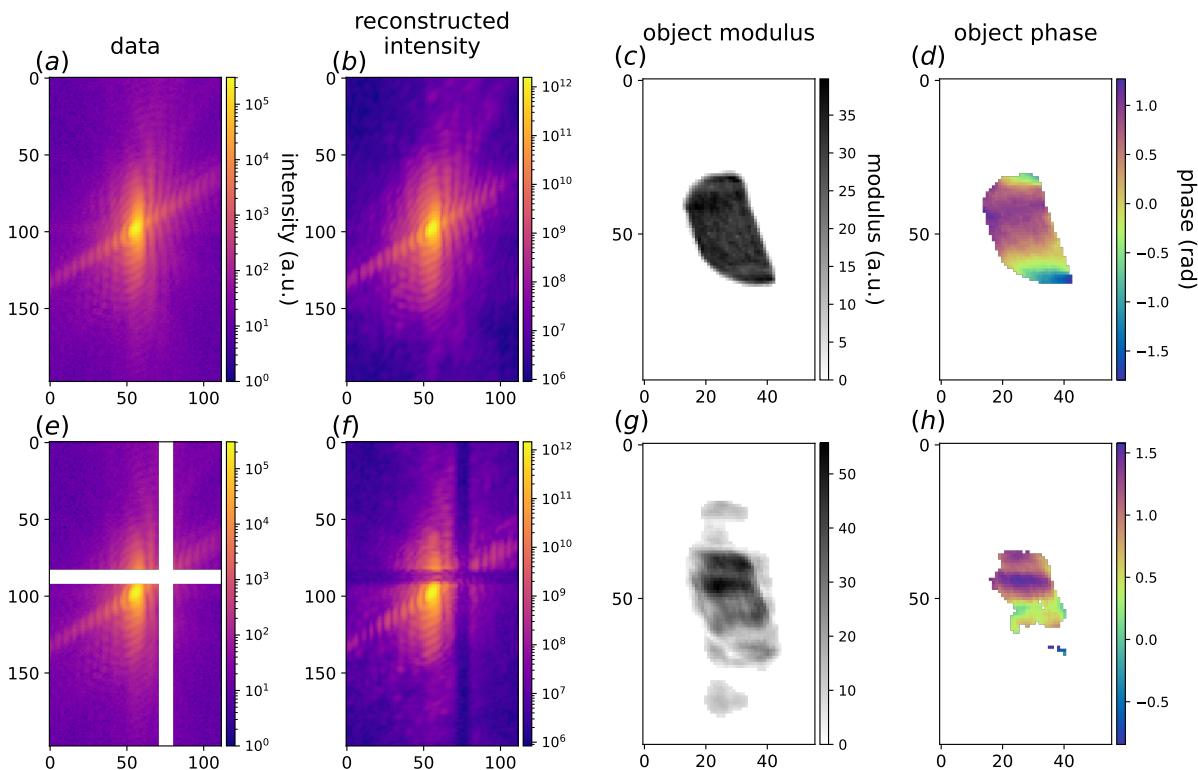
1214 In this chapter, the “detector - gaps problem” in Bragg Coherent Diffraction Imaging and  
 1215 the approach to solve it using Deep Learning (DL) are discussed. The main state-of-the-  
 1216 art methods are presented briefly and the topic of image inpainting with DL is introduced.  
 1217 The focus will then shift to the works that led eventually to the optimal “Patching-based”  
 1218 approach that can also be found in the published paper entitled “*Patching-based deep learning*  
 1219 *model for the Inpainting of Bragg Coherent Diffraction patterns affected by detectors’ gaps*”  
 1220 (<https://doi.org/10.1107/S1600576724004163>). The chapter is closed with the analysis of the  
 1221 performance of the DL models in a variety of simulated and experimental cases.

1222 **5.1 The “Gap Problem”**

1223 At time of writing, standard BCDI experiments employ pixelated photon counting detectors to  
 1224 record the diffraction patterns. These detectors can guarantee high spatial resolution, low-noise  
 1225 counting and fast read-out times. Two examples of these devices, currently used at the ID01  
 1226 beamline are the MAXIPIX and EIGER detectors [50, 144]. These detectors are often built by  
 1227 tiling together several sensing chips in order to cover a larger area, and are typically bonded to  
 1228 an Application-Specific Integrated Circuit (ASIC) using bump bonding. This implies always  
 1229 some physical connection which is not a pixel on one side of a chip. Additionally, intra-chip  
 1230 edges also constrain larger pixels which disrupt the oversampling condition, hence cannot  
 1231 always be used. These issues result in the presence, in the overall sensing region, of vertical  
 1232 and/or horizontal stripes that are not filled with useful signal. The width of these lines varies  
 1233 depending on the device but normally does not exceed the equivalent of some tens of pixels.  
 1234 Specifically, the MAXIPIX detector, with sensing area of  $516 \times 516$  pixels of  $55\mu m \times 55\mu m$ , is  
 1235 composed of four modules separated by  $220\mu m$  wide gaps (equivalent of 4 pixels) while the  
 1236 edge pixels measure  $165\mu m$  (i.e. 3 pixels), thus making a gap, when adjacent, of 6 pixels width.

1237 The EIGER detector instead has two types of larger gaps of 12 pixels and 38 pixels width.  
 1238 The detector gaps problem does not affect BCDI only, but it is shared among other X-ray  
 1239 techniques that deal with single photon-counting pixelated detectors and/or beamstops. We  
 1240 have seen in chapter 2 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 [145] (see Fig.5.1).



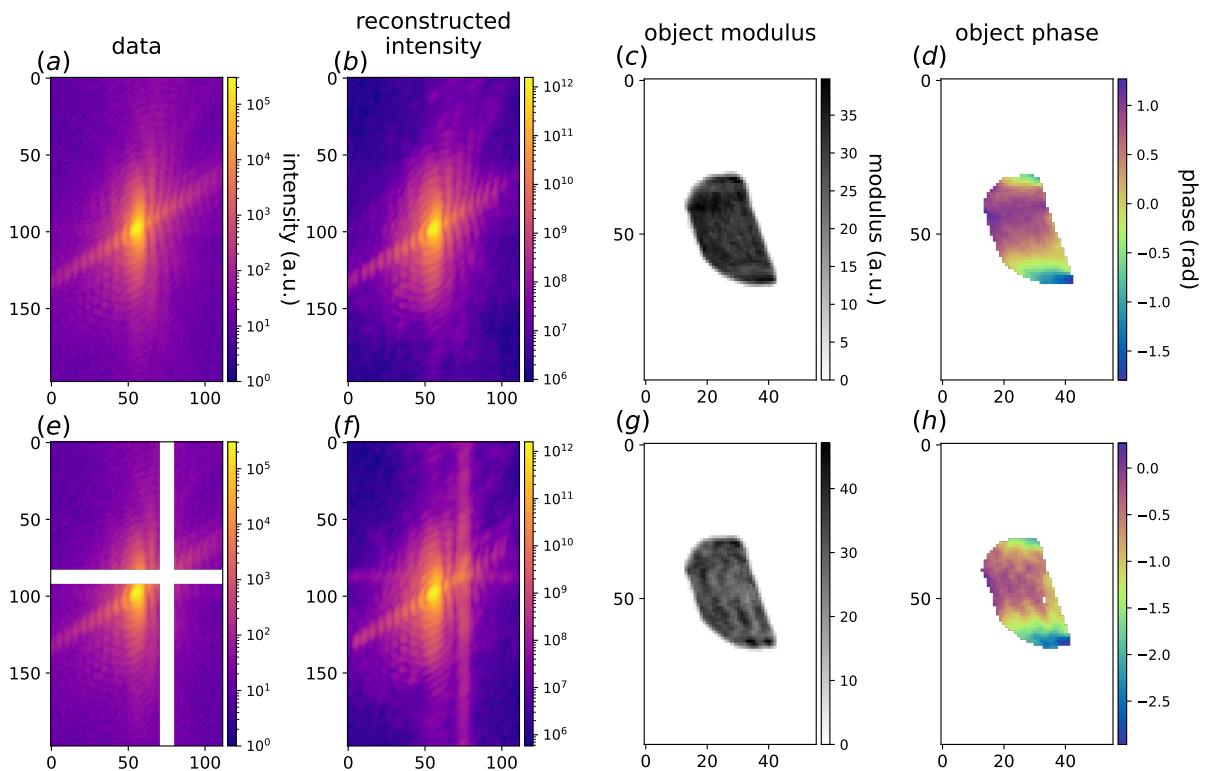
**Figure 5.1: Effect of detector gaps in BCDI reconstructions** (a) The central slice of an experimental diffraction pattern. (b) The same slice of the diffracted intensity calculated from the retrieved object. (c - d) slice of the modulus and phase respectively of the particle obtained from the phasing 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 of 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.

## 1252 5.2 State of the art

1253 Here we will discuss the current strategies employed to treat the detector gaps. As someone  
 1254 could argue the simplest, yet not practical, solution would be to slightly move the detector  
 1255 sideways and acquire a second full scan with the gap hiding a different region of the same Bragg  
 1256 peak, and then merge the two measurements into a single gap-less one. This would at least  
 1257 double the acquisition time making it, de facto, not an option during standard experiments.

1258 The PyNX software, routinely used for the BCDI phase retrieval at ID01, allows the user to  
 1259 define a mask of the gap regions and ignore those pixels during the execution. This improves  
 1260 the quality of the reconstruction, but one can still notice the presence of oscillations appearing  
 1261 in both the object's modulus and phase. The origin of these artifacts can be found in the  
 1262 diffracted intensity calculated from the reconstructed particle as one can clearly see that the  
 1263 gap-regions is filled with non-physical high intensity (see Fig. 5.2(f))



**Figure 5.2: 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 5.1(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.

1264 Methods relying on compressive sensing and TV regularization were proposed by He in  
 1265 [146] and Malm [147] respectively. Another, more invasive, option is to *fill* these gaps with an  
 1266 estimate of the intensity distribution that would be there, before the phase retrieval. The task  
 1267 of filling gap in images is usually referred to as “inpainting”.

---

**1268 5.2.1 Background on Image Inpainting Research**

1269 Computational image inpainting has been widely studied in the field of photography and  
1270 imaging for many years [148, 149]. The inpainting problem can be defined as the task of using  
1271 known information extractable from the image, to repair the parts where this information is  
1272 missing, where for known information the colors, the textures and the semantic features are  
1273 intended. In the history of image inpainting a clear-cut can be observed when deep learning  
1274 methods have started to be employed. For traditional inpainting, different techniques have  
1275 been explored, from the texture synthesis methods pioneered by Efros and Leung [150] to the  
1276 use of PDEs as Navier-Stokes equations proposed by Bertalmio *et al.* [151] and then again from  
1277 sparse representations [152] to hybrid methods combining variational and statistical methods  
1278 [153].

1279 More recently, Deep Learning models, headed by Convolutional Neural Networks (CNN),  
1280 have taken the place of more traditional methods as they can attain higher accuracy for more  
1281 complex inpainting tasks. By undergoing a training process, CNNs can “learn” to recognize  
1282 and reproduce the semantic features of the training dataset, and thus leverage them during  
1283 inference as additional information beside the colors and textures of the specific image to  
1284 restore. As we have seen in Chapter 4, the typical CNN architecture for image generation  
1285 consists of an encoder, which retains the features of the input image and compresses them into  
1286 a lower dimensional latent space, and a decoder, which is responsible for the generation of  
1287 the output image starting from the latent space. The model is then trained according to a loss  
1288 function that pushes the model’s predictions to be close to a given ground truth reference. In  
1289 some cases, the loss function can be replaced by another CNN that is trained to discriminate  
1290 true images from the ones predicted by the model. These complementary networks are known  
1291 as Generative Adversarial Networks (GAN), firstly proposed by Goodfellow *et al.* [138], and  
1292 have also been used for image inpainting (e.g. [154]).

1293 Since reviewing the vast amount of works about CNN for image inpainting is beyond the  
1294 scope of this thesis and for more information, we redirect the reader to the reviews written by  
1295 Elharrouss *et al.*, Xianget al. and Xu *et al.* [149, 155, 156]. For what concerns the application  
1296 of DL based inpainting for scientific imaging, early works date back to 2018 as in the case of  
1297 Sogancioglu *et al.* for X-ray human chest 2D radiographic images [157] and to 2020 for 2D  
1298 microscopic images [158]. A couple of years later Tanny Chavez and coauthors published a  
1299 paper comparing the performance of different CNN models for the inpainting of 2D X-ray  
1300 diffraction images [159]. The work is precisely addressing the gap problem for X-ray detectors  
1301 used for powder diffraction measurements and is awarding U-Net and Mixed Scale Dense  
1302 (MSD) models for the best performance on experimental data. The DL models outperform  
1303 interpolations obtained with biharmonic functions across 7 and 17 pixel-wide gaps. This work  
1304 has been of inspiration for the design of our DL model for BCDI gaps inpainting. In the same  
1305 year, another work on DL based inpainting for X-ray detector gaps was published by Alfredo  
1306 Bellisario and coauthors [160]. The authors tested a U-Net-like model on the inpainting of  
1307 2D simulated, noiseless coherent diffraction patterns against gaps of different sizes (2 to 20  
1308 pixels) along the central row. The gaps were placed such that the center of the peak was cov-  
ered, a choice that, as we will see later, yields better results than predictions on peripheral areas.

1310

## 1311 5.3 Model design: 2D case

1312 For simplicity, I have begun with the 2D case, using simulated diffraction patterns and inpainting  
 1313 randomly placed vertical gaps of different width. First, a training set of simulated data was  
 1314 created, composed of pairs of gap-affected images and corresponding gap-free ground truths,  
 1315 then a U-Net-like model was built and trained in a supervised fashion.

### 1316 5.3.1 Dataset creation

1317 The creation of training datasets of simulated 2D BCDI patterns for both the gap-inpainting  
 1318 and phase retrieval tasks has followed the procedure described in this paragraph.  
 1319 First, once the size of the array was chosen, a randomly shaped polygon was created in the  
 1320 center using `scipy.spatial.ConvexHull` function. This guarantees the object to have  
 1321 a compact support with homogeneous electron density as assumed for BCDI. Subsequently,  
 1322 a random phase field of the same size with variable phase range and correlation length was  
 1323 generated thus the complete complex object was formed.

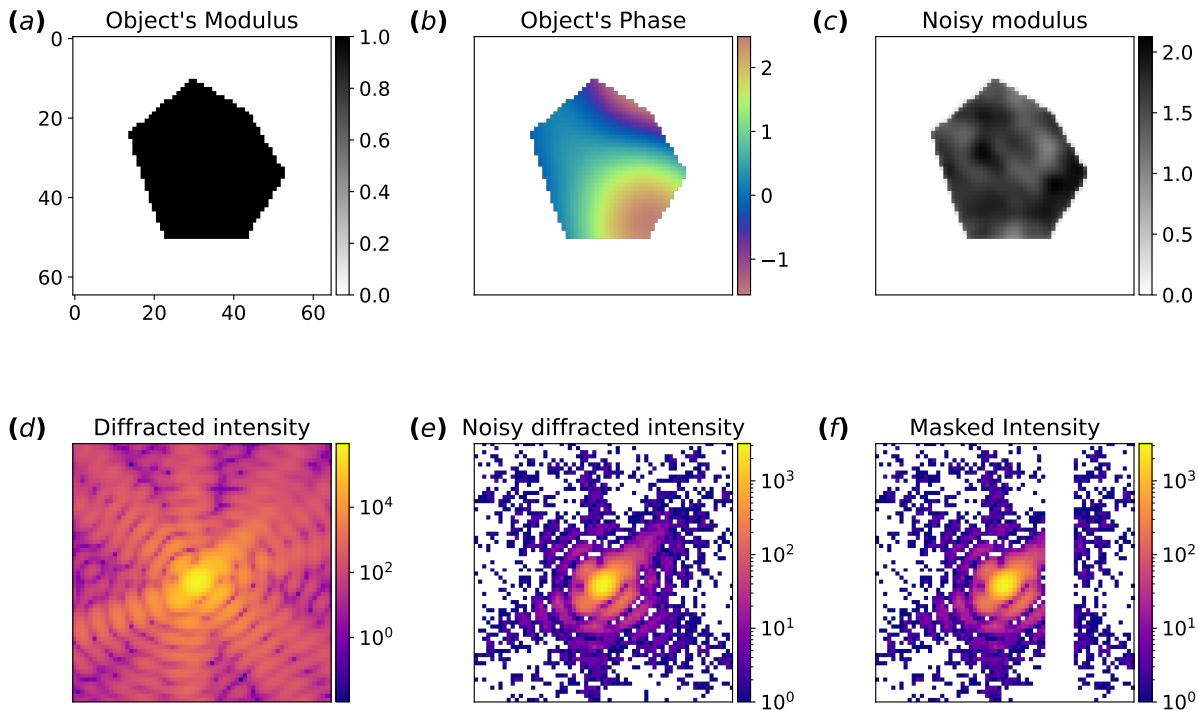
1324 In order to make the object more realistic a Gaussian filter and Gaussian - correlated noise  
 1325 [161] have been applied to the object's modulus, to smoothen the edges and simulate real cases  
 1326 respectively. At this point the object was resized to the shape required to match the chosen  
 1327 oversampling ratio and the 2D Discrete Fourier Transform was computed. Finally, Poisson  
 1328 noise was added to the diffraction patterns with different magnitudes to simulate various X-ray  
 1329 flux conditions (see Fig.5.3).

1330 Datasets contain a number of diffraction patterns in the order of thousands and for each of  
 1331 them the random variables have been varied as well as the oversampling ratios. In the datasets  
 1332 for the training of phase retrieval models (presented later in Chapter 6), the reciprocal space  
 1333 phase corresponding to each diffraction pattern was saved as well and used as ground truth  
 1334 label. For inpainting tasks a randomly located vertical gap mask was created and applied  
 1335 to the intensity data. In some cases cross-shaped gaps were added instead to simulate the  
 1336 experimental condition of the Bragg peak in the vicinity of the corner of the sensing area. The  
 1337 size of the gaps was chosen to be consistent across the dataset and four different cases were  
 1338 studied (3px, 6px, 9px, 12px). Fig.5.3.

### 1339 5.3.2 2D Model design

1340 The 2D model that I have implemented is a U-Net that takes in input batches of 32 simulated  
 1341 BCDI patterns affected by both vertical and cross-shaped gaps. Each diffraction pattern was  
 1342 transformed into logarithmic scale to enhance the spatial features and then normalized between  
 1343 0 and 1. This normalization was proven to be convenient to any DL model [162]. Regarding  
 1344 the logarithmic transformation, it is important to notice that in order to avoid problems for  
 1345 zero intensity values, the  $\log(I + 1)$  was taken. The shape of each image was chosen to be of  
 1346  $128 \times 128$  pixels.

1347 The encoder of part of the models was built as follows. The inputs go through five convolutional  
 1348 blocks inside each of which a convolutional layer, a Leaky ReLU activation function  
 1349 and a MaxPooling operation are applied. The feature map's size is therefore reduced down



**Figure 5.3: 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.

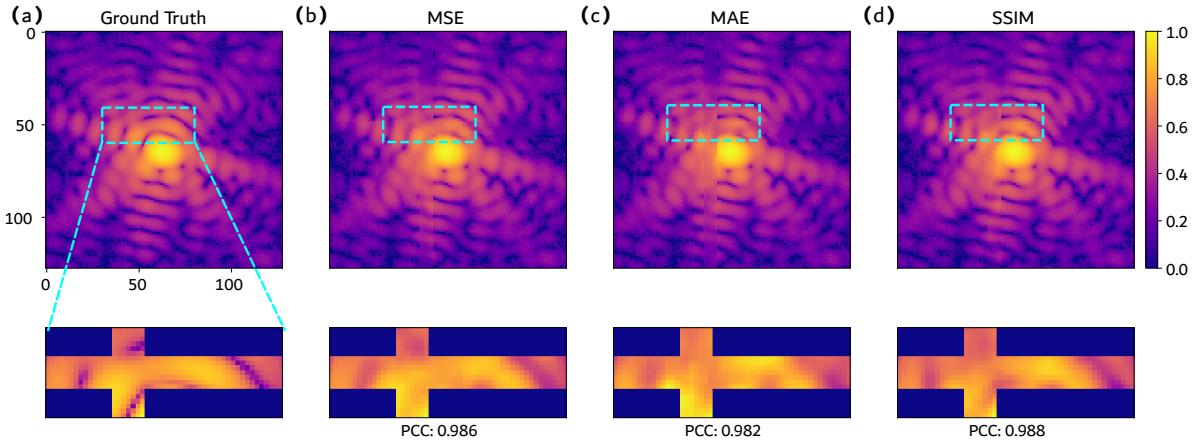
to  $2 \times 2$  while the channel dimension is brought up to 768 filters, and the kernel size is kept at  $3 \times 3$ . At the end of the encoder the feature map passed to the decoder is a  $(32, 2, 2, 768)$  tensor. The decoder, mirroring the encoder, is composed of five blocks inside each of which 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 [143]. The last activation layer of the model is a sigmoid function that guarantees an output bounded between 0 and 1.

1358

A simple Mean Squared Error (MSE) was used as loss 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  $10^{-4}$ . The Mean Absolute Error (MAE) and the Structural Similarity Index Measure (SSIM) [163] loss functions were tested as well and the corresponding results obtained after the same training were compared. Here in Fig. 5.4 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).

$$PCC = \frac{\sum_{i \in \text{gap}} (\mathbf{I}_i^{\text{true}} - \langle \mathbf{I}^{\text{true}} \rangle)(\mathbf{I}_i^{\text{pred}} - \langle \mathbf{I}^{\text{pred}} \rangle)}{\sqrt{\sum_{i \in \text{gap}} (\mathbf{I}_i^{\text{true}} - \langle \mathbf{I}^{\text{true}} \rangle)^2} \sqrt{\sum_{i \in \text{gap}} (\mathbf{I}_i^{\text{pred}} - \langle \mathbf{I}^{\text{pred}} \rangle)^2}}, \quad (5.1)$$

1366 Where  $\mathbf{I}$  is the intensity inside the gap. The PCC measures the linear correlation between  
 1367 two sets of data, and it is naturally normalized between -1 and 1 for which the two datasets are  
 1368 said to be linearly correlated. On the contrary the 0 value implies uncorrelated sets.



**Figure 5.4: 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 U-Net 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.

1369 In the light of these results, the MAE metric was discarded and the sum of MSE and SSIM  
 1370 adopted instead. Finally, another term that computes the MSE between the *gradients* of the  
 1371 ground truth and predicted intensity inside the gap region was added in the definitive loss  
 1372 function.

1373

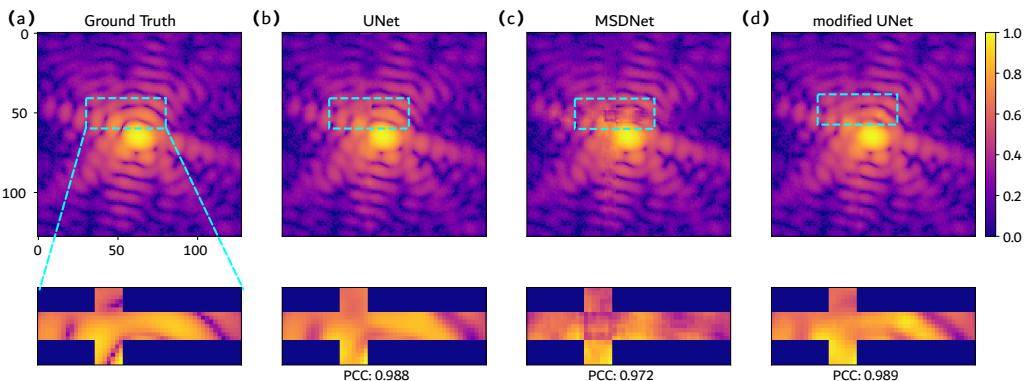
1374 Once established what was considered the best loss function, different models have been  
 1375 explored . Following the work of Chavez *et al.* mentioned above ([159]), a Mixed-Scale Dense  
 1376 Network (MSD-Net) was used [164]. The advantage of this type of network is the significant  
 1377 reduction of trainable parameters, and the use of *dilated* convolutions with respect to U-Net  
 1378 ones. While the former property guarantees faster training and lower chances of overfitting, the  
 1379 latter enhances the capture of long-range correlations. Moreover, in an MSD-Net, the image's  
 1380 spatial dimensions are kept constant throughout the whole network as no downsampling nor  
 1381 upsampling is made. The MSD-Net consists of sequential blocks in each of which the input is  
 1382 transformed by two different convolutional layers with growing dilation rates. Each output of  
 1383 the convolutional layers is concatenated to the input feature map and the result is passed to  
 1384 the following block. While the kernel size is kept constant to  $3 \times 3 \times 3$  pixels the dilation rate  
 1385 increases linearly from 1 to 30. The last layer is a sigmoid function as well as for the U-Net.  
 1386 The total number of trainable parameters is of the order of 320'000, two orders of magnitude  
 1387 lower than the U-Net.

1388 In order to combine the hierarchical dimensionality reduction of the U-Net with the fine-  
 1389 features capturing of the MSD-Net we have implemented a modified U-Net that adopts dilated  
 1390 convolutions inside the first three encoder blocks. In particular, they return the input tensor  
 1391 concatenated with the outputs of four dilated convolutional layers computed from the input.  
 1392 Dilation rates of (16,8,4,2), (10,5,3,1) and (5,3,2,1) were chosen respectively. As the MaxPooling

1393 operation down-samples the feature maps into smaller sizes, we limited the dilated convolutions  
 1394 to the first three blocks. Moreover, they were used in the encoder layers only as they are mostly  
 1395 used for feature extraction [165]. The characteristics of each model are summarized in form of  
 1396 pseudo - code in Table 5.1.

1397

1398 The three different models have been trained with a combined loss function (MSE + SSIM +  
 1399 MSE on the gradients) on the same training dataset for 10 epochs each. The results showed  
 1400 poor performances of the MSD-Net with respect to the two U-Nets. Slightly higher accuracy  
 1401 was achieved by the modified U-Net (see Fig. 5.5).



**Figure 5.5: 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.

1402 We conclude here the introductory studies on 2D simulated data. These preliminary tests  
 1403 allowed to explore different DL architectures and loss functions and select the optimal choices  
 1404 for the inpainting of BCDI detector gaps.

### 1405 5.3.3 Accuracy VS Gap position

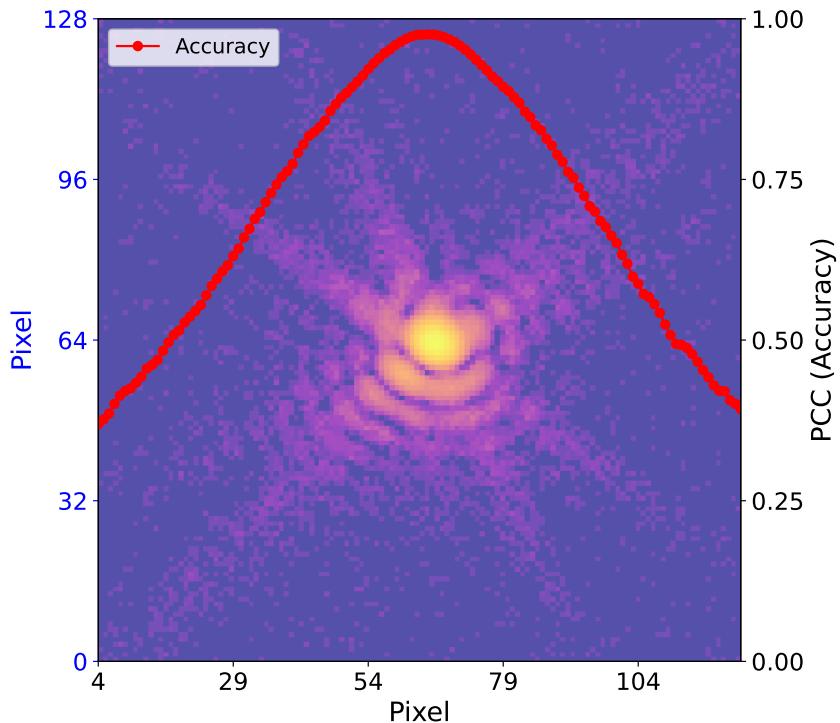
1406 Before moving to the 3D case, it is worth spending a few words on the assessment of the DL  
 1407 model upon different conditions. Namely, the accuracy as function of the position of the gap  
 1408 inside the diffraction pattern and as a function of the oversampling ratio. For the first test  
 1409 150 2D diffraction patterns were simulated starting from random particles shapes, random  
 1410 oversampling ratios and Poisson noise intensity. For each diffraction pattern a vertical 9 pixel-  
 1411 wide gap was placed at all positions from left to right, then the DL prediction and corresponding  
 1412 accuracy score when compared to the ground truth were calculated. The accuracy was again  
 1413 calculated with the Pearson Correlation Coefficient. This score was then averaged for each gap

	U-Net	MSD-Net	U-Net_mod
<b>block1</b>	<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>
<b>block2</b>	<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>
<b>body</b>	<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>
<b>parameters</b>	31,827,673	322,458	32,652,337

**Table 5.1:** Comparison of U-Net, MSDNet, and U-Net\_mod components.

position, over the 150 diffraction patterns and the result was plotted as a function of the gap position. Fig. 5.6 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.

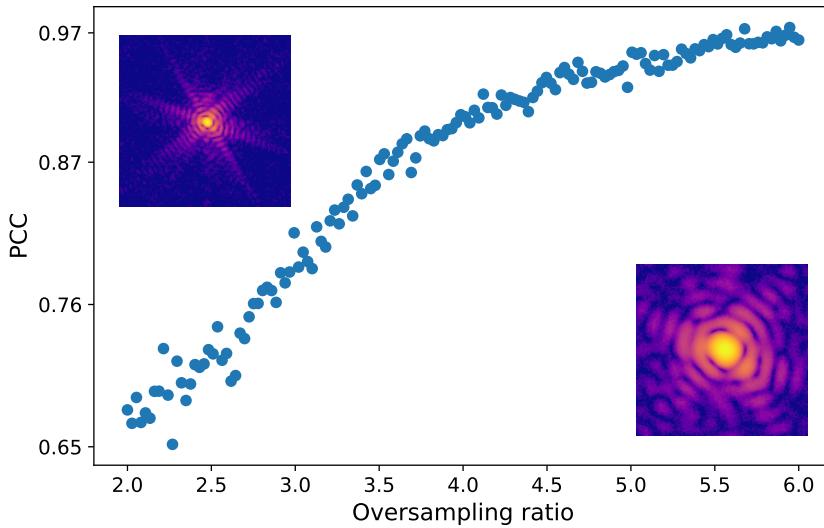


**Figure 5.6: (Accuracy VS Gap position)** Average Pearson Correlation Coefficient calculated over 150 9 pixel-wide vertical predicted gaps for each position of the gap inside the diffraction patterns. The model shows higher accuracies when the gap masks high intensity regions.

To conduct the second test, 150 diffraction patterns were simulated for the same particle varying gradually the oversampling ratio (see Sec.3.1) 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. 5.7). 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 theorem<sup>1</sup>. 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.

<sup>1</sup>Interpreted 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 (a 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 5.7: (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.

## 1448 5.4 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.

1461

The regularity of the oscillations within BCDI datasets, given by the periodicity of the fringes in reciprocal space, is peculiar property of this coherent diffraction technique. It naturally

emerges 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. Such structured periodicity constitutes a crucial element for effective pattern recognition by CNNs and significantly facilitates the inpainting process. The patch-based approach leverages this intrinsic regularity at smaller spatial scales, inherently assuming a certain degree of information redundancy within individual patches. 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 [166] we opted for 32 as a 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).

#### 5.4.1 Dataset creation

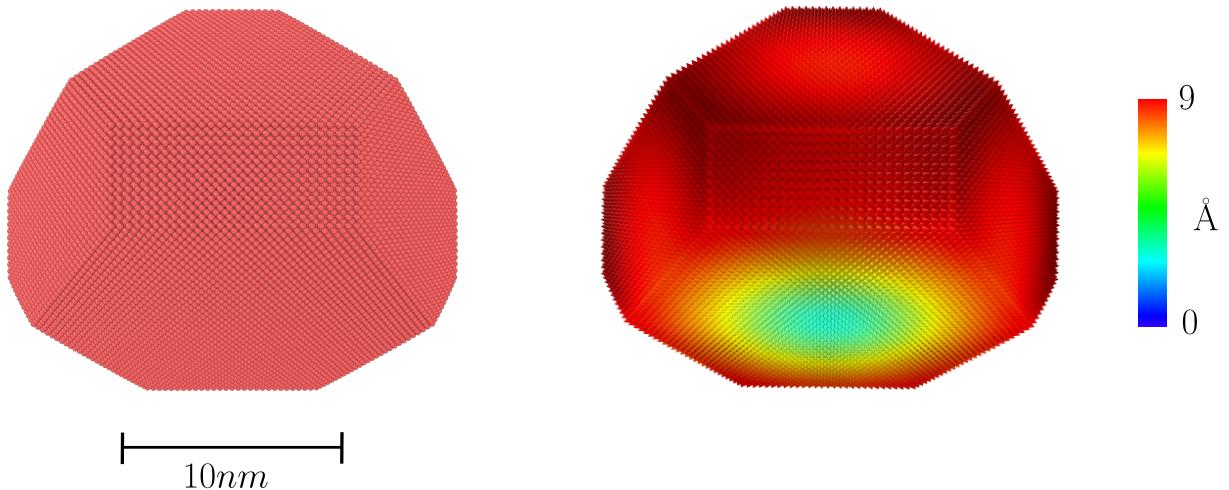
The training dataset consists of 50% patches from experimental data and 50% from simulated data. The experimental measurements were acquired at the ID01 beamline of the ESRF during different beamtimes on different particles. Namely, (i) Pt particles de-wetted on sapphire and YSZ (yttria-stabilized zirconia) 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 [167]. 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 [168]. 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 [56]. 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 2, 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

1508 results in different oversampling ratios.

1509 Before taking portions of these simulated BCDI patterns, Poisson noise was applied, ran-  
1510 domly scaling the  $\lambda$  parameter for each image.

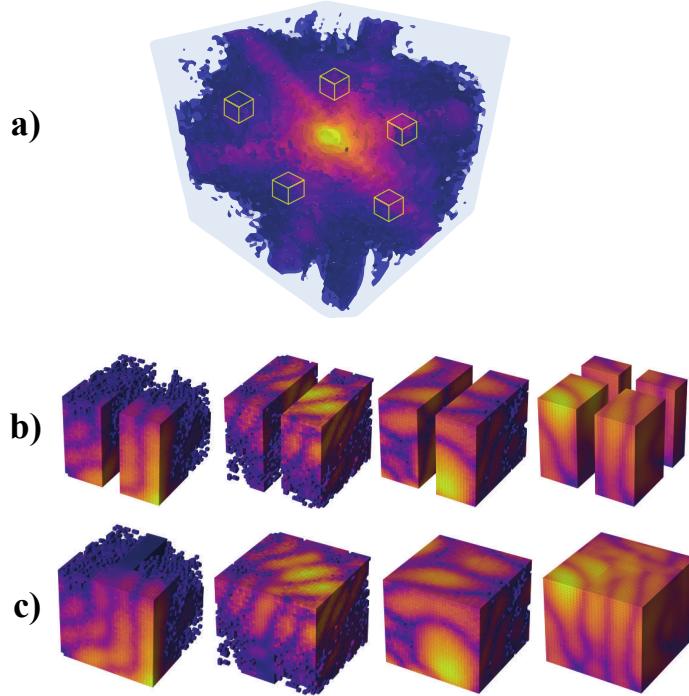


**Figure 5.8:** **Left:** Simulated Au particle with Winterbottom shape (134114 atoms). **Right:** Atomic displacement field of the same particle after LAMMPS energy relaxation showing the typical distribution at the interface with the substrate.

1511 At this point, I proceeded with the extraction of patches taken at *pseudo*-random locations  
1512 inside each 3D pattern. The selection in fact was not totally random as the extraction of patches  
1513 from the outer regions, far from the center of the peak was favored. There are mainly two  
1514 reasons for this choice, namely (i) compensate the inherent uneven accuracy score against  
1515 the position of the gap (see Fig.5.6) by increasing the training data far from the center and  
1516 (ii) emulate as much as possible the experimental conditions, in which unavoidable gaps are  
1517 typically far from the center of the peak. Patches extracted from experimental data were  
1518 selected avoiding possible regions with gaps or dead pixels. For each sub-volume a 3D mask of  
1519 the gap was created for different gap sizes (3,6,9,12 pixel-wide). The gap was placed vertically,  
1520 in the center, along the third dimension, resulting in a “empty slab”. Notice that, while in  
1521 practice a gap may obscure the diffraction pattern at any position, it is unnecessary to consider  
1522 other placements within a patch, since one can always select a patch symmetrically around the  
1523 gap. Cross-shaped gaps were also included in the training dataset, with a population ratio of 1:5  
1524 compared to vertical gaps. They were created by adding a horizontal gap at a random height to  
1525 an existing vertical gap (Fig. 5.9). The final training dataset consisted of 30'000  $32 \times 32 \times 32$   
1526 pairs of patches, with gap and ground truth, created as described above.

## 1527 5.5 3D model architecture

1528 The DL architecture used for the 3D patching inpainting is illustrated in Fig. 5.10. Given  
1529 the reduced size of the inputs, the encoder was composed of four blocks only, in each of  
1530 which there were convolutional layers and max pooling layers. The feature map was thus  
1531 reduced to a  $2 \times 2 \times 2 \times 478$  tensor before being passed to the decoder. Notice that, as  
1532 introduced above in section Sec.5.3, dilated convolutions were employed in the first two blocks  
1533 to enhance the extraction long-range correlated features. After four decoder blocks three simple



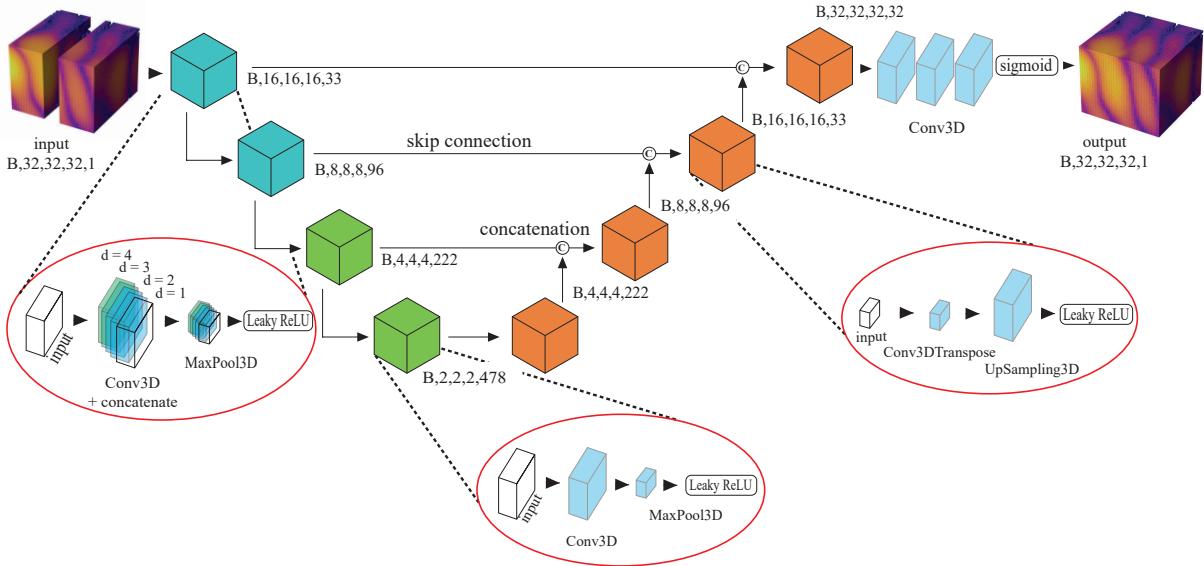
**Figure 5.9: Schematic of the patches extraction.** a) The 3D BCDI diffraction pattern and the patches. b)  $32 \times 32 \times 32$  pixel-size patches with 9 pixel-wide vertical and cross-shaped gaps. c) Same patches with the DL inpainted gaps.

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 ADAM optimizer [122]. 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.

## 5.6 Results in detector space

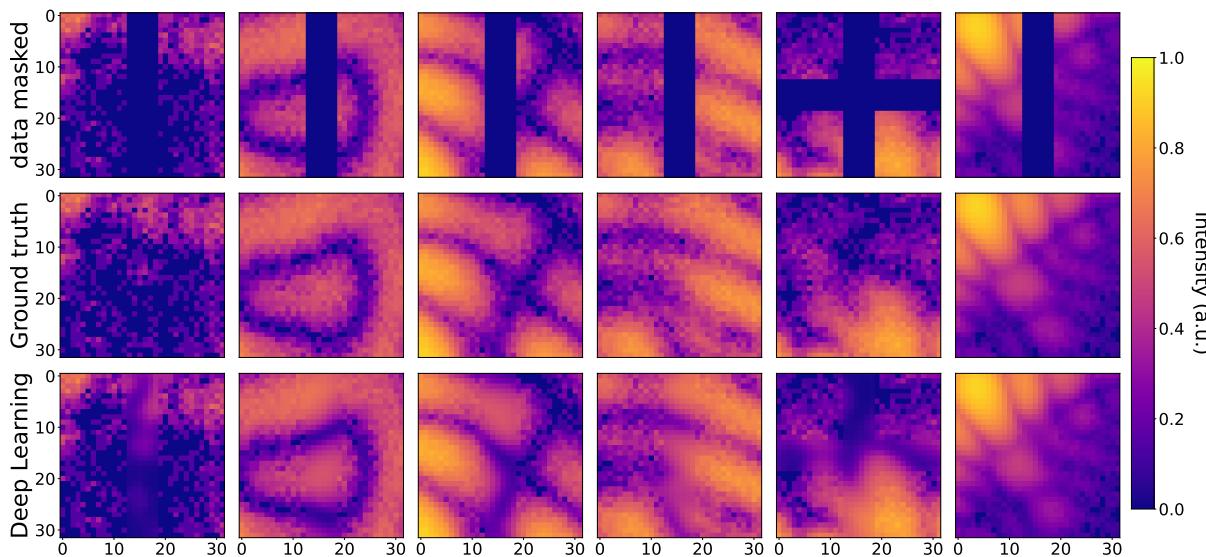
In this section the results of the DL model on both simulated and experimental diffraction patterns are presented. The following section will focus on the results in real space and the reduction of the artifacts in the reconstructed objects.

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. 5.11 - 5.12). 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. 5.7. Another curious effect that can be observed, is the “smoothing” of features around noisy areas (see first column in Fig. 5.11 and last column in Fig. 5.12). In fact,

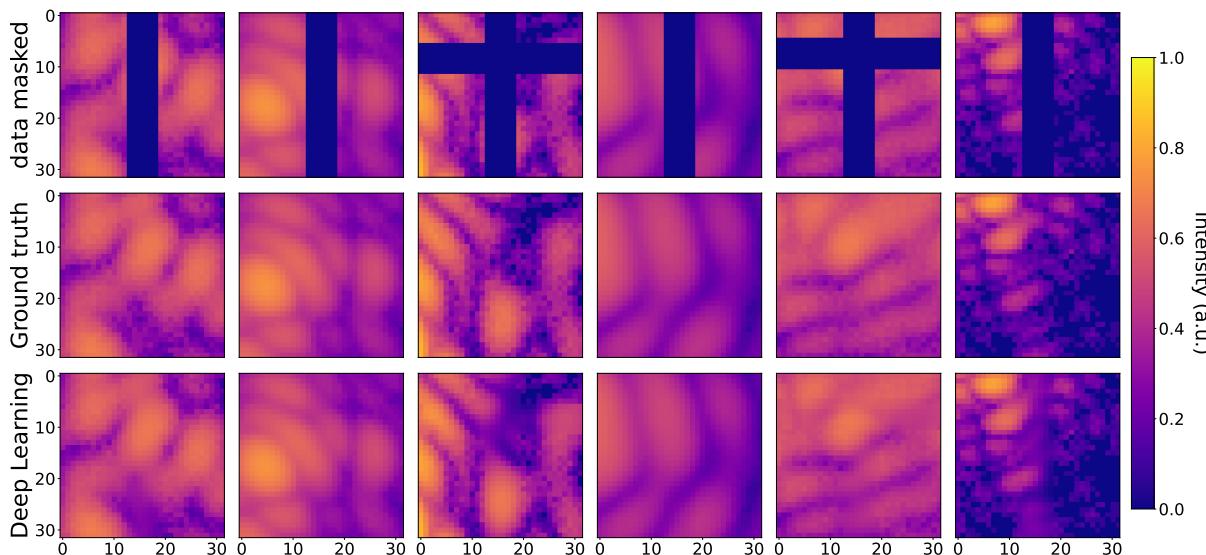


**Figure 5.10: Schematic of the 3D model architecture** The model uses a modified U-Net structure. In the first two encoder blocks (highlighted by the left red circle), dilated convolutions are applied where the original input is concatenated with its convolutions at various dilation rates ( $d = 4, 3, 2, 1$ ) 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.

1554 the “grainy” aspect of these regions is caused by Poisson noise which cannot be predicted by  
 1555 the DL model as it is uncorrelated. In those regions the DL performs therefore an averaging  
 1556 that “smoothes” the features and acts like a denoiser. This effect has been already studied in  
 1557 the literature and exploited for denoising applications like the Noise2Void model [169].



**Figure 5.11: 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.

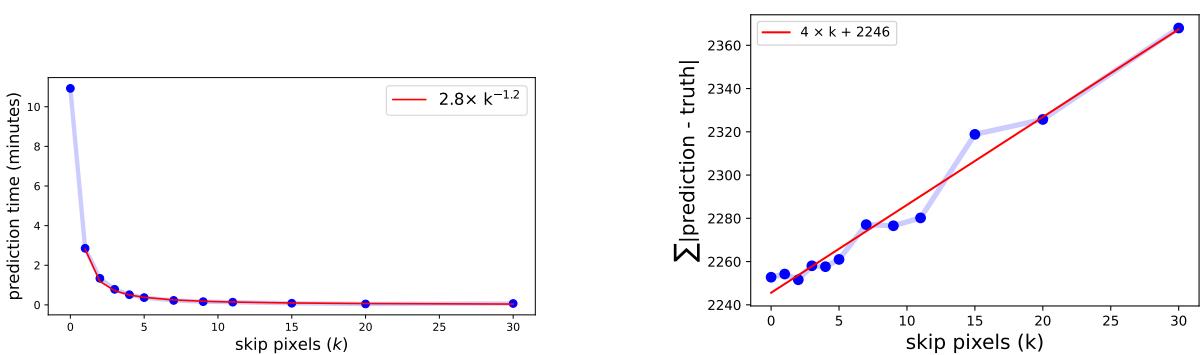


**Figure 5.12: Results on portions of test experimental data.** Central slices of portions taken from the experimental test dataset. Masked input with 6 pixel-wide gap in the first row, corresponding ground truth and DL inpainted in second and third row respectively.

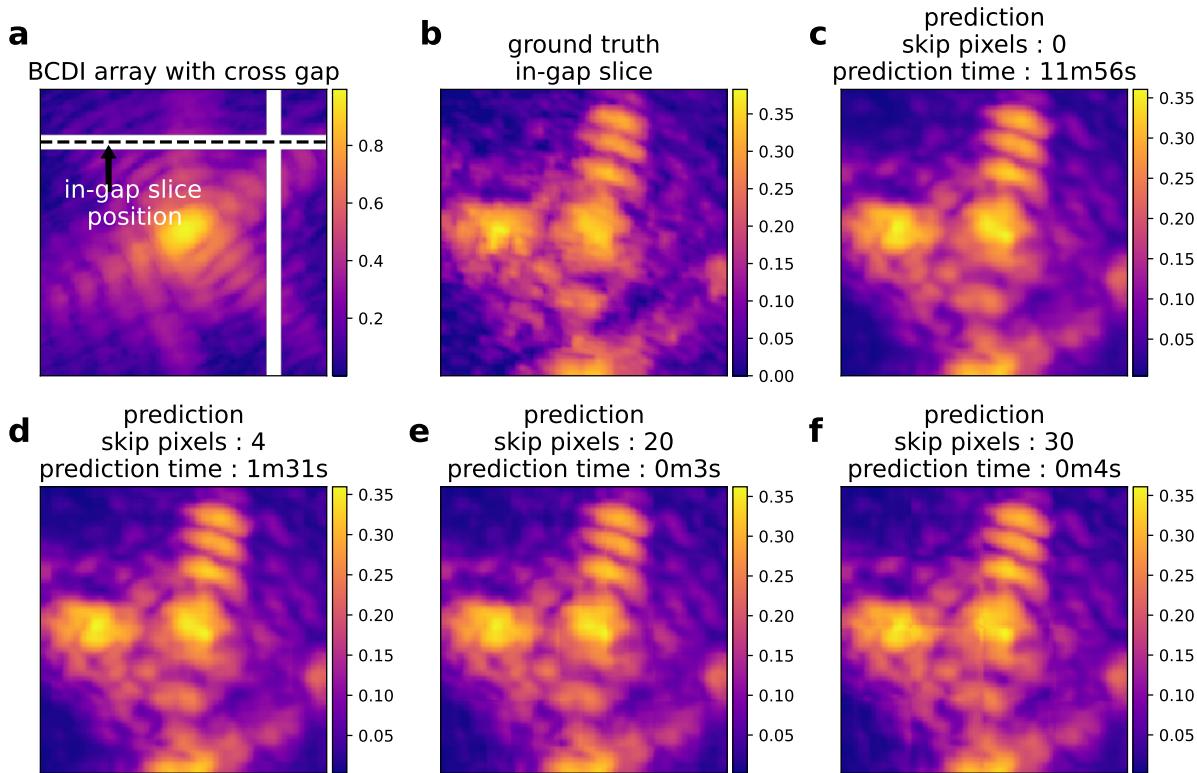
### 1558 5.6.1 Full gap inpainting

1559 For the inpainting of a gap inside a full 3D BCDI pattern it is sufficient to apply repeatedly  
1560 the DL model on sub-volumes cropped such that the gap plane lies vertical in the center of  
1561 the array perpendicularly to the third dimension. Each sub-volume needs to be preprocessed  
1562 exactly in the same way described above, i.e. transformed into logarithmic scale and normalized  
1563 between 0 and 1. Moreover, it is advised to apply a mask on the gap, to match exactly the  
1564 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. 5.14). It was proven that the amount of time for the full inpainting follows a power law (see Fig. 5.13a) and the accuracy starts dropping significantly for more than 5 pixels skipped at the time (see Fig. 5.13b).



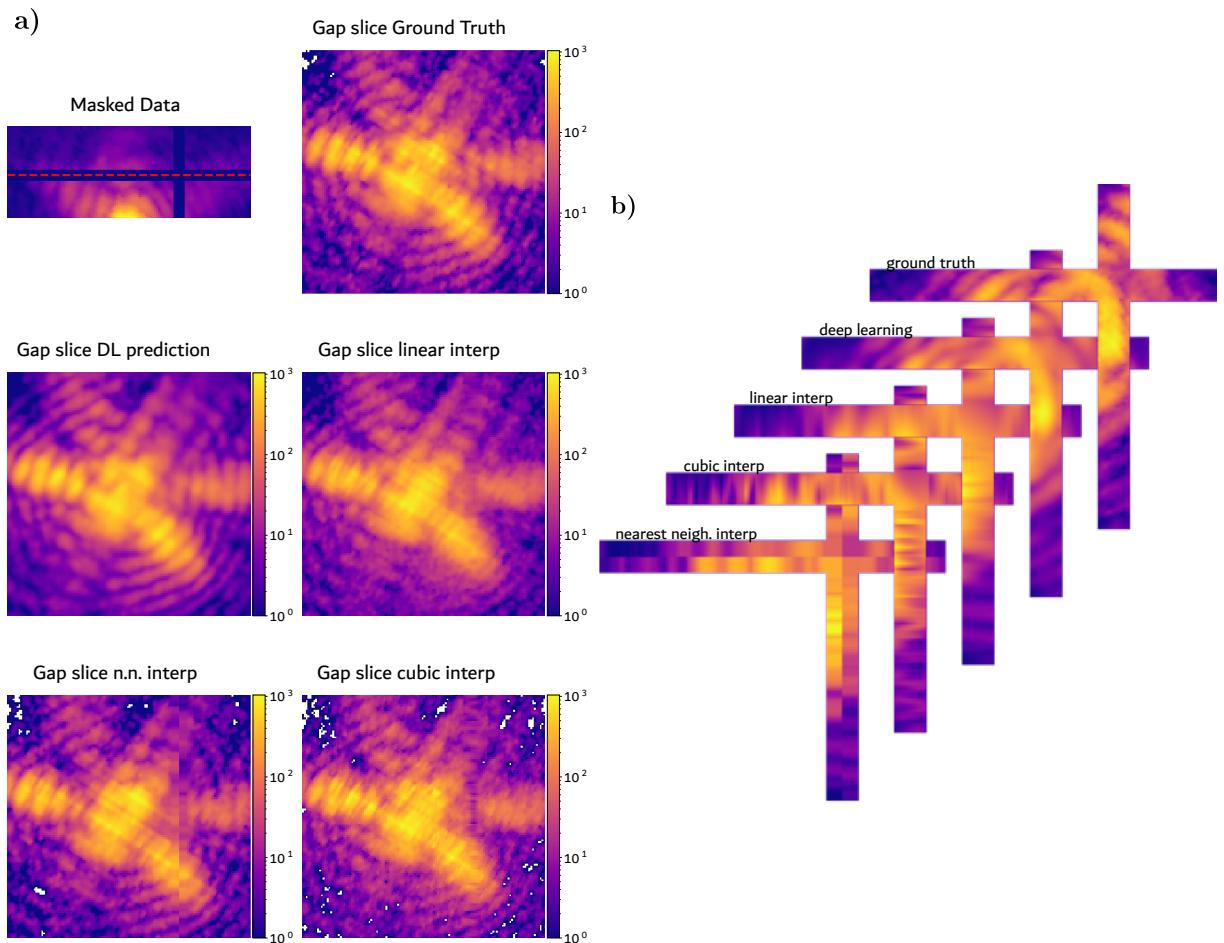
**Figure 5.13:** **Left** Full inpainting time for a 6 pixel-wide cross-shaped gap on a  $128 \times 128 \times 128$  pixel-size diffraction pattern as function of the step size, i.e. the amount of pixels skipped between patch DL predictions along the gap. **Right** Sum of the absolute errors as function of the skipped pixels.



**Figure 5.14:** 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.

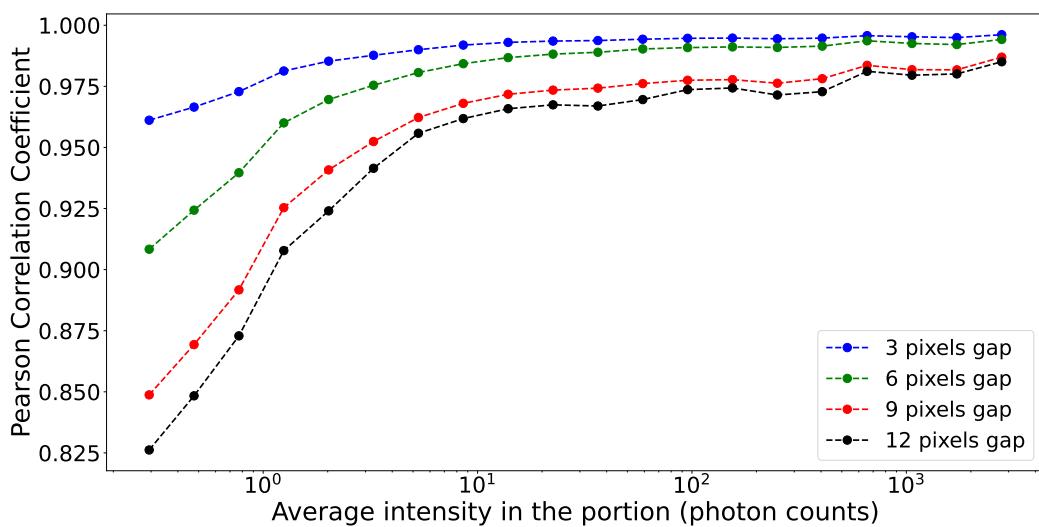
## 1575 5.7 Performance assessment

1576 In order to assess the DL model's performance with respect to other inpainting methods, it was  
 1577 tested against conventional interpolation methods. Specifically, an experimental BCDI pattern  
 1578 with a 6 pixel-wide cross-shaped gap was considered and the inpainting results of the DL model  
 1579 with (i) linear interpolation (ii) cubic interpolation (iii) nearest-neighbor interpolation were  
 1580 compared. These techniques allow for a quick estimation of the intensity distribution inside  
 1581 the gaps but fail to recover fine features (see Figs. 5.15). In particular, it can be noticed in  
 1582 the *in-gap slice* (Fig. 5.15a) that linear interpolation for instance doesn't retrieve correctly the  
 1583 space curvature of the fringes while nearest neighbor and cubic interpolations show artifacts  
 1584 in correspondence of the perpendicular gap. When considering the central slice perpendicular  
 1585 to the gap planes (along the rocking curve dimension) one can notice even more how the DL  
 1586 model outperforms conventional interpolations (Fig. 5.15b).



**Figure 5.15: 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 [169]. 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.

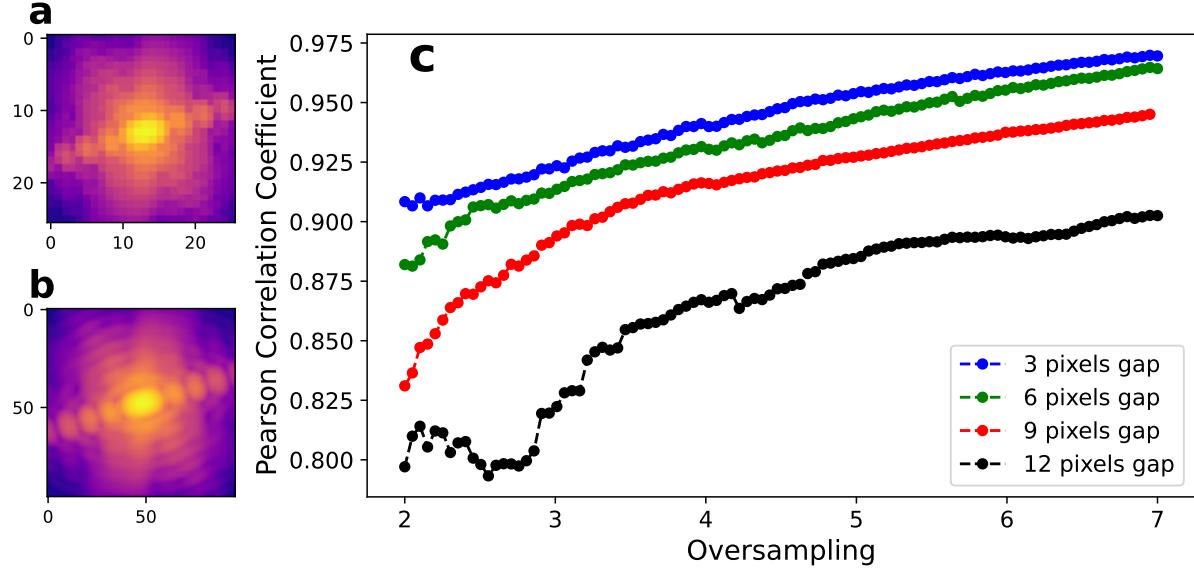
Similarly to the 2D case above, the performance of the model has been evaluated against the amount of intensity inside the sub-volume and against the oversampling ratio. The test was repeated for different gap widths, namely 3,6,9,12 pixel-wide, using vertical gaps placed in the center of each patch. 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. 5.16. 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 5.16:** 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. 5.6.1). 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. 5.17. 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).



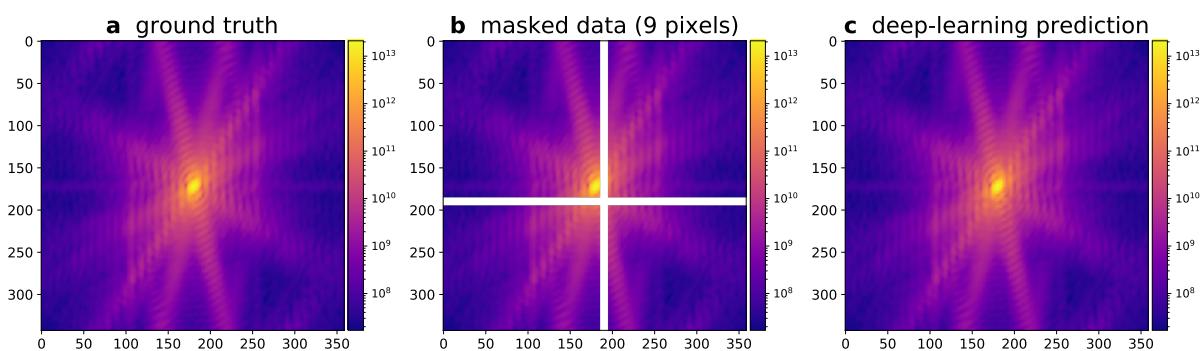
**Figure 5.17: Accuracy scores (PCC) of the DL patching model against the oversampling ratio.** **a)** Simulated diffraction pattern with low oversampling ratio. **b)** The same pattern simulated on a larger grid for an increased oversampling ratio. **c)** Pearson Correlation Coefficient (PCC) of the full inpainted diffraction pattern for the four different gap sizes considered. The trend overall confirms the expectations.

## 5.8 Results in real space

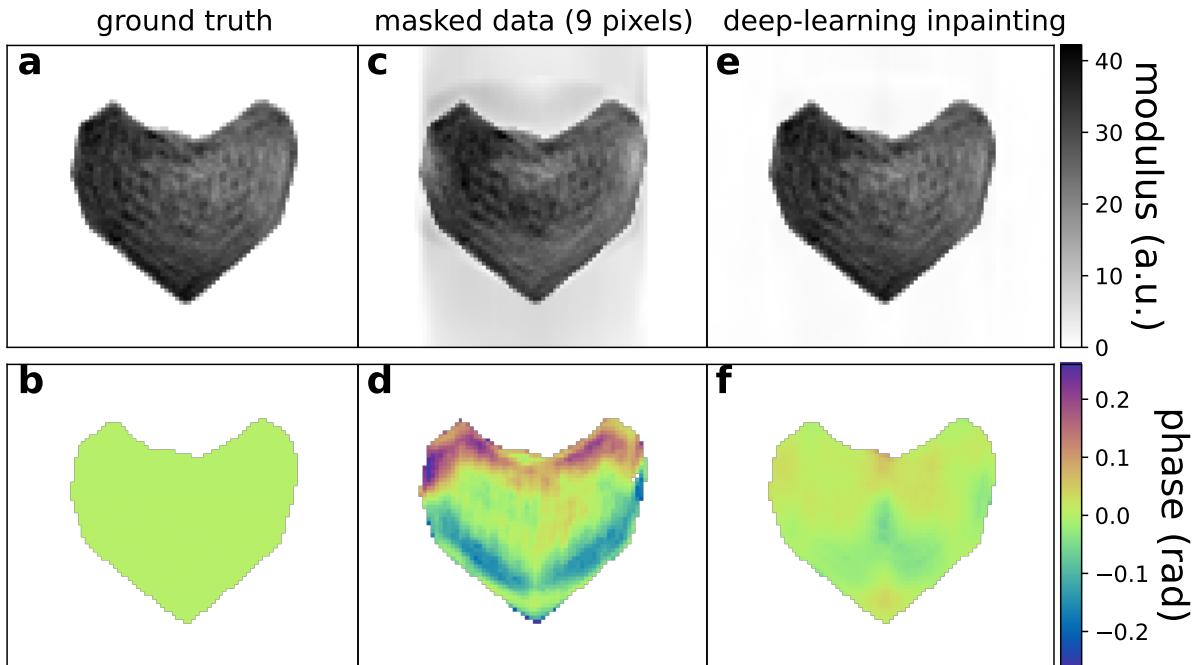
In this section the effects of DL inpainting on the reconstructed objects will be discussed for both simulated and experimental data. In particular, the assessment, both qualitatively and quantitatively, of the gap induced artifacts in the modulus, phase and strain fields of the reconstructions and their reduction thanks to the DL inpainting are presented. 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 [145]. 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.

1634 The procedure was repeated for four different gap sizes (3, 6, 9, 12 px-wide) matching exactly  
1635 the cases mentioned in the work of Carnis and coauthors. Figure 5.18 shows the projection  
1636 along the rocking curve axis (XY slice in this case) of the ground truth, the gapped and the DL  
1637 inpainted diffracted intensity for the 9 pixel-wide gap case.

1638 Notice that in all cases of Fig 5.18, there are vertical and horizontal lines of higher intensity  
1639 not attributable to the typical streaks from the truncation rods. These arise from the detector  
1640 gaps in the original dataset, which are in fact responsible for the oscillatory stripe artifacts  
1641 visible on the object's modulus for all three cases in Fig. 5.19, ground truth included. The DL  
1642 inpainting of these original gaps is presented later in Sec. 5.8.1.

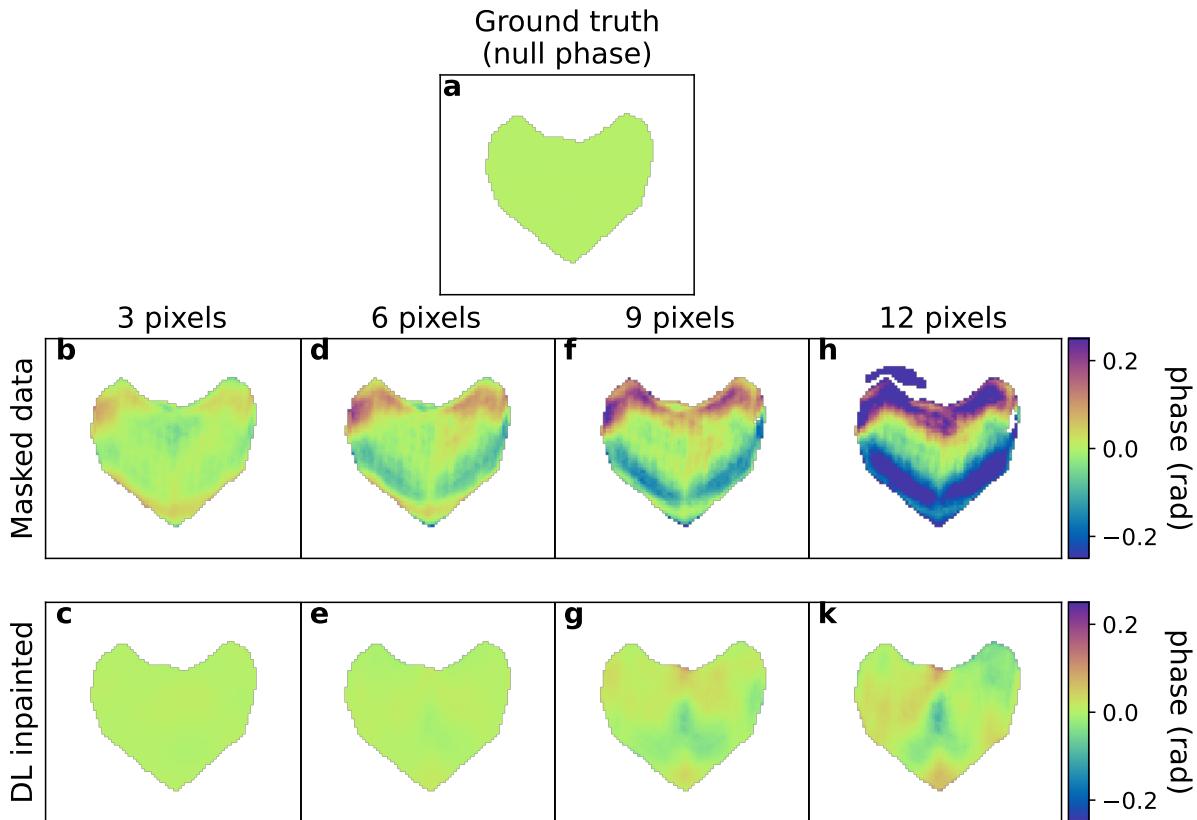


**Figure 5.18: Projections along the rocking curve axis of the studied diffraction pattern in log scale.** **a** Ground truth pattern obtained from the  $|\mathcal{F}(\mathbf{O})|^2$ . **b** Pattern with a 9 pixel-wide cross shaped gap. The position close to the center of the peak is experimentally unlikely 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.

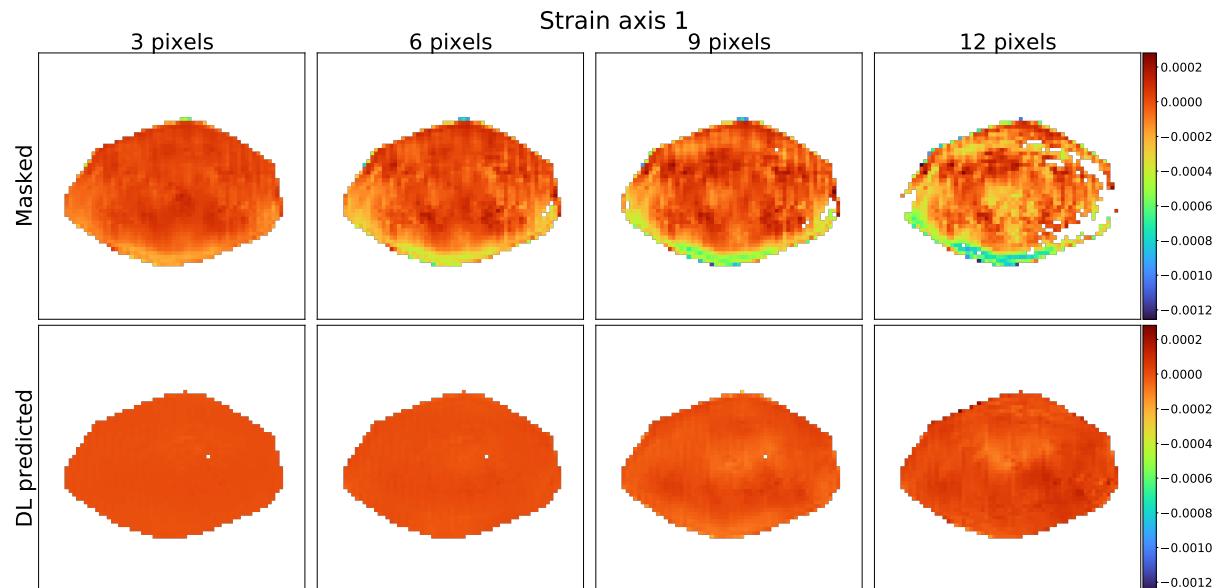


**Figure 5.19: Reconstructed objects (central slice).** a-b XY central slice of ground truth modulus and phase obtained from the reconstruction of an experimental gap-less dataset. The object's phase has been set to zero artificially after the reconstruction for better assessment of the gap-induced artifacts. 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.5.18. 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.

1643     Figure 5.19 illustrates instead the central slice (XY) of the reconstructed objects for the  
 1644 three cases. It is evident that while  $\mathbf{O}_{gap}$  shows significant abnormalities in both modulus and  
 1645 phase,  $\mathbf{O}_{DL}$  is much closer to the ground truth. In particular one can notice that the gap plane,  
 1646 horizontal in the XY plane, induces artifacts along its perpendicular direction. The result is  
 1647 indeed a stripe of non-zero modulus outside the support and, most importantly, an overall  
 1648 phase variation of 0.4 radians along the vertical direction. This phase variation results in an  
 1649 error of  $\pm 7$  pm in the lattice displacement field for the 111 Pt reflection, with more intensity  
 1650 around the surface. These artifacts are particularly problematic in the cases of (electro-)catalytic  
 1651 experiments [22] or in situ gas experiments [170–174], where the particle's surface is primarily  
 1652 involved in the reaction and one could follow the process by monitoring the evolution of the  
 1653 strain in that region. As one could expect the artifacts become more severe as the gap size  
 1654 increases. Fig.5.20 depicts the phases of  $\mathbf{O}_{gap}$  and  $\mathbf{O}_{DL}$  for the different gap sizes considered in  
 1655 this study while Fig.5.21 the strain distribution in the XY plane.

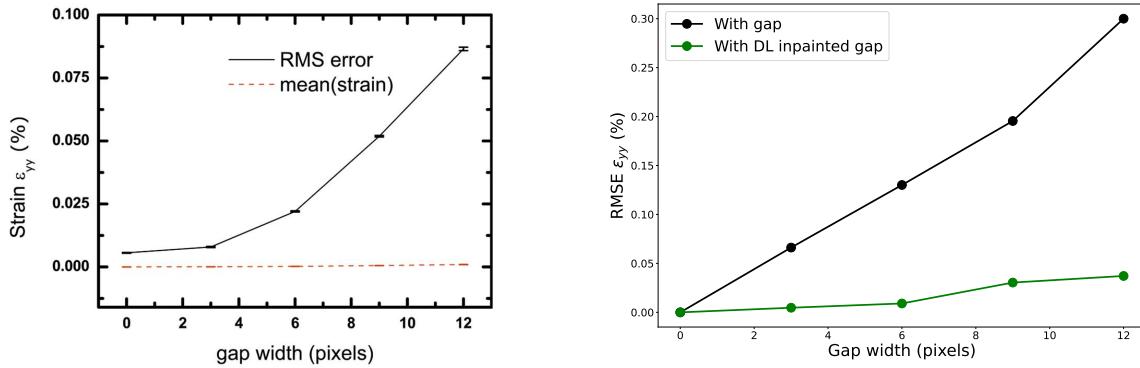


**Figure 5.20: Comparison of the phase artifacts vs gap size.** XY central slice of the phase of  $\mathbf{O}_{gap}$  for different gap sizes (b-d-f-h), and phase of the corresponding  $\mathbf{O}_{DL}$  (c-e-g-k). The comparison with the ground truth null phase (a) shows a significant increment of the 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 5.21: 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.

1656 The deviation from the ground truth zero value of the retrieved strain for both  $\mathbf{O}_{gap}$  and  
 1657  $\mathbf{O}_{DL}$  can be measured with the root mean squared error (RMSE) across all the different gap sizes.  
 1658 This calculation was already proposed in the aforementioned work of Carnis and coauthors in  
 1659 which the results were plotted in Fig.4. We have reproduced a similar figure adding the results  
 1660 of our DL model (Fig.5.22). The trend of the strain RMSE resembles the curve shown in [145],  
 1661 increasing significantly with the gap size while the DL equivalent curve lies below, dampening  
 1662 the error by approximately a factor 5.



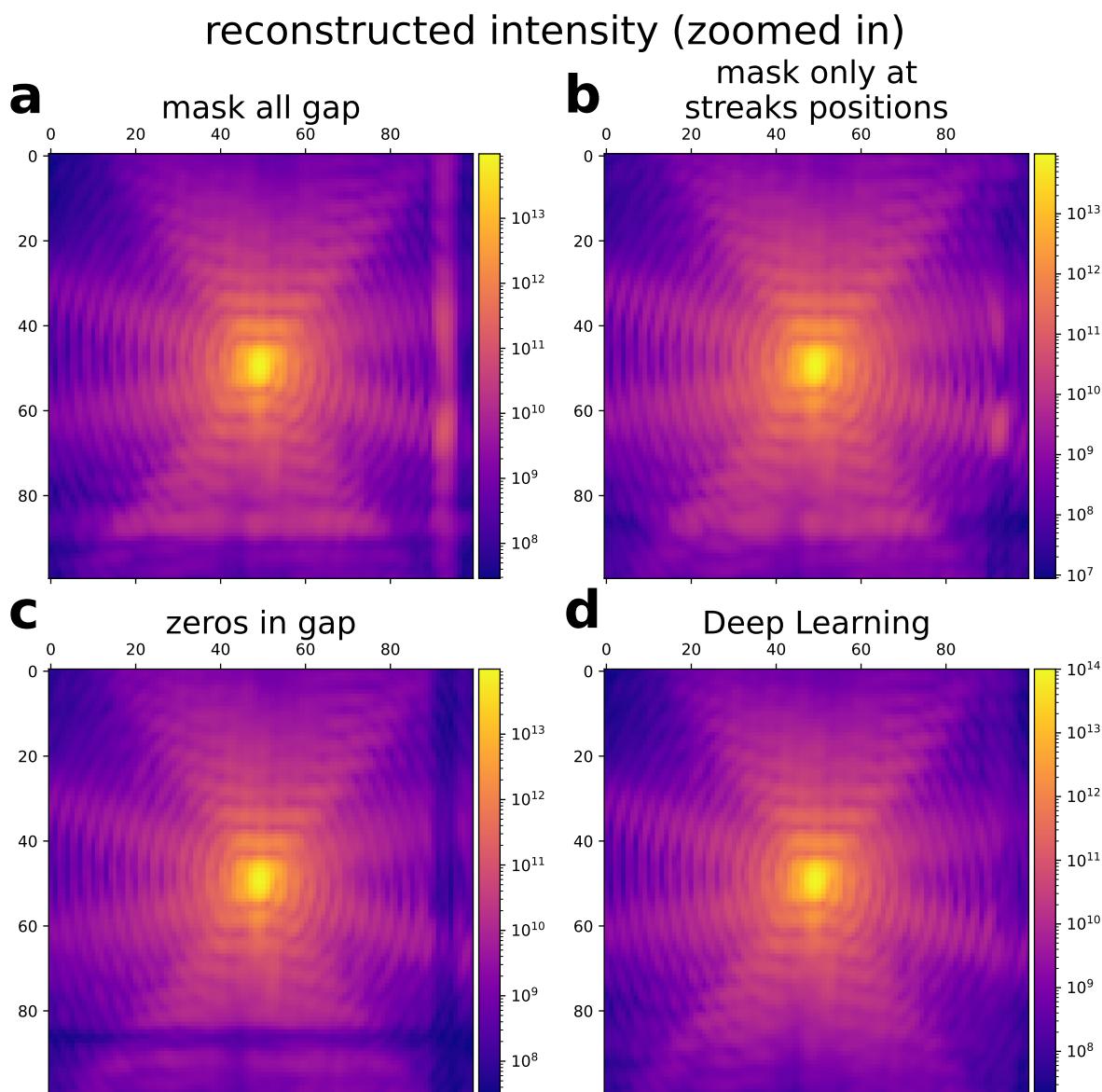
**Figure 5.22: RMSE of the strain field versus gap size(Left)** The plot realized by Carnis *et al.* in [145] depicts the RMSE of the  $\varepsilon_{yy}$  strain component for the objects reconstructed from gapped diffraction patterns with different gap-size. The mean strain around zero corresponds to the null object's phase set artificially. **(Right)** Plot of the RMSE of the  $\varepsilon_{yy}$  strain component versus same gap-size as **(Left)** for gapped (blue line) and DL inpainted (orange line) diffraction patterns.

### 1663 5.8.1 DL inpainting for high resolution BCDI

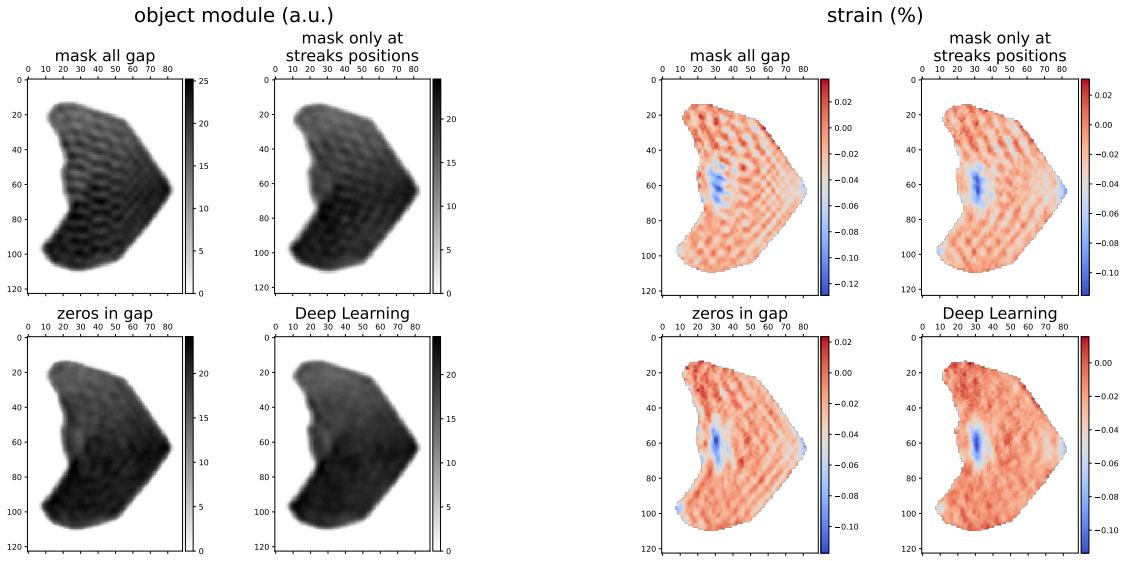
1664 As anticipated in the introduction to the chapter, the most prominent case in which a BCDI  
 1665 datasets is affected by a detector gap is a “high-resolution” dataset. The acquired data here  
 1666 extend for large  $q$  ranges in all directions resulting in parts of the diffracted signal that cross a  
 1667 region on the detector with a vertical or horizontal gap, hence the need for gap-inpainting. All  
 1668 the more so, it is convenient to use a patching approach since treating the full volume would  
 1669 be computationally too expensive. Moreover, any binning or interpolation to smaller sizes will  
 1670 induce information loss, and is not advised.

1671 An example of high-resolution BCDI dataset of this type is the one we have used so far from  
 1672 the work of Carnis and coauthors. The original dataset is indeed a large  $(256 \times 300 \times 300$  pixels)  
 1673 array that contains a cross-shaped, 6 pixel-wide gap. Here we show how the artifacts  
 1674 can change depending on the type of masking of the gaps is chosen during the phasing and  
 1675 how our DL model can outperform these methods. A common approach, when using PyNX  
 1676 software, is to mask the gap such that those pixels don't contribute during the phasing and  
 1677 are left free to evolve **(a)**. Moreover, one could mask only near the intensity streaks affected  
 1678 by the gap **(b)** or simply leave the gap with zeros and remove the contribution of the gap  
 1679 voxels during the whole phasing **(c)**. These strategies have been used during the phasing of the  
 1680 cited Pt diffraction pattern and the results in object space compared with that obtained from  
 1681 the DL inpainted pattern. The results, illustrated in Figs. 5.23 - 5.24, show that the amount

1682 of oscillatory artifacts progressively decreases as we go from method **a** to **c**, proving the DL  
 1683 inpainting to be the optimal method among them.



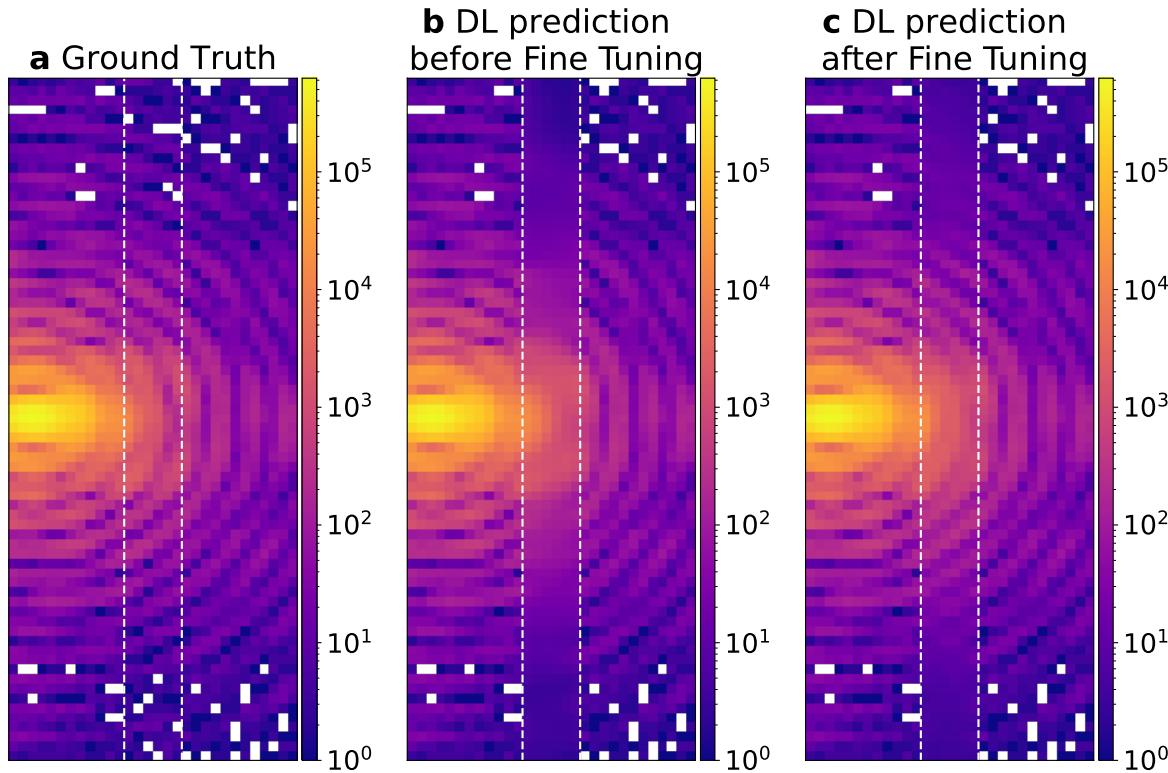
**Figure 5.23:** Zoom on the projection along the rocking curve axis of the Pt BCDI pattern in Fig.5.18 calculated from the reconstructed object obtained with PyNX software using **(a)** a mask on the gaps, **(b)** a mask on the streaks only, leaving zeros inside the gaps **(c)** and inpainting the gaps with the DL model **(d)**. Inside the regions where the mask was applied the intensity is free to vary throughout the PR iterations.



**Figure 5.24:** Modulus (**Left**) and strain (**Right**) of the object's reconstructions for each case mentioned above in Fig. 5.23. The oscillatory artifacts are smallest for the object obtained after DL inpainting (bottom right corner on both left and right images).

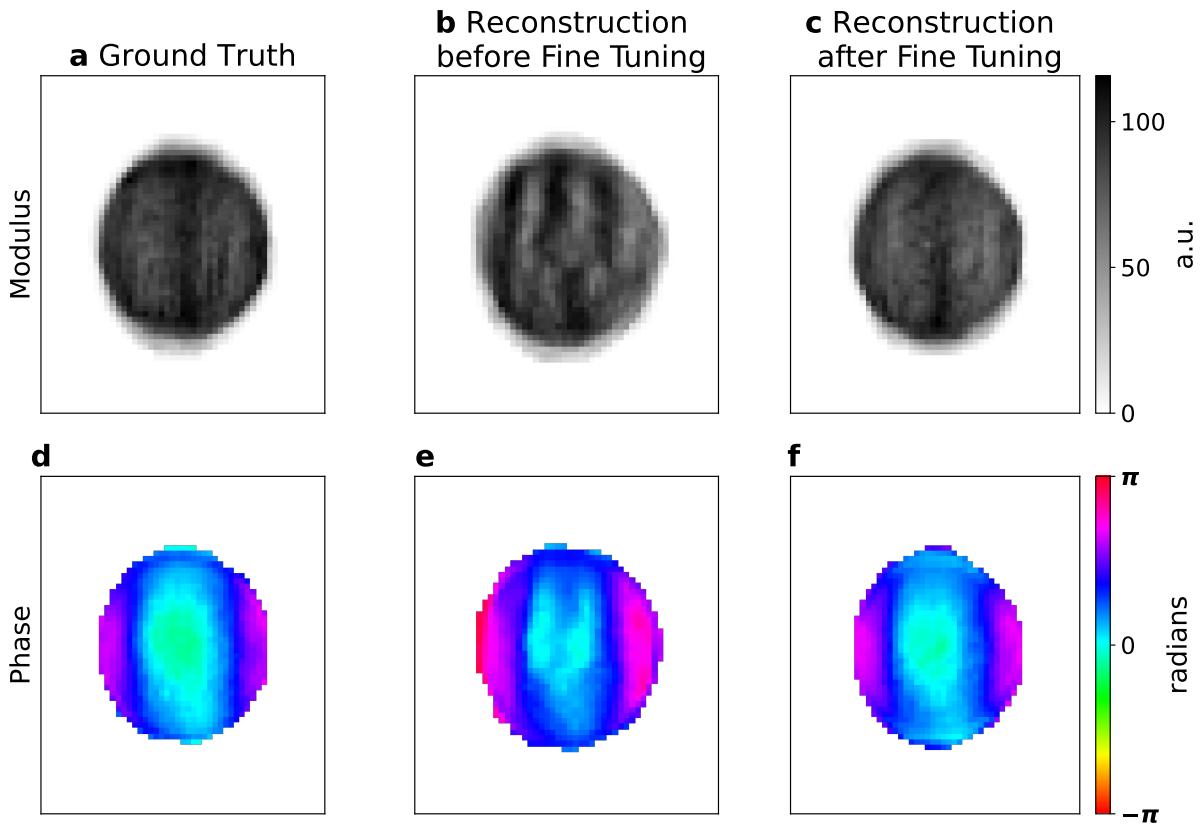
## 1684 5.9 Fine-tuning

1685 For those cases in which the DL model does not yield satisfactory results when inpainting a new  
 1686 experimental BCDI pattern a fine-tuning of the model was conceived in order to improve the  
 1687 accuracy of the prediction. This fine-tuning is enabled by the patching approach as it consists  
 1688 of a secondary short training of the general model on a small dataset made of portions extracted  
 1689 from the new BCDI pattern to be inpainted. In particular, after loading the gap affected BCDI  
 1690 pattern, 2000 portions were randomly cropped out of it, paying attention not to include the  
 1691 gap region. The model has been trained for the corresponding gap width for 2 epochs with  
 1692 a learning rate of  $10^{-5}$  and batches of 32 images ( $\sim 2$  mins on a NVIDIA Tesla V100-SXM2  
 1693 GPU with 32GB RAM). Biassing the model to fit the features of that specific diffraction pattern  
 1694 (oversampling ratio, particle shape, noise level, fringes shape) better results can were obtained  
 1695 on the real gap. An example is shown in Fig.5.9. There, the general DL model was not able to  
 1696 predict the fringes with the correct periodicity inside the gap. After the fine-tuning instead,  
 1697 the model properly recovers the fringes improving the accuracy. This fine-tuning technique is  
 1698 a further example of the advantage of using a patching approach and its usage depends on the  
 1699 user judgement on the quality of the general model inpainting.



**Figure 5.25: 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.

1700 The effect of the fine-tuning can also be assessed on the reconstructed object. Here, the  
 1701 diffraction patterns showed in Fig.5.25 has been phased with PyNX and the results are compared  
 1702 in Fig.5.26. For the phase retrieval, in all the three cases a single run of 400 HI0 + 1000 RAAR +  
 1703 300 ER iterations has been performed starting from the autocorrelation of the object and with  
 1704 the shrinkwrap threshold of 0.2. As expected, already from visual inspection, the modulus and  
 1705 phase of the object reconstructed from BCDI pattern inpainted after the fine-tuning are more  
 1706 similar to the ground truth ones. On the contrary, the one obtained before the fine-tuning  
 1707 shows some oscillatory artifacts caused by the not correctly restored fringes.



**Figure 5.26: Reconstructed object before and after fine-tuning of the DL model** From the diffraction pattern in Fig. 5.25, **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. 5.25**b** produce artifacts in the reconstructed object that are eliminated with the inpainting of the fine-tuned DL model.

## 1708 5.10 Conclusions

1709 A novel DL model for the inpainting of BCDI patterns with detectors' gaps has been presented  
 1710 and tested for different experimental circumstances. The “patching approach” has proven to be  
 1711 adequate for BCDI for different reasons here summarized.

- 1712     • Due to the oversampling condition, the BCDI patterns exhibit spatially regular and well-  
 1713       separated oscillations. This local “redundancy” in detector space can be exploited by  
 1714       isolating smaller sub-volumes of the diffraction pattern (*patches*). In the presence of gaps,  
 1715       the missing signal within each patch can then be reconstructed from neighboring voxels,  
 1716       with the reconstruction accuracy depending on the relative size of the gap compared to  
 1717       the patch.
- 1718     • The patching approach naturally allows for larger training datasets, faster training  
 1719       compared to full-image direct inpainting approaches. Moreover, no lossy resizing is  
 1720       needed to adapt the whole BCDI pattern to the DL model size, as in full-image direct  
 1721       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, at the cost of only a few extra minutes of computation.

The presented DL model is planned to be implemented in the PyNX package in the near 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.

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## 1742 DEEP LEARNING FOR PHASE RETRIEVAL

1743 We enter now the core topic of the thesis. Most of the efforts during this PhD have been  
 1744 dedicated to the study of the Phase Problem for Bragg Coherent Diffraction Imaging using  
 1745 DL based approaches. Here I will discuss the main steps of this journey, starting off from the  
 1746 analysis of the most relevant works in literature and concluding with some comments on the  
 1747 final version of a DL model for highly strained particles. The latter has become the subject  
 1748 of an article, currently submitted, entitled “*Phase Retrieval of Highly Strained Bragg Coherent*  
 1749 *Diffraction Patterns with Supervised Convolutional Neural Network*” [175]. The process that led to  
 1750 the final version of the model will be unraveled, and particular attention will be given to  
 1751 elucidating the key steps and the critical issues encountered along the way. The structure of  
 1752 the chapter can be summarized as follows:

- 1753 • **State of the art and Reciprocal space phase phasing:** introduces the works on DL for  
 1754 phase retrieval in BCDI and discusses the novel concept of the prediction, using DL, of  
 1755 the reciprocal space phase (RSP) lost during the measurement.
- 1756 • **2D case and Weighted Coherent Average loss function:** presents the preliminary  
 1757 studies on the 2D case, for low-strain and high-strain simulated particles. Introduces the  
 1758 novel “Weighted Coherent Average” loss function, designed specifically for the prediction  
 1759 of complex phases.
- 1760 • **3D case - Patching approach:** discusses a patching-method for the low-strain and  
 1761 high-strain cases in 3D.
- 1762 • **Final 3D model:** presents the final DL model for the prediction of the RSP of highly  
 1763 strained 3D BCDI patterns. Results on simulated and experimental data are shown as  
 1764 well as the combination of DL prediction and iterative algorithms for refinement.
- 1765 • **Performance assessment:** the final DL model and standard algorithms are tested on  
 1766 simulated data with different strain distributions to assess the benefits of the DL approach.

## 1767 6.1 State of the art

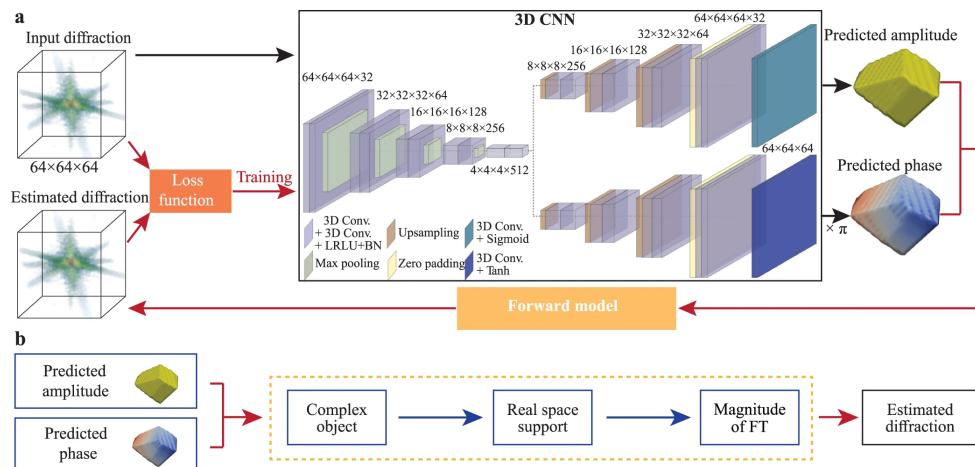
1768 In this paragraph I will focus on the state of the art for what concerns the Phase Retrieval of  
1769 BCDI diffraction patterns with deep-learning, tensor-computation and automatic differentiation  
1770 methods. Conventional phase retrieval iterative algorithms are discussed in the introduction  
1771 chapter as well as other approaches.

1772  
1773 Given the relatively new development of neural networks and more specifically even more  
1774 recent for BCDI phase retrieval, I will try to give a chronological overview of the main works  
1775 in the literature pointing out strengths and weaknesses. The use of deep neural networks for  
1776 inverse problems in imaging was proven to be successful in lensless computational imaging by  
1777 Sinha in 2017 [176] and holography by Rivenson [177]. However, the first work pioneering  
1778 the field for BCDI is “Real-time coherent diffraction inversion using deep generative networks”  
1779 published by Cherukara *et. al* in 2018 [178]. The paper presents two CNNs for the phase  
1780 retrieval of small ( $32 \times 32$  pixels) 2D simulated BCDI patterns, one predicting the support  
1781 and the other the phase. A U-Net like architecture with encoder-decoder was implemented,  
1782 and the model was trained for 10 epochs in a supervised fashion with a cross-entropy loss  
1783 function. The results showed an excellent agreement between prediction and ground truth also  
1784 in presence of relatively strong phases. The potential of this new approach for phase retrieval  
1785 becomes immediately clear when considering the drastic reduction of computational time and  
1786 resources needed for the model inference. Once the model is trained, the reconstruction can be  
1787 obtained within a few milliseconds on a desktop machine.

1788 In 2020 Scheinker and Pokharel proposed another approach [179] that employs a CNN model  
1789 for 3D diffraction patterns. The fundamental difference is that the object’s support was defined  
1790 by its surface only, as it is assumed to be *compact* and *homogeneous* inside. Moreover, the surface  
1791 was parametrized by spherical harmonics and the DL model was trained to predict 28 of the first  
1792 even coefficients of the spherical harmonics. The model architecture was therefore essentially  
1793 different since, while the encoder is just transposed to a 3D one, the decoder is replaced by  
1794 a flattening and dense layer with 28 different classes as output. The model showed good  
1795 performance on both simulated and experimental data, marking the first DL-based approach  
1796 capable of real 3D BCDI phase retrieval. In the same year, Wu and coauthors, [180], opted  
1797 for an architecture made of a single encoder and two identical decoders for the prediction  
1798 of amplitude and phase of single crystals from the central slice of the BCDI pattern. They  
1799 conducted the study on simulated data and tested it on one experimental case as well. What is  
1800 evident from their work is the winning combination of DL prediction and iterative refinement.  
1801 The speed and generalization capabilities of the CNN allows for fast and good estimations of the  
1802 object’s support and phase. In addition, the precise and well established iterative methods can  
1803 bring this initial guess to a more polished and accurate solution in fewer cycles than without  
1804 DL prediction. This successful combined approach has been later adopted in other works, ours  
1805 included.

1806 In 2021 two important works were published. First, Chan *et al.* in [181] extended the  
1807 encoder/2-decoders architecture to the 3D case. In their work they first created a “physics-  
1808 informed” training set obtained building particles by clipping planes from a cubic FCC structure  
1809 of atomic positions, relaxing them with LAMMPS software for molecular dynamics and com-  
1810 puting the BCDI pattern around the (111) Bragg peak. The procedure is very similar to the  
1811 one adopted by Lim *et al.* in [167] and described above in Section 5.4.1. 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.* [182] 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 [183], 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.



**Figure 6.1: Schematic of the AutoPhaseNN model for unsupervised BCDI Phase Retrieval.** **a** The convolutional neural network is divided into an encoder and two parallel decoders for object's modulus and phase respectively. **b** The loss function is calculated in reciprocal space, between the observed and calculated intensities, therefore enabling unsupervised training. Adapted from [183]

Although of different nature, it is worth mentioning the work of Zhuang and coauthors [184] in which two CNNs are used in the “deep image prior” (DIP) framework. DIP [134] 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 [185] (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 the presence of strong phases. The last interesting contribution is the work of Yu and *et al.* [186]. 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 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 6.1 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.

	Architecture	Last Activation Layer	Loss Function	Refinement
Cherukara - 2018 [178]	Two different UNets	Sigmoids	Cross Entropy	-
Wu - 2020 [180]	Encoder / 2 Decoders	ReLU	MSE on mod and phase + PCC on magnitudes	Iterative
Chan - 2021 [181]	Encoder / 2 Decoders	ReLU	MAE on normalized magnitudes	Automatic Differentiation
Wu - 2021 [182]	Encoder / 2 Decoders	LeakyReLU	MSE on mod and phase + PCC on magnitudes	Transfer learning + unsupervised training
Yao - 2022 [183]	Encoder / 2 Decoders	Sigmoid and Tanh	MAE on normalized magnitudes	Iterative (50 ER)
Yu - 2024 [186]	Complex encoder-decoder + skip connections	ReLU	MAE on real + MAE on imaginary	Transfer learning + unsupervised training

**Table 6.1:** Comparison of deep learning-based phase retrieval approaches.

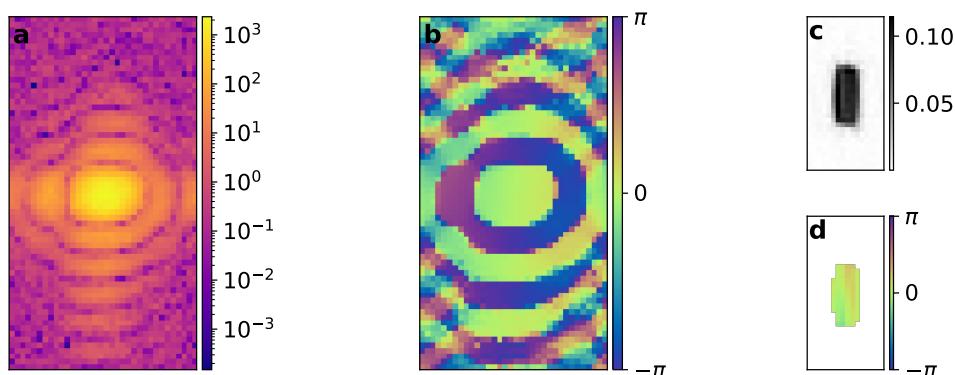
First, it is interesting to notice that the architecture's choice, from treating the object's modulus and phase separately with two different detached networks, moved over the years to a single "standard" U-Net that accounts for the complex nature of the data. Second, I noticed that the choice of the 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.

1875

1876 One of the most important components of the model is the loss function, as its gradients  
 1877 regulate the model's parameters, hence the prediction. The first work that employs a cross  
 1878 entropy loss, normally used for classification tasks, whereas other works opt for Mean Absolute  
 1879 Error (MAE) and Mean Squared Error (MSE), which are standard for regression tasks and  
 1880 Pearson Correlation Coefficient (PCC). Typically, when the loss is calculated between intensities  
 1881 the MAE and the PCC are used as they are more suitable for the high dynamic range of  
 1882 the diffraction patterns. MSE in fact, “would overly de-emphasize errors in mid-intensity  
 1883 regions of the images” [181]. Lastly, I have listed the different ways used to refine the DL  
 1884 predictions. Here we can notice that GPU accelerated gradient descent methods have been  
 1885 used to replace of conventional iterative algorithms. The unsupervised training allows to  
 1886 easily switch from inference to refinement using the same model in the same GPU optimized  
 1887 computing environment guaranteed by machine learning libraries like PyTorch and Tensorflow.

## 1888 6.2 Reciprocal space phasing

1889 Taking inspiration from these works, but significantly changing the perspective, it was de-  
 1890 cided to predict the “reciprocal space” phase (RSP) that is lost during the measurement of  
 1891 the BCDI pattern rather than the complex object in real space. The main, intuitive, reason  
 1892 behind this choice is the visual similarity between the morphology of the diffraction pattern  
 1893 and its corresponding RSP. Furthermore, it is common that many samples studied with BCDI  
 1894 have facets that happen to be, to some degree, parallel with each other, thus interfering like a  
 1895 single slit, with the typical fringes of intensity that correspond to constructive interference,  
 1896 interspersed with dark regions arising from destructive interference. In these specific cases,  
 1897 the RSP shows a regular pattern in which there is always a  $\pi$  shift between two crests of the  
 1898 fringes as shown in Fig.6.2. Once the RSP is retrieved, one can then recompose the full complex  
 1899 diffracted wave-function and obtain the complex object via inverse Fourier transform.  
 1900



**Figure 6.2:** 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  $\pi$  (or  $-\pi$ ) in correspondence of the intensity fringes.

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 (see Section 3.2) that I have encountered during the development of the algorithms and that will be discussed in the next sections.

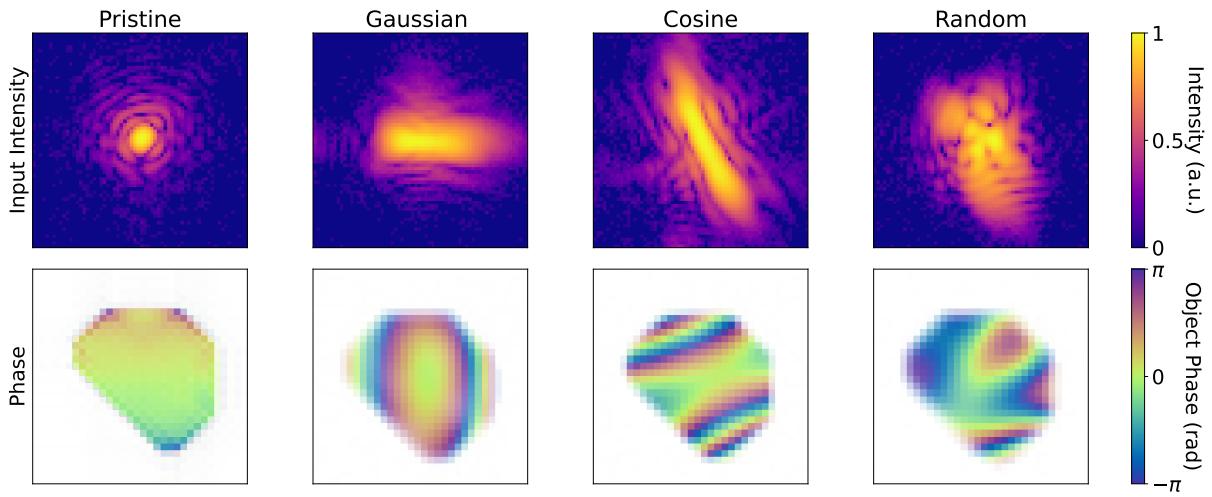
Ultimately, the goal of this DL model for phasing is to facilitate the reconstruction of highly strained particles. While other works in literature have mostly leveraged the gain in computing time, here the model aims at tackling those reconstructions for which conventional algorithms struggle to find convergence because of the high strain in the particle. However, in this case, the aforementioned RSP  $\pi$ -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 2.6).

## 6.3 Dataset creation

I have trained the 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 5.3.1 and 5.4.1 for the 2D and 3D cases, respectively. However, in this case 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 [161]. In each case, centers, amplitudes, variances, frequencies, and correlation lengths were randomly chosen to ensure a phase variation within the particle ranging between  $\pi$  and  $5\pi$ . By doing this, strongly distorted BCDI patterns, similar to experimental high-strain ones, were obtained. In particular, the two Gaussian and cosine functions phase can closely emulate the effect of the substrate induced strain inside Winterbottom particles. (see Fig.6.3)

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.



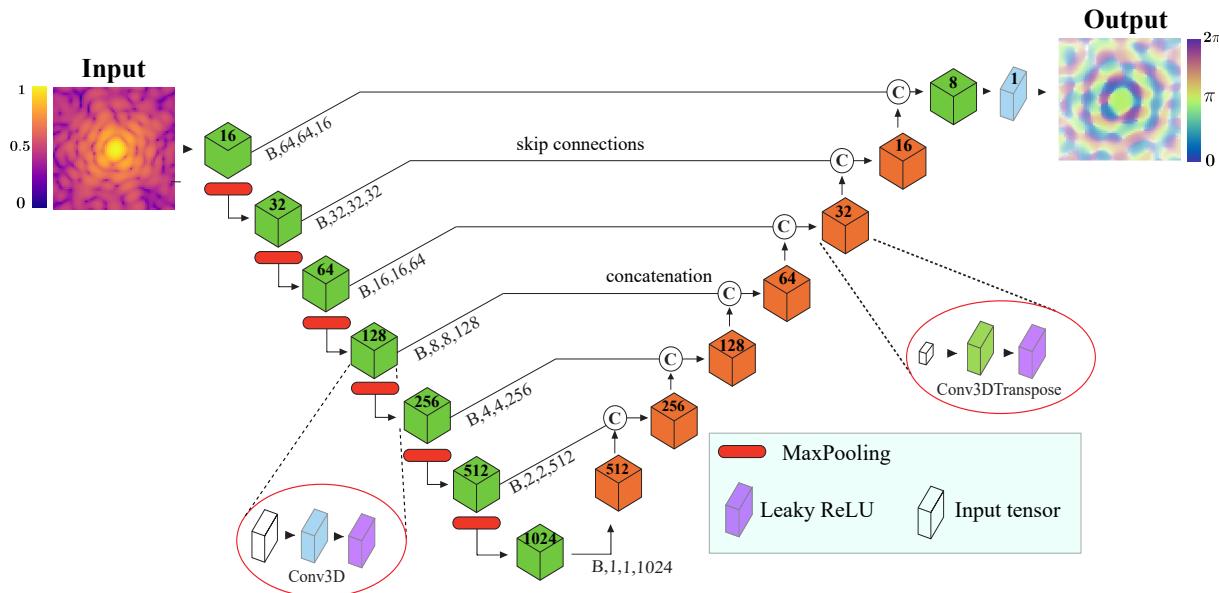
**Figure 6.3: Different simulated strain distributions.** Examples of each of the three adopted methods to simulate the high-strain. From the left, non-strained diffraction pattern and the corresponding pristine particle (central slice); the diffraction pattern obtained from the same particle with a phase built with two Gaussian functions, and relative object's phase slice; the diffraction pattern when the phase is simulated with two cosine functions and corresponding object's phase slice; the diffraction pattern obtained from the same particle with an applied gaussian-correlated random phase field. It is interesting to notice that for the first two objects, the variation of the phase are more pronounced along a specific direction, the same for which the diffraction pattern show an elongation of the Bragg peak, causing a visual “fusion” of the fringes. This phenomenon is often observed experimentally for particles grown on crystalline substrates (see Fig.6.40) [96, 101, 187].

## 1933 6.4 2D case low strain

1934 Similar to the approach for the inpainting case, I have first conducted some preliminary studies  
 1935 in 2D, on noise-less low strain data. Here I will briefly show the model’s architecture, the loss  
 1936 function and the results.

### 1937 6.4.1 Model structure

1938 The architecture that I used has a U-Net like structure with an encoder and a decoder. The  
 1939 encoder is composed of six convolutional blocks through which the input diffracted intensity is  
 1940 progressively reduced from the 64 pixel-side squares to a 1D flattened vector. Each convolutional  
 1941 block is composed of a convolutional layer, a LeakyReLU activation function and a MaxPooling  
 1942 layer that halves the feature’s map dimensions (see Fig. 6.4 for details). At the end of the encoder  
 1943 the so-called bottleneck, composed of a convolutional layer followed by a LeakyReLU activation,  
 1944 processes the feature map before passing it to the decoder which by means of transposed  
 1945 convolutions (with double stride to upsample each image) and LeakyReLU activations, brings  
 1946 back the feature map to the input’s size. Skip connections between encoder and decoder blocks  
 1947 are employed as well. The output tensor is the result of a last single-channeled convolutional  
 1948 layer with no activation function. In this way we let the model predict unbounded tensors to  
 1949 account for the phase symmetries.



**Figure 6.4: U-Net like architecture of the 2D model for phase retrieval.** The log-scaled, normalized 2D diffraction pattern is first passed through the encoder, composed of six convolutional blocks (green cubes) in each of which the feature map is filtered by a single stride convolutional layer with a  $3 \times 3$  pixels kernel. The number of filters, shown in bold inside each block, scales from 16 to 512. A LeakyReLU (alpha = 0.2) activates the output of each convolutional layer. A MaxPooling layer halves the size of the feature map down to 1D. The bottleneck, built with another convolutional block with 1024 filters, passes the feature map to the decoder. This last, mirroring the encoder, is composed of an equivalent number of de-convolutional blocks (orange cubes) in each of which the feature map is filtered and up-sampled by a two-strides de-convolutional layer with a  $3 \times 3$  pixels kernel and same LeakyReLU activation. Skip connections are employed as well between encoder and decoder. Copies of the feature map after each convolutional block are concatenated with same size feature maps in the decoder before transposed convolutions. A last convolutional layer with no activation function produces the RSP prediction. The total number of trainable parameters is of the order of 6.8 million.

## 1950 6.4.2 Loss function

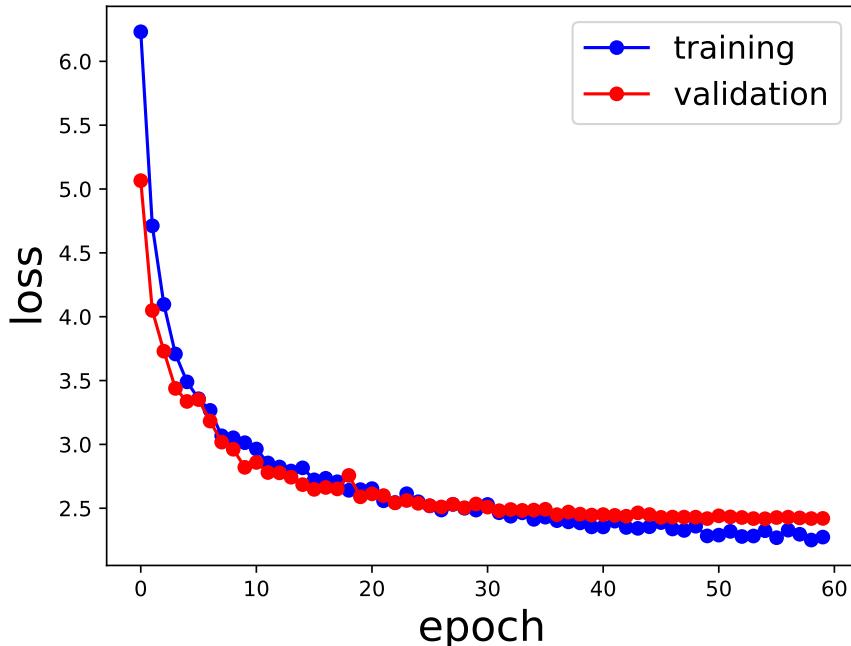
1951 The choice of the loss function was first based on what was used in literature. A sum of the MSE  
 1952 computed on the objects' amplitudes and one on the phases has thus been used (Eq. 6.2). The  
 1953 ground truth objects were indeed available from the simulated data while the predicted objects  
 1954 have been first calculated with a 2D inverse Fourier transform from the diffracted amplitude  
 1955 and the predicted RSP (Eq. 6.1).

$$\hat{O}(\mathbf{r}) = \mathcal{F}^{-1}\{\sqrt{I(\mathbf{q})} e^{i\varphi_{\text{pred}}(\mathbf{q})}\}(\mathbf{r}) , \quad (6.1)$$

$$\mathcal{L} = \frac{1}{N} \sum_{\mathbf{r}} \left( |\hat{O}(\mathbf{r})| - |O(\mathbf{r})| \right)^2 + \frac{1}{N} \sum_{\mathbf{r}} \left( \phi(\mathbf{r}) - \phi_{\text{gt}}(\mathbf{r}) \right)^2 , \quad (6.2)$$

1956 **6.4.3 Results**

1957 The training of the model was conducted on 8500 simulated BCDI patterns over 30 epochs with  
 1958 a learning rate of 0.0003 and monitored both training and validation loss. Here, Fig.6.5 shows  
 1959 the model's loss during the 30 epoch long training.



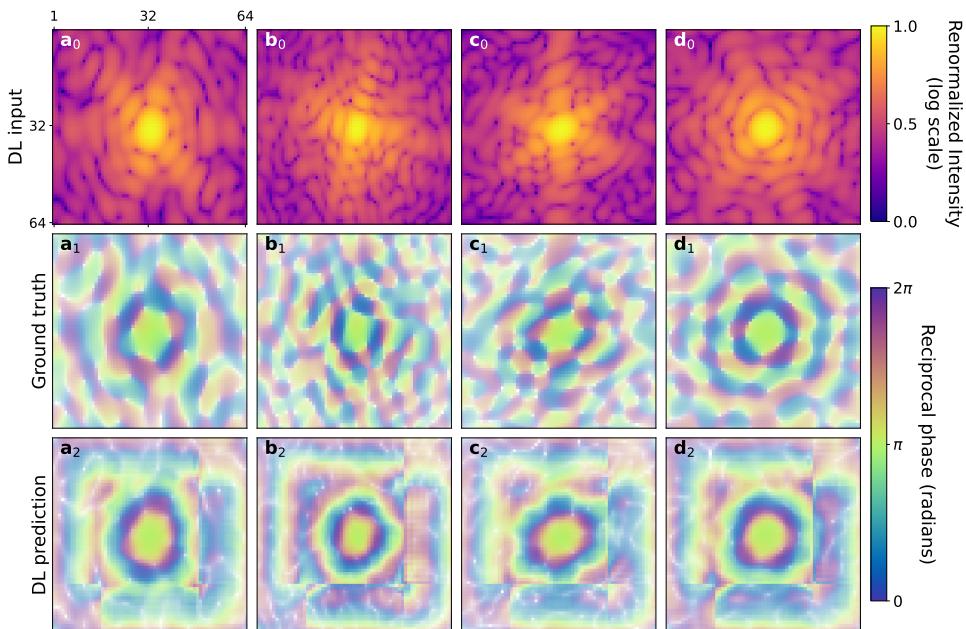
**Figure 6.5:** Training and validation loss over 30 epochs. The curve suggests a proper learning with no overfitting as both losses are decreasing reaching a plateau and the validation loss follows the same trend of the training loss.

1960 Fig.6.6 illustrates the results of the predicted RSP of some test simulated BCDI patterns. Note  
 1961 that the displayed predicted RSP has been wrapped between 0 and  $2\pi$  for better comparison  
 1962 with the ground truth but the raw output of the model is in fact an “unwrapped” array. This is  
 1963 expected since no activation layer was applied to the last convolutional layer, meaning that  
 1964 the last operation is the multiplication of the last feature map with the real values inside the  
 1965 convolutional kernel, hence linear.

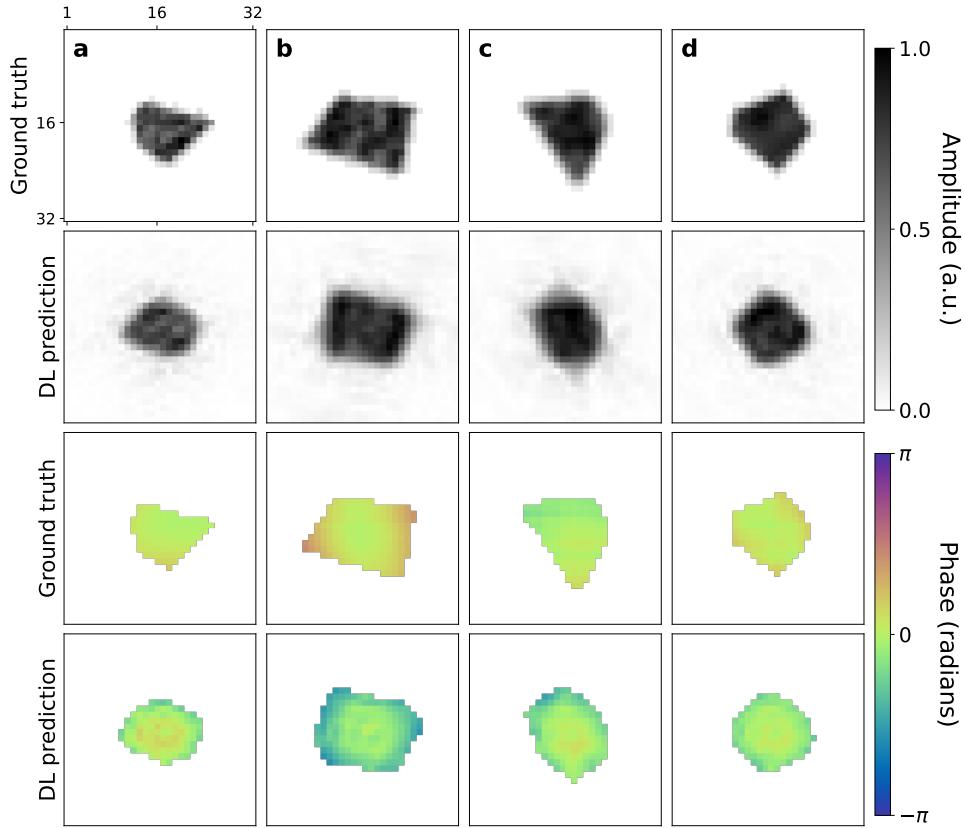
1966 When comparing the reconstructed objects obtained from the predicted RSP with the ground  
 1967 truth ones (Fig. 6.7) one can draw some interesting conclusions about the model’s learning  
 1968 performance. First it can be observed that the model learns the approximate shape and size of  
 1969 the particle, it produces images that resemble reasonable particles, sometimes similar to the  
 1970 ground truth. The amplitude is concentrated inside the support with little noise outside and  
 1971 the phase is overall correct around zero.

1972  
 1973 However, when looking more carefully, it is clear that the shape is not quite correct,  
 1974 especially for highly non-centrosymmetric objects. For instance, if we consider the object in  
 1975 Fig. 6.7 c, we see that the predicted shape seems to be derived from the incorrect superposition  
 1976 of the correct shape and its twin, also a correct solution since an object and its conjugate

symmetric have the same Fourier transform amplitude. More generally, it seems that the model tends to predict centrosymmetric objects. According to Sicairos *et al.* [91], if we name  $\varphi(\vec{q})$  the correct RSP, this phenomenon arises from a predicted RSP phase  $\phi$  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.* [185] and Zhang and coauthors in [188]. 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.

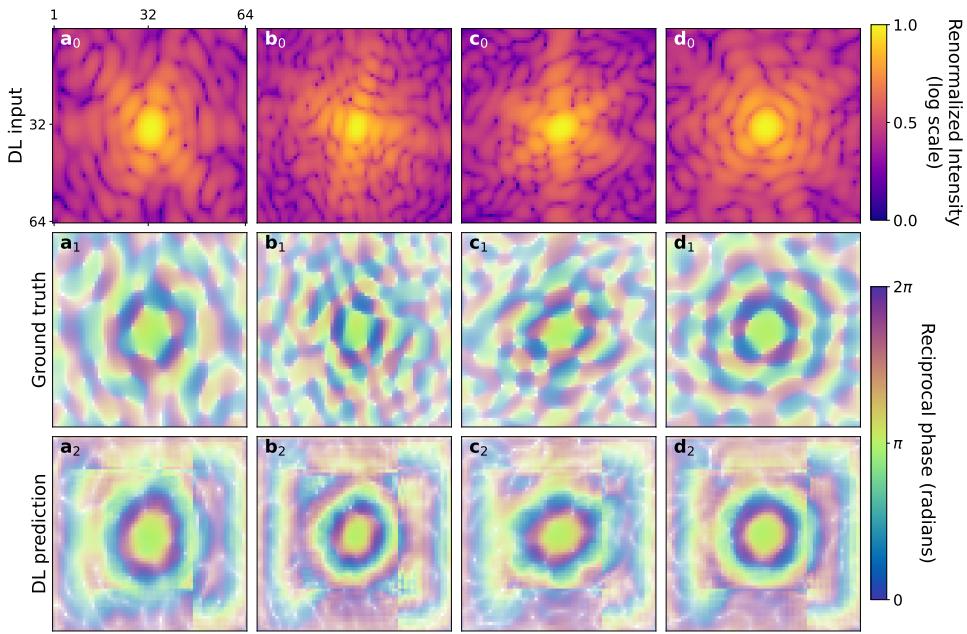


**Figure 6.6: Model testing on new 2D data using MSE loss function.** First row shows four simulated BCDI patterns, second row the ground truth RSP corresponding to the pattern and last row the DL prediction. The superposition of the two twin solutions resulting in the object space can be inferred here 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.

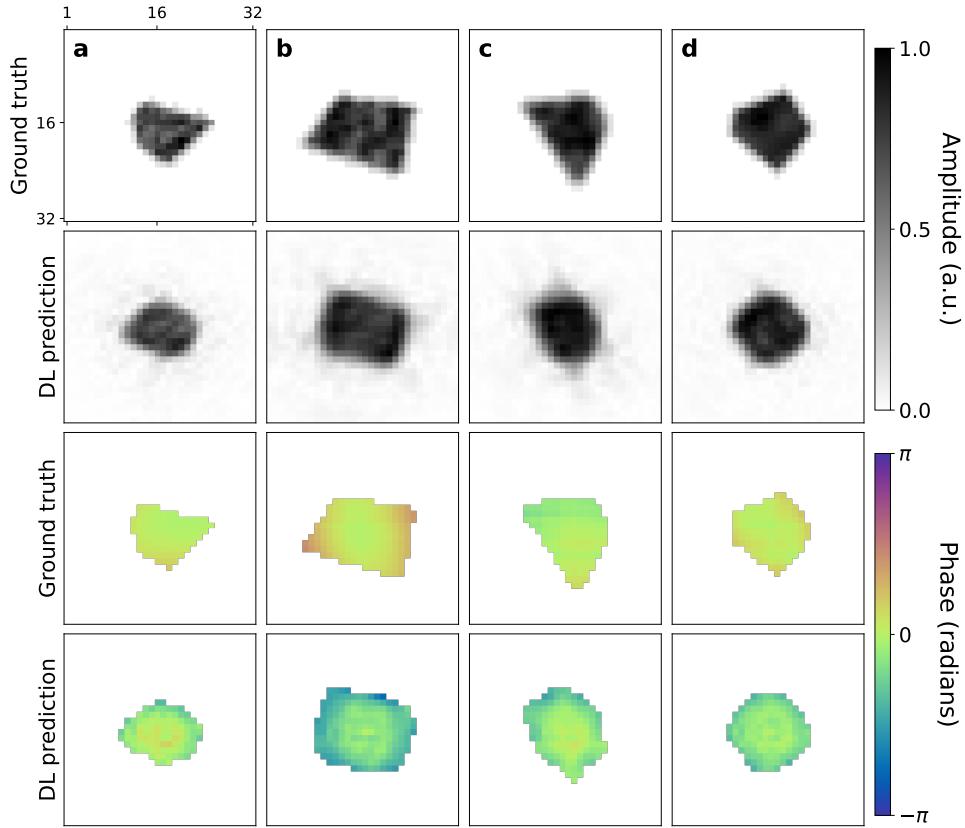


**Figure 6.7: Corresponding reconstructed objects.** Ground truth and predicted objects' amplitudes (first two rows respectively) and ground truth and predicted objects' phases (first two rows respectively). Note that the predicted objects resemble the superposition of the two twin true solutions.

1987      The procedure presented in the article for the removal of the phase symmetries consists in:  
 1988    (i) the centering of all the objects in real space (phase ramp removal), (ii) the shift of the RSP  
 1989    such that the  $k$ -th pixel for which  $\varphi_k = 0$  is the same across the dataset (phase offset removal),  
 1990    and lastly, (iii) the flip of the sign of the whole RSP when its value corresponding to a fixed  
 1991    position across the dataset is negative (if  $\varphi_{k+1} < 0$  then  $\varphi = -\varphi$ ). In our case the phase ramp  
 1992    symmetry was already broken by simulating particles with the center of mass in the center of  
 1993    the array. In this way the model is already biased towards the prediction of RSPs that yield  
 1994    centered objects. For the offset and the sign, the method proposed by Zhang *et al.* has been  
 1995    implemented in the model and the results are shown in Fig. 6.8 for the RSP and Fig. 6.9 for the  
 1996    reconstructed objects.



**Figure 6.8: Model testing using MSE loss function and biased dataset.** Here, the training dataset was biased with the method proposed in [188] in order to break the RSP sign symmetry. 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. 6.6.



**Figure 6.9: 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.

1997        Unfortunately, the proposed method did not seem to solve the sign ambiguity of the RSP.  
 1998        The model is still unable to discriminate between the plus/minus sign of the RSP and the result  
 1999        is the incorrect overlap of the object with its twin obtained by the inversion symmetry. The  
 2000        phase, though small for this case, is also showing a kind of centro-symmetry as its variations  
 2001        tend to spread radially from the center of the array.

#### 2002        6.4.4 The Weighted Coherent Average loss function

2003        At this point in the study, and in anticipation of applying the model to portions of the RSP,  
 2004        it became necessary to consider a loss function that would operate directly on the phase,  
 2005        without requiring transformations into direct-space. However, the main challenges were posed  
 2006        by the symmetries inherent to the phase. Upon further reflection, it was concluded that a  
 2007        Mean Squared Error (MSE) is not an appropriate metric for comparing the phases of complex  
 2008        functions. Indeed, MSE fails to account for the  $2\pi$  periodicity and the possibility of a global  
 2009        phase offset. One could argue that  $2\pi$  wraps can be fixed with a modulo  $2\pi$  operation and the  
 2010        offset can be removed by shifting the tensor by a constant. However, the modulo wrapping  
 2011        function jumps abruptly by  $2\pi$  every time the phase crosses an integer multiple of  $2\pi$ , meaning  
 2012        that the gradients are infinite thus not advised for gradient-based optimizations. Moreover, the  
 2013        MSE (or MAE and other *divergent* metrics) will have problems at the  $0-2\pi$  boundary. In fact,  
 2014        when considering the phase mapped in the  $0-2\pi$  range, if we suppose a  $\varphi_{pred}^0 = -0.1$  where

2015     $\varphi_{G.T.}^0 = 0$ , the wrap will move the  $\varphi_{pred}^0$  to the value  $2\pi - 0.1 = 6.183$  amplifying the error (2016     $\Delta\varphi$ ) from  $0.1^2$  to  $6.183^2$  improperly.

2017

2018    In order to bypass these shortcomings a new loss function was designed. Here it follows  
2019    the reasoning process that leads to the mathematical expression of the loss.

2020    The best way to account for the periodicity and the wrap, without discontinuities and error  
2021    unbalances, is to evaluate the ground truth - predicted phase differences ( $\Delta\varphi_k$ ) on the unit  
2022    circle. To do such, it is necessary to express  $\Delta\varphi_k$  as angles of a complex exponential. This  
2023    means that if  $\varphi_{pred}$  is an array of random values, each complex number  $z_k = e^{i\Delta\varphi_k}$ , when  
2024    represented on the Argand plane, can be seen as a vector pointing at a random coordinate on  
2025    the unit circle (Fig. 6.10c). Now, the goal of the optimization is not to minimize  $\Delta\varphi_k$  for all  $k$   
2026    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  
2027    same direction, but it does not necessarily lie on the x-axis ( $\Delta\varphi_k = 0$  condition). Therefore,  
2028    the loss function should ultimately drive all the  $z_k$  from randomly distributed to coherently  
2029    aligned along a common direction.

2030    A helpful quantity in this case can be the complex average vector  $\langle z \rangle = \frac{1}{N} \sum_{k=1}^N z_k =$   
2031     $\frac{1}{N} \sum_{k=1}^N e^{i\Delta\varphi_k}$  where  $k$  runs over all the  $N$  pixels. In particular the length of  $\langle z \rangle$ , represented  
2032    by the modulus  $|\langle z \rangle|$ , is an efficient metric for the measurement of the *degree of alignment* of all  
2033    the  $z_k$  between each other. In other words it measures the “coherence” between  $\varphi_{GT}$  and  $\varphi_{pred}$ .

2034    In fact,  $|\langle z \rangle|$  scores 0 for randomly oriented  $z_k$ , as opposite contributions cancel out each  
2035    other because incoherent, while it scores 1 for perfectly aligned ones. It follows that one wants  
2036    to maximize  $|\langle z \rangle|$  during the optimization. Moreover, given the natural normalization between  
2037    0 and 1 of this metric, it follows naturally that the loss function can be expressed as  $L = 1 - |\langle z \rangle|$ .

2038

2039    Additionally, an importance mask can be applied during the averaging process. In particular,  
2040    we know that the brightest pixels of the BCDI pattern are the ones contributing the most to the  
2041    object’s reconstruction. For this reason one could weight the complex average by multiplying  
2042    by the input magnitudes. The effect of this operation is to “give a direction” to the optimization,  
2043    meaning that the  $\langle \Delta\varphi \rangle$  the model will tend to converge to, will be mostly steered close to the  
2044     $\Delta\varphi_k$  of the brightest  $k$  pixels. The loss can now be expressed as:

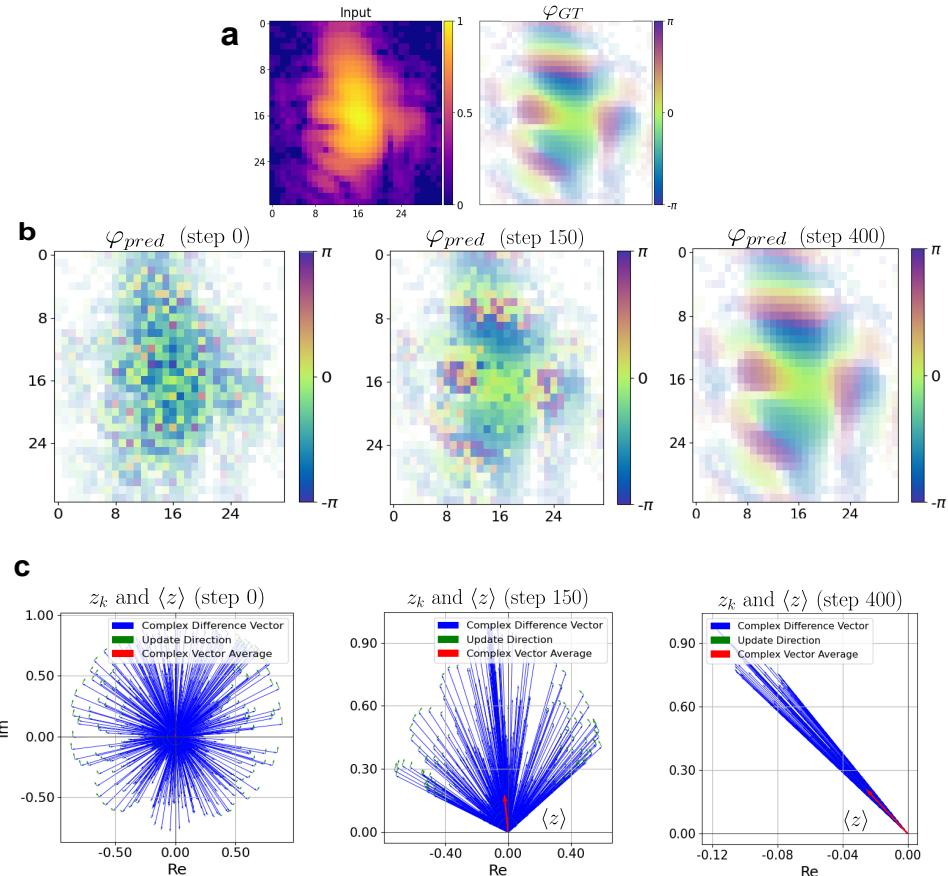
$$L = 1 - \left| \frac{1}{N} \sum_{k=1}^N \sqrt{I_k} \exp(i(\varphi_{GT,k} - \varphi_{pred,k})) \right| \quad (6.3)$$

2045    Where  $N$  is the total number of pixels in each RSP array and  $k$  is the pixel index.  $\sqrt{I}$  is the  
2046    magnitude of the BCDI pattern normalized between 0 and 1 with respect to the sum, and  $\varphi_{GT}$   
2047    and  $\varphi_{pred}$  the ground truth and predicted RSP.

2048    The last missing piece is the removal of sign symmetry. Rather than biasing the dataset  
2049    preferring one sign over the opposite, the function  $L$  is computed for both  $\varphi_{GT}$  and  $-\varphi_{GT}$   
2050    and in a second passage, the minimum of the two along the batch dimension is kept for  
2051    backpropagation. The final form of the Weighted Coherent Average (WCA) loss is then given  
2052    by:

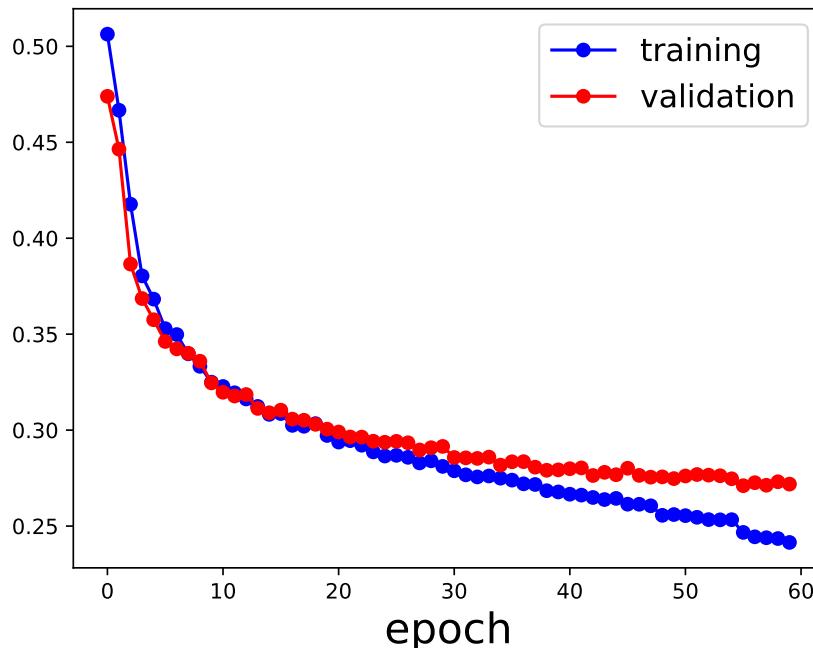
$$L_{WCA} = \min(L_+, L_-) \quad (6.4)$$

2053 To better visualize the functioning of the WCA loss function, a simple model has been  
 2054 trained to fit the ground truth phase of a single 2D BCDI pattern using the WCA. The complex  
 2055 phase differences vectors were extracted at each step of the optimization together with the  
 2056 updates obtained from the gradients of the WCA with respect to the trainable parameters. Fig.  
 2057 6.10 shows the evolution of the predicted RSP as well as the progressive alignment of the  $z_k$ .



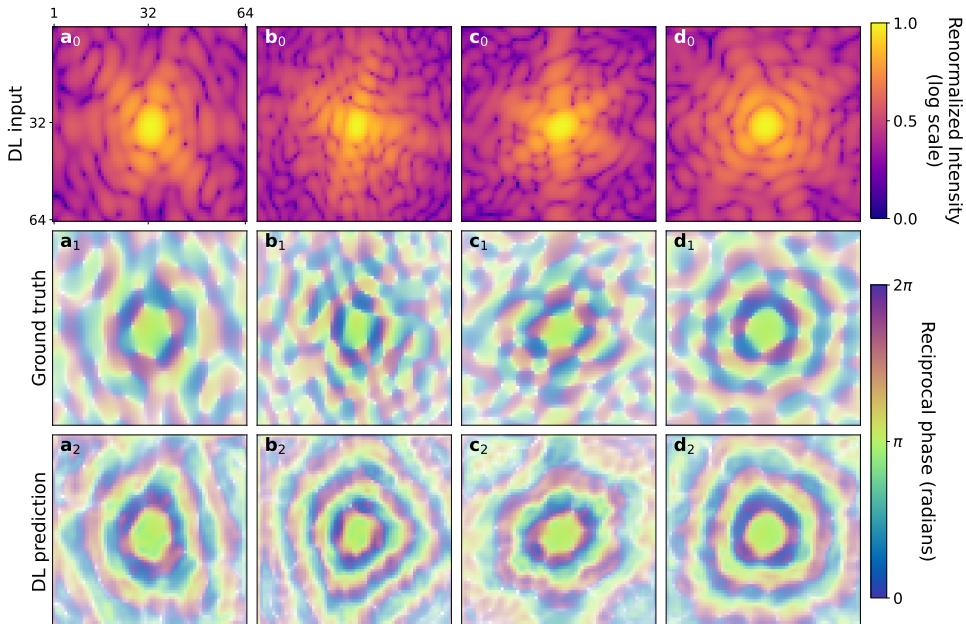
**Figure 6.10: Illustration of the WCA loss function.** **a** Input intensity (log-scale normalized) and ground truth RSP. **b** Predicted RSP in steps 0 - 150 - 400 of the optimization. **c** Corresponding complex phase-differences vectors  $z_k$  on the Argand plane (blue arrows), together with the 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. Notice in **c** the vectors align around a  $\Delta\varphi \neq 0$  resulting in a  $\varphi_{pred}$  shifted by a constant offset. The offset has been removed before plotting in **b** for better comparison with the ground truth.

2058 The same model has been trained using the WCA for the same number of epochs on the  
 2059 same dataset and here the results are shown. First, it can be noted in Fig.6.11 that the training  
 2060 and validation loss values throughout the training are following different trends with respect  
 2061 to the model trained with the MSE loss (Fig. 6.5)

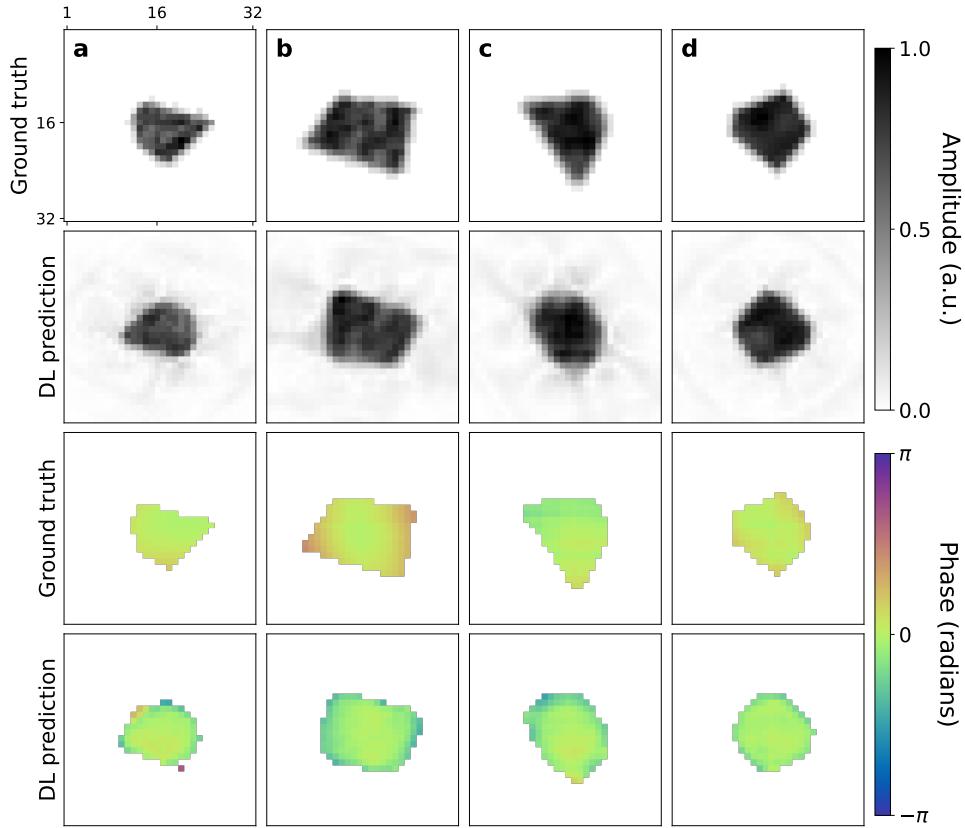


**Figure 6.11:** Training and validation loss curves over 60 epochs using the WCA loss function.

2062 In this case the correct learning curve does not reach a plateau within the first 25 epochs  
2063 but maintains a negative slope for longer, indicating a better learning. This suggests indeed  
2064 better results when used on test data. In particular, for the same input diffraction patterns  
2065 tested above in Figs.6.8 - 6.9 the model trained with the WCA yields the prediction shown in  
2066 Fig.6.12 for the RSP and Fig.6.13 for the corresponding reconstructed objects.



**Figure 6.12: 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. Here the squared artifacts disappeared, suggesting a better learning.

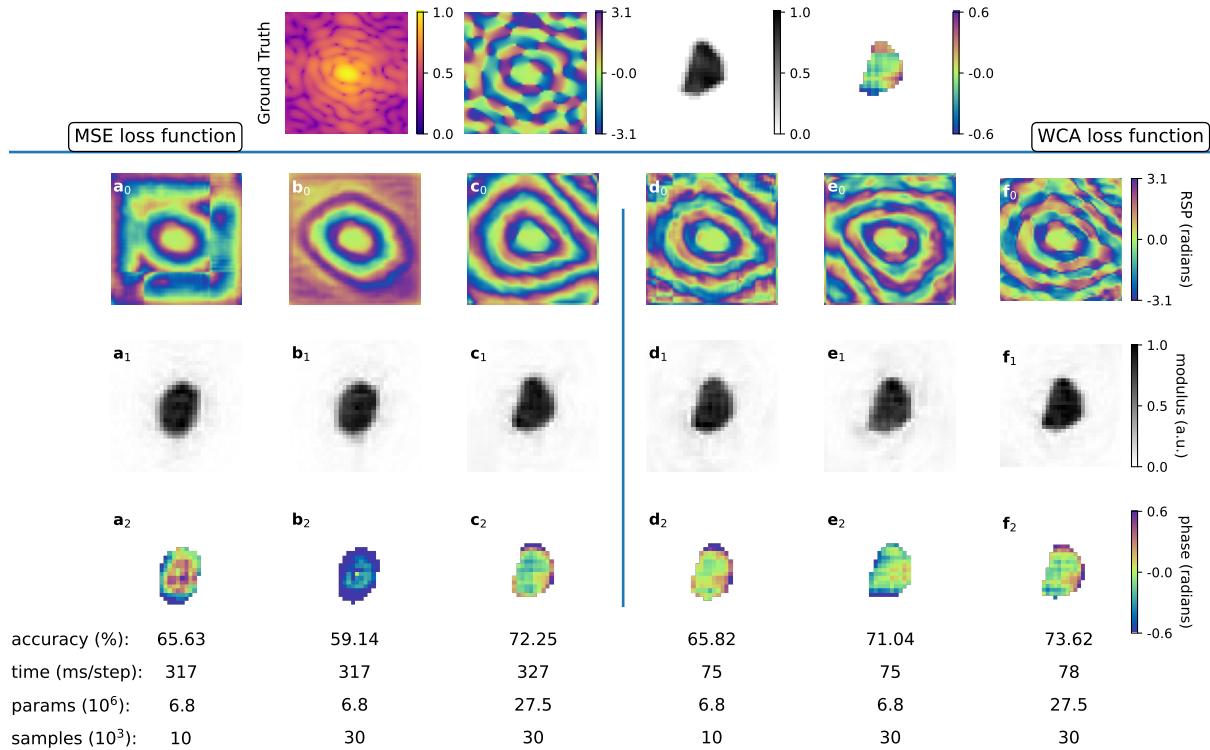


**Figure 6.13: Corresponding reconstructed objects.** Ground truth and predicted objects' amplitudes (first two rows respectively) and ground truth and predicted objects' phases (first two rows respectively). Here the twin problem mentioned above and visible in Figs.6.7 - 6.9 is less pronounced, the model seems to manage the two solutions.

2067     The results obtained from the model trained with the WCA loss function are visually better  
 2068     than the MSE ones. Although not completely removed, the sign symmetry that gives rise to  
 2069     the superposition of the object with its twin, is less pronounced. For example, particles in Fig.  
 2070     6.13(a-b-d) have a clear orientation and a shape that matches the ground truth. In all those  
 2071     cases though, the model has opted for the conjugate solution as the predicted object are flipped  
 2072     with respect to the ground truth ones. In Fig. 6.13(c) instead the symmetry is not broken and  
 2073     the result is still a superposition of the particle with its twin. This suggests that the symmetry  
 2074     breaking method implemented in the WCA, and the one proposed by Zhang and coauthors, is  
 2075     only partially playing a role in the actual model learning. It is interesting to notice indeed that  
 2076     when the training dataset or the model trainable parameters are increased, the sign symmetry  
 2077     is completely removed in the most difficult cases as well. Fig. 6.14 shows the effect of the  
 2078     dataset and models sizes for both MSE and WCA loss functions on the same simulated test  
 2079     data. The first important piece of information this figure shows is that the model trained with  
 2080     the WCA reaches higher accuracy. Moreover, it is much faster to compute since no FFT or  
 2081     IFFT is involved (see Fig. 6.14), thus the training time is drastically reduced. For what concerns  
 2082     the accuracy metric, in order to properly account for both modulus and phase, it has been  
 2083     calculated using

$$\left( \frac{PCC(\rho_{pred}, \rho_{GT}) + WCA(\rho_{pred}, \phi_{pred}, \phi_{GT})}{2} \right) \times 100 \quad (6.5)$$

where  $PCC(\rho_{pred}, \rho_{GT})$  is the Pearson Correlation Coefficient on the object's moduli and  $WCA(\rho_{pred}, \phi_{pred}, \phi_{GT})$  is the WCA function applied to the object's phase weighted by the normalized predicted modulus (Eq.6.3-6.4). 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. Finally, 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.

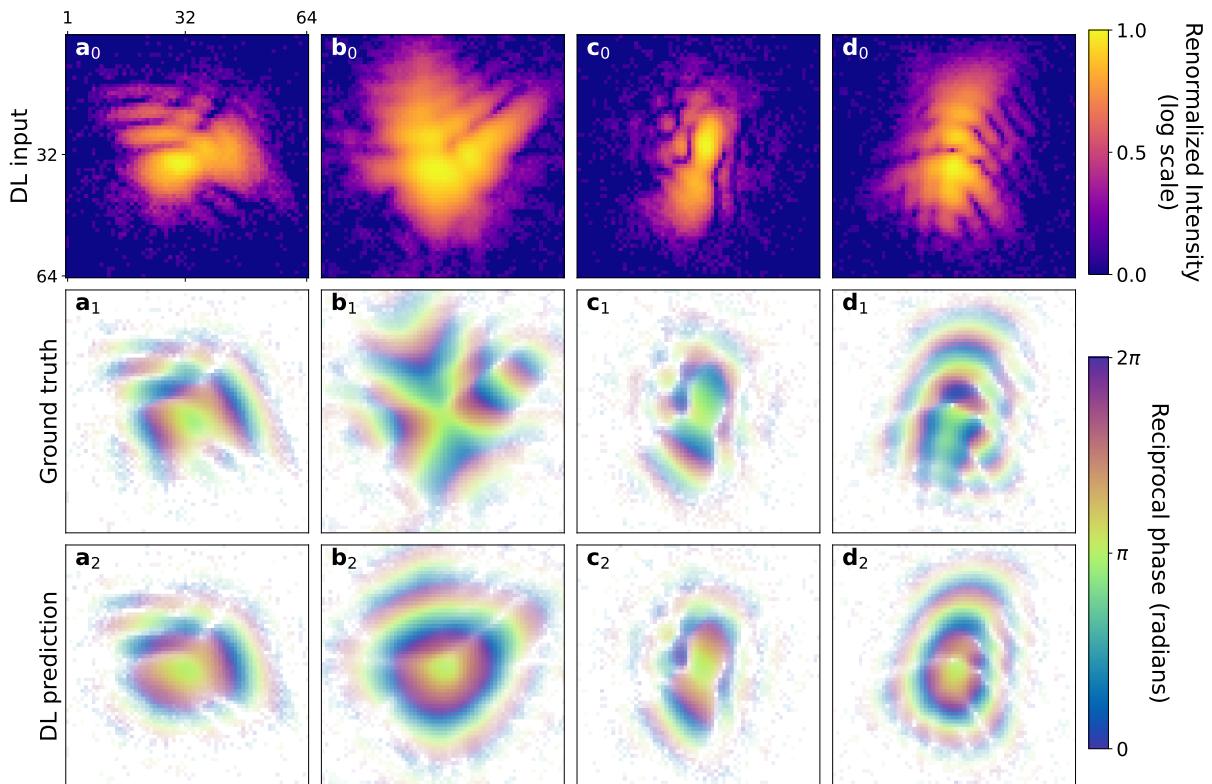


**Figure 6.14: Comparison of MSE and WCA loss function for different model and training dataset sizes** In the first row from left to right the input intensity, the ground truth RSP and the corresponding object (modulus and phase) are represented.  $a_0, b_0, c_0$  are the results of the predicted RSP 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. The WCA yields better results and requires shorter trainings.

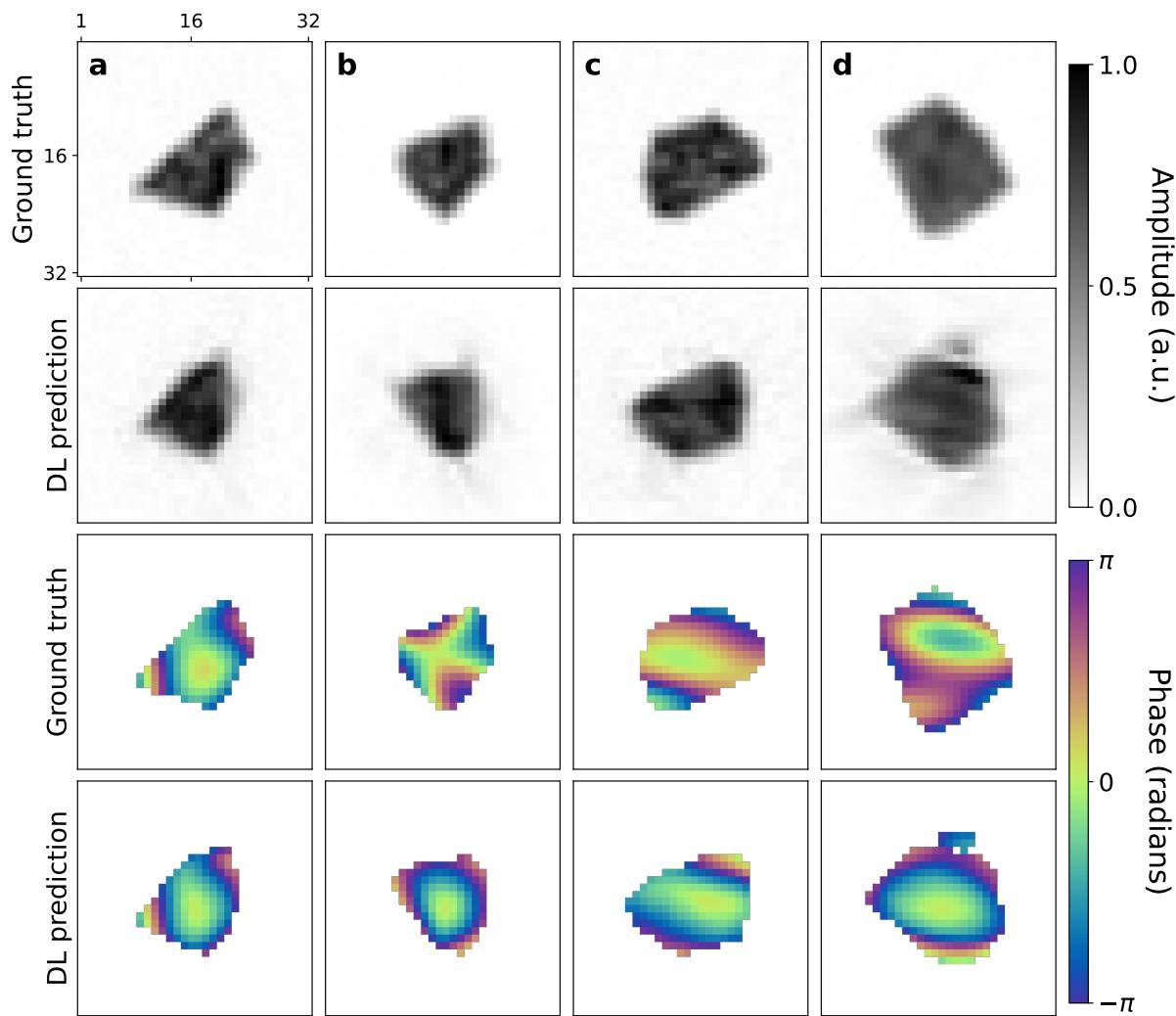
## 2091 6.5 2D high strain case

In this paragraph the model training was performed on a dataset of highly strained 2D BCDI pattern simulated as described above in section 5.3.1. In this case Poisson statistics 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 6.14 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

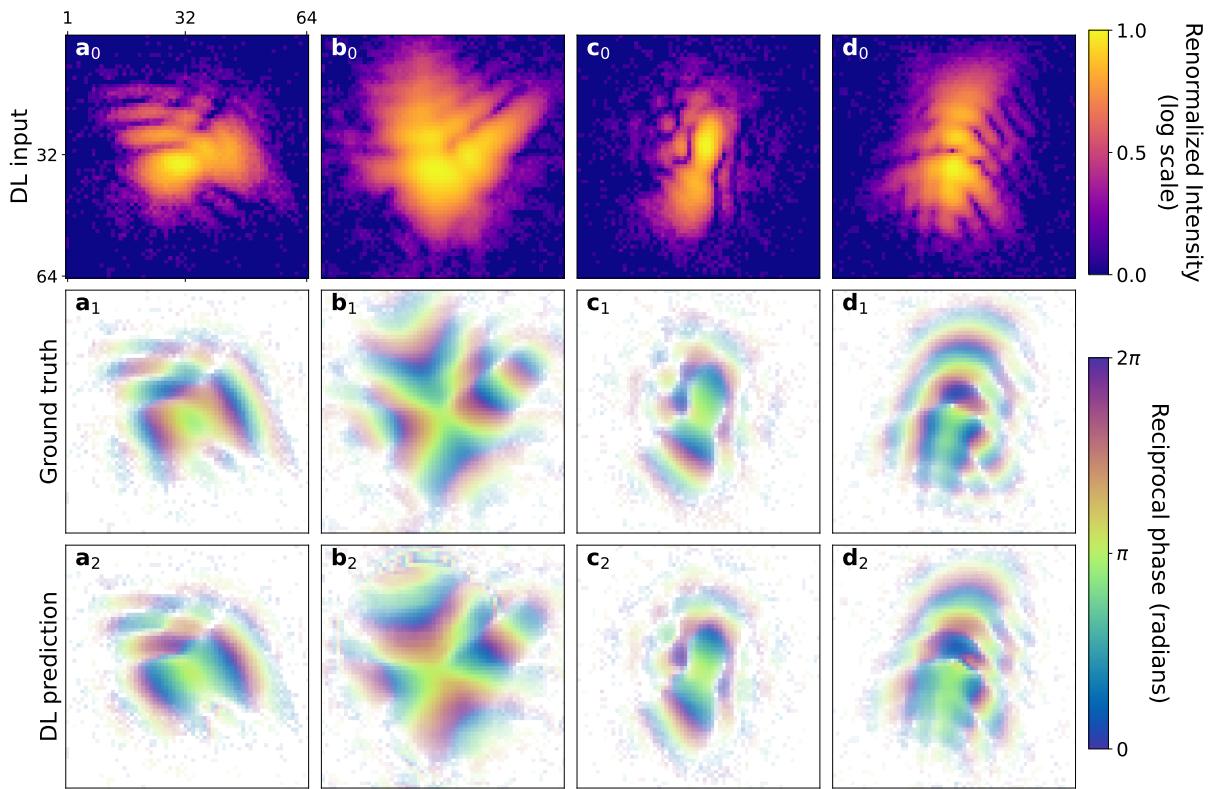
2099 of the model when the complexity of the task increases. The results on 4 test BCDI pattern are  
 2100 shown in Figs. 6.15-6.16 for the MSE one and 6.18-6.18 for the WCA.



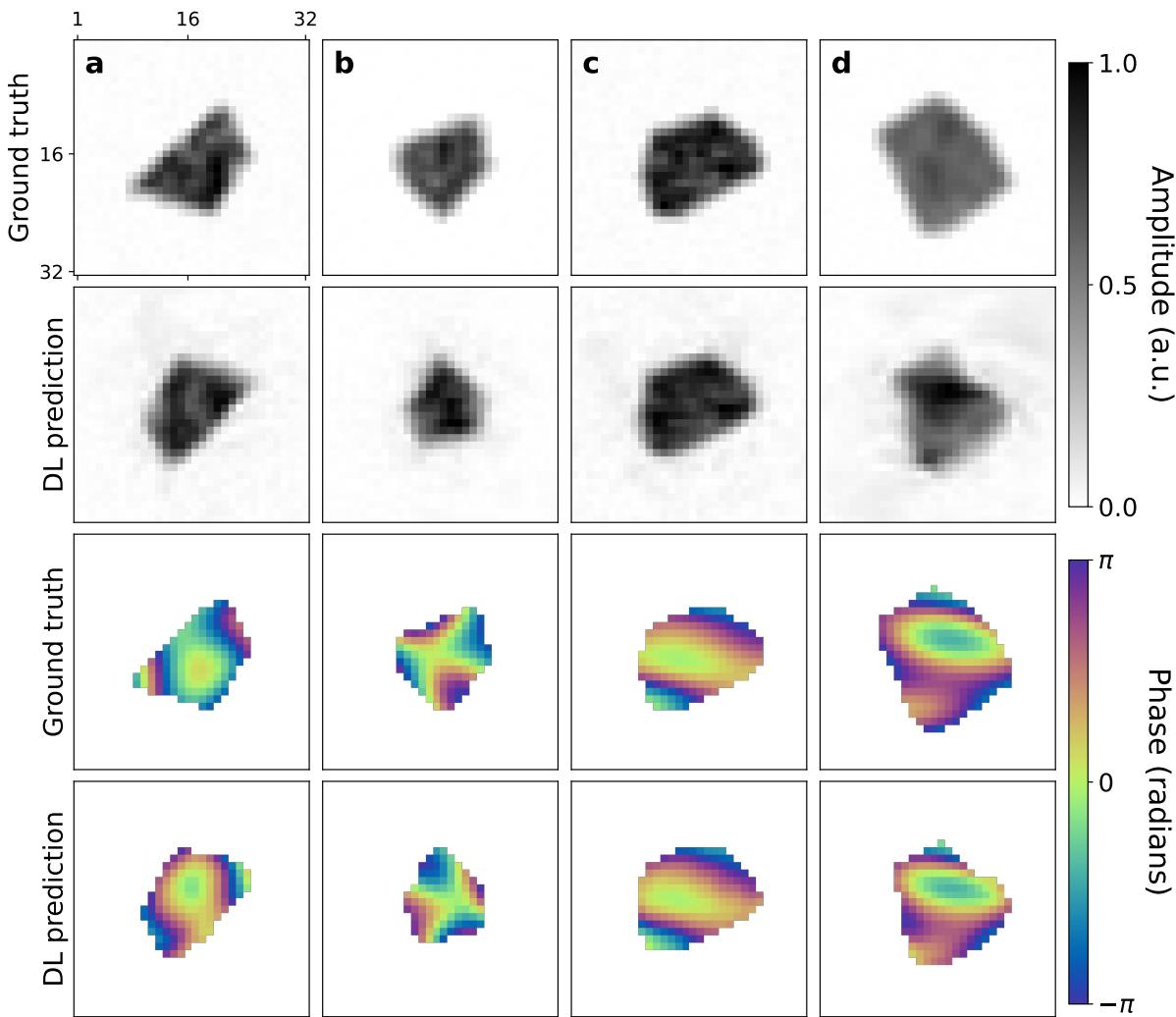
**Figure 6.15:** RSP predicted by the model trained with the MSE loss function calculated on both modulus and phase of the reconstructed object. First row: simulated 2D strained BCDI patterns (test dataset). Second row: corresponding ground truth RSP. Third row: predicted RSP. Once can notice that the model cannot predict correctly the RSP where the “iso-phases” do not have a circular symmetry (see **b-d**).



**Figure 6.16:** Reconstructed objects corresponding to Fig 6.15. 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 6.17:** 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 6.18:** Reconstructed objects corresponding to Fig 6.17. Compared to the objects obtained from the DL model trained with the MSE loss function (Fig.6.16) here the objects' phases are correctly recovered, also when the symmetry is not circular (**b**).

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 patches of the reciprocal space.

## 6.6 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 patterns 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 a unique mapping exists,

2113 barring some trivial RSP symmetries, between the diffracted intensity and the RSP in 3D [189].  
2114 What is interesting to investigate is whether this relationship exists also for subsets of the  
2115 reciprocal space, and in particular if it can be retrieved by a DL model. Moreover, the use of  
2116 patches is further motivated by the local behavior of the phase, which typically oscillates in an  
2117 approximately linear fashion from 0 to  $2\pi$ , mirroring the intensity fringes. This local periodicity  
2118 implies a degree of spatial consistency and tractability that justifies the use of localized patches  
2119 for learning and reconstruction. (From now on the term “patches” will be used to refer to cubic  
2120 subsets of the reciprocal space).

2121

2122 When deciding to work with patches, there is a number of questions that arise and the  
2123 answer to which is not straightforward nor unique in many cases. Namely:

- 2124 • What size is best?  
2125 • Can the patches be extracted at random positions or should there be an order?  
2126 • What about the normalization of the intensity range inside the patch?  
2127 • How are the patches stitched together into the full RSP eventually?  
2128 • How are the phase symmetries taken into account during the stitching?

2129 Here I will present the approach that allowed me to address these questions.

### 2130 6.6.1 The choice of the size

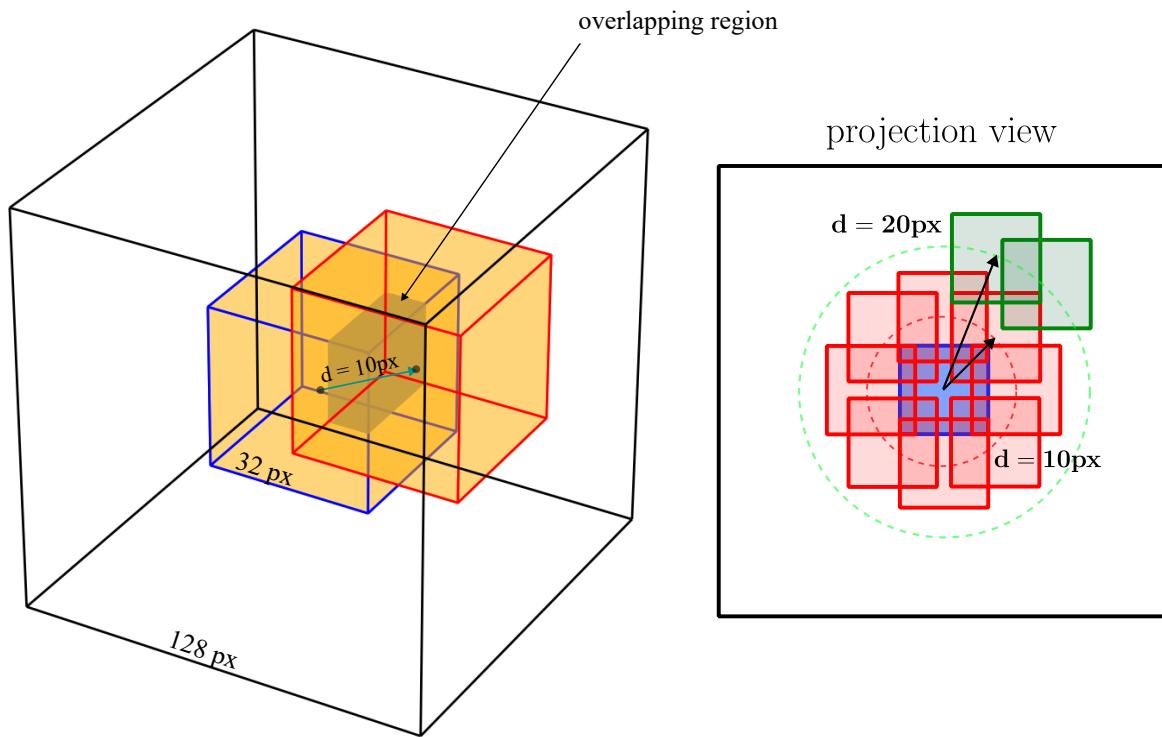
2131 Similarly to the inpainting case, 32 pixel-side cubic patches, cropped out of 128 pixel-side  
2132 simulated BCDI patterns were considered. The choice was supported by the following reasons:

- 2133 • The good results obtained for the inpainting case suggested that the amount of informa-  
2134 tion contained inside a 32 pixel-side patch of reciprocal space is enough for the model to  
2135 grasp spatial correlations.  
2136 • The average oversampling ratio of BCDI experimental data is such that in a 32 pixel-side  
2137 volume a sufficient amount of fringes is contained, meaning intuitively that the model  
2138 can predict the corresponding RSP.  
2139 • An even number multiple of 2 is usually considered GPU-friendly since it facilitates  
2140 shared calculations across different threads.

### 2141 6.6.2 Patches division and stitching

2142 At first, the patches were extracted randomly from the full BCDI pattern as for the inpainting  
2143 case. However, by doing so the RSP of each patch would in principle have different offsets and  
2144 different wraps than their neighbors and this would complicate the stitching of the patches  
2145 back into the full RSP. For this reason, and considering the approximate spherical symmetry

2146 of the average BCDI pattern it was decided to crop patches radially, starting from the region  
 2147 around the center of the Bragg peak and the progressively moving outwards to higher q-values.  
 2148 In this configuration, an integer step (10 pixels in our case) was chosen beforehand and the first  
 2149 patch around the center of the Bragg peak was selected together with all the patches centered  
 2150 in distances of integer multiples of the chosen step. Fig. 6.19 shows a simplified schematic of  
 2151 the patches' extraction. For the DL model training the patches of the intensity pattern need to  
 2152 be selected as well as the for the corresponding RSP for ground truth comparison.



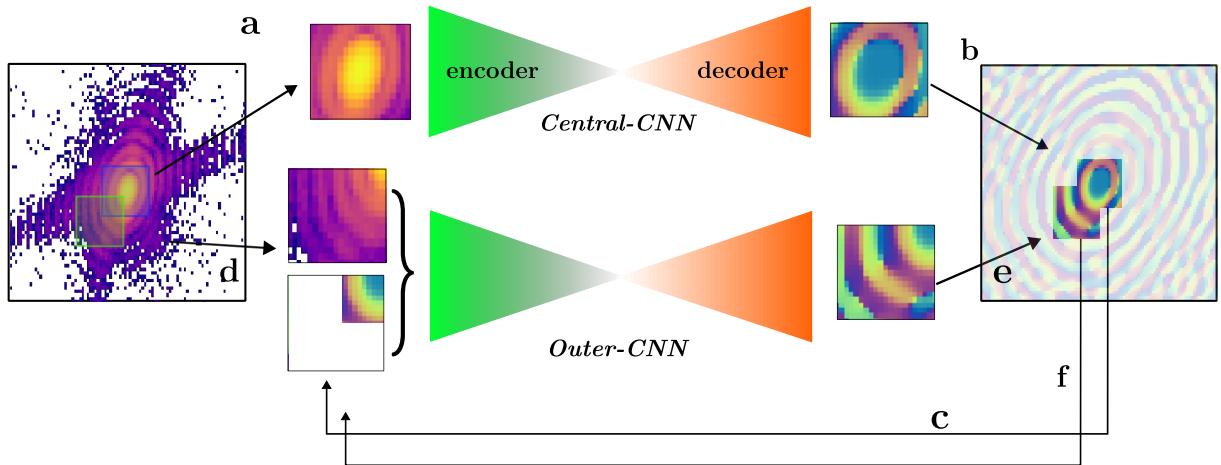
**Figure 6.19: Schematic of the cropping of patches (not in scale).** **(Left)** From the full BCDI pattern (white 128 px-side cube) the first patch is cropped out in the center and (orange 32 px-side cube with blue outline). Other patches are extracted radially from concentric shells separated by a 10 pixels distance. The gray shaded area highlights the overlapping volume between the two patches. **Right** The projection view shows the full first shell and two patches from the second shell for simplicity.

2153 Being the step size smaller than the semi-diagonal of the 32 pixel-sided patches, it follows  
 2154 that the patches of adjacent cells have overlapping volumes. These common regions can have a  
 2155 twofold purpose. Firstly, they reduce the complexity of the stitching procedure since when this  
 2156 is executed progressively starting from the central patch, the sign and the offsets of the RSP  
 2157 are unambiguously fixed for all the following ones. Secondly, during the DL model training,  
 2158 for patches belonging to the outer shells, the overlapping volume of RSP belonging to the  
 2159 innermost adjacent shell can be provided as initial guess along with the input intensity patches.  
 2160 This of course cannot be exploited for the central patch that necessarily has to be predicted  
 2161 without initial RSP guesses.

2162 The last question to be answered concerns the normalization. Since each patch is processed  
 2163 independently of the others by the DL model, it was decided to normalize each patch between

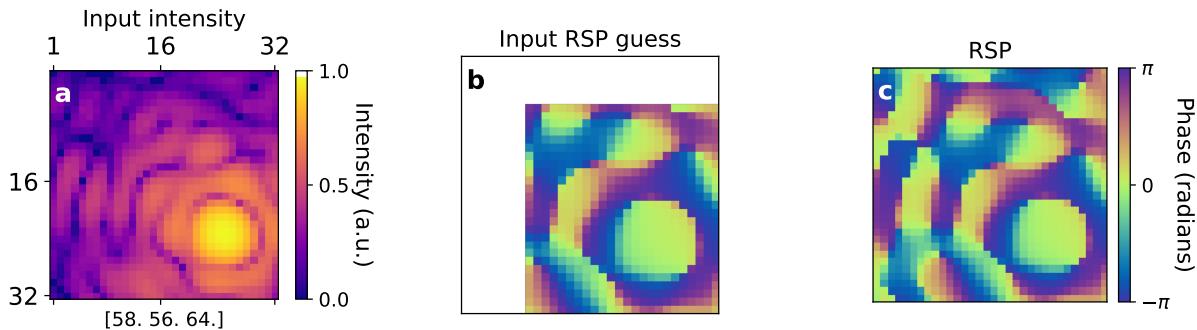
2164 0 and 1 (always in log scale).

2165 To summarize, the final design implied the use of two distinct training datasets and two  
 2166 different CNNs as depicted in Fig.6.20.



**Figure 6.20: Schematic of the patching model for phase retrieval.** The central patch of the BCDI pattern is cropped and sent through the *Central-CNN* (a) for the prediction of the corresponding RSP. During training the corresponding ground truth RSP is used to compute the WCA loss function and update the model's parameters. During inference predicted central RSP is placed in the center of an empty 3D array (b). The outer patches on the first shell at 10px distance from the center are extracted from the BCDI pattern and concatenated with the RSP patch which is non-zero only in those voxels overlapping with the innermost shell. During training this initial RSP guess is taken from the ground truth simulated RSP while during inference this is taken from the previous predictions (c - d). The *Outer-CNN* takes as input the outer intensity patch and the initial guess RSP and completes the RSP patch with the prediction inside the empty voxels. During inference, the output is stitched with the previous predictions (e) to form the full RSP. The process is repeated until the outermost shell (f).

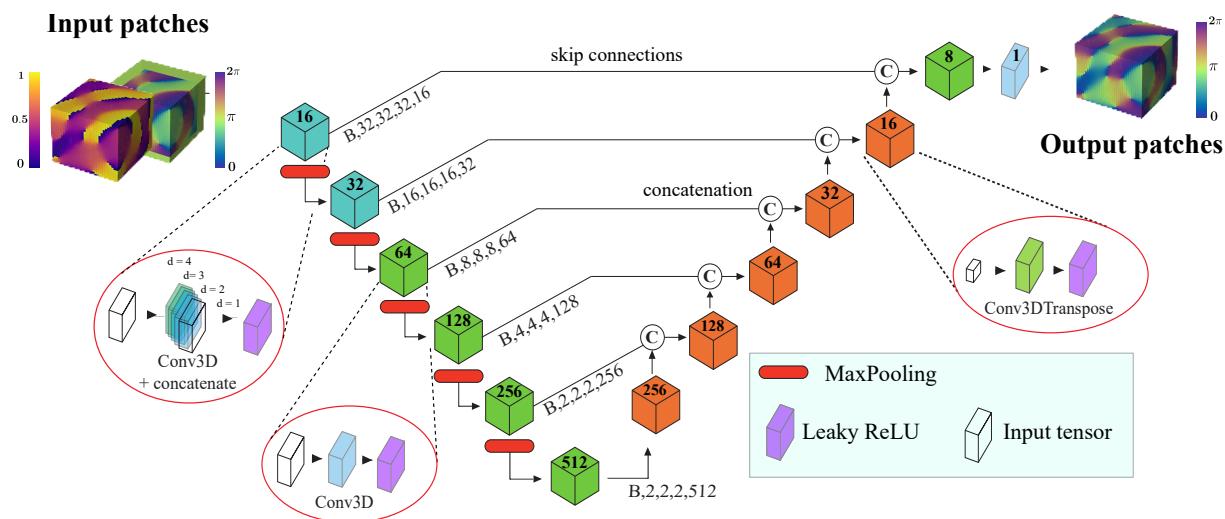
2167 The first dataset was dedicated to the central portion, therefore the first CNN (*Central-CNN*)  
 2168 was provided with 3D intensity patches in input (normalized log scale) and corresponding  
 2169 RSP patches as ground truth labels. A second training dataset containing patches from outer  
 2170 shells (5 concentric ones for a 128 pixel-sided full BCDI pattern) was created. Here each file  
 2171 was made of the pair intensity-RSP initial guess, from the closest neighbor patch belonging to  
 2172 the innermost shell, as input and the full RSP ground truth patch corresponding to the input  
 2173 intensity. This second dataset was used to train a second CNN (*Outer-CNN*) identical to the  
 2174 first one. One observation regarding the datasets is that there is an intrinsic imbalance between  
 2175 the number of central patches and the outer ones. In fact, for a single full BCDI pattern, the  
 2176 number of patches in the first shell is 1, while the number of outer portions can go up to several  
 2177 hundreds. Moreover, the central patch is the most important one as it contains a low resolution  
 2178 representation of the particle in real space. In order to balance the training, the first dataset  
 2179 was augmented with more simulated data.



**Figure 6.21:** 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**.

### 2180 6.6.3 Model architecture

2181 The model architecture is similar to the one used for the inpainting case, with a U-Net like  
 2182 structure (see Fig. 6.22). 5 encoder blocks reduce the feature map to 512 one-dimensional  
 2183 vectors in the bottleneck and 5 decoder blocks upsample the feature map back to the original  
 2184 size. Skip connections are used as well and similarly to the 2D case for Phase Retrieval, the last  
 2185 layer has been left with no activation function.



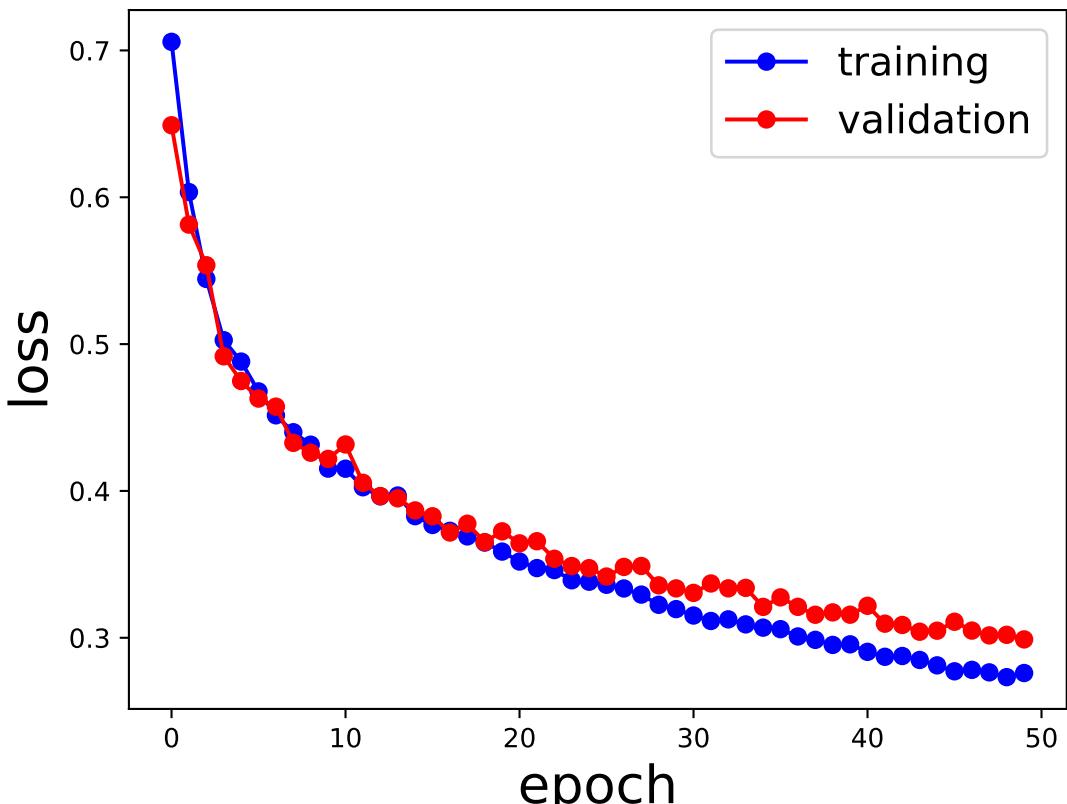
**Figure 6.22: Patching-model architecture.** Both *Central-CNN* and *Outer-CNN* share the typical U-Net like encoder-decoder structure for 3D images. Here only *Outer-CNN* is depicted. The description of each block can be found in the captions of Figs.5.10 - 6.4.

2186 The total number of trainable parameters is of the order of 3.5 million.  
 2187 For the second CNN the layers and features are identical to the first one but three modifications  
 2188 were made. Namely:

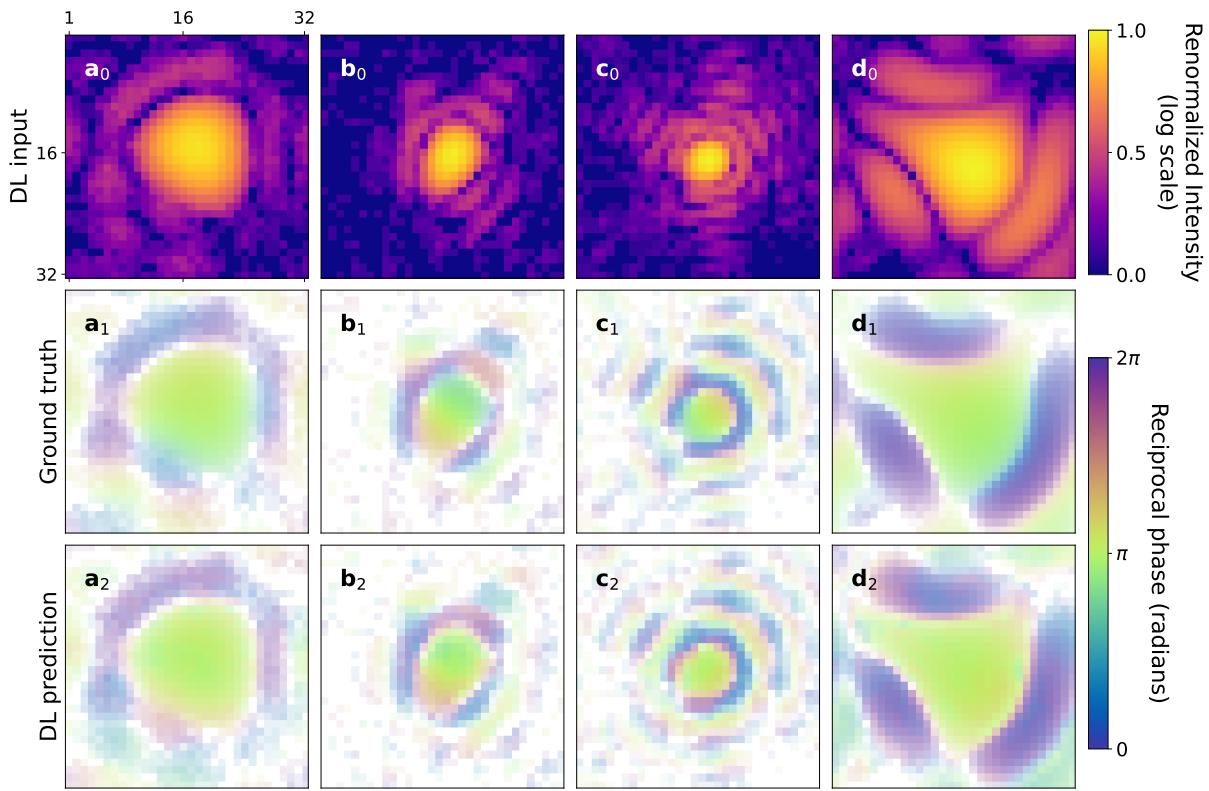
- 2189 • The input tensor was composed of the intensity patch concatenated with the initial RSP  
 2190 guess and a binary mask marking the RSP guess voxels from the others.
- 2191 • The mask was used at the exit of the decoder as well to select the new predicted voxels  
 2192 only for the back-propagation.
- 2193 • The WCA loss function has been restricted only to the positive sign of the RSP since no  
 2194 sign ambiguity is left when fixing an initial guess

2195 **6.6.4 Results on central patches**

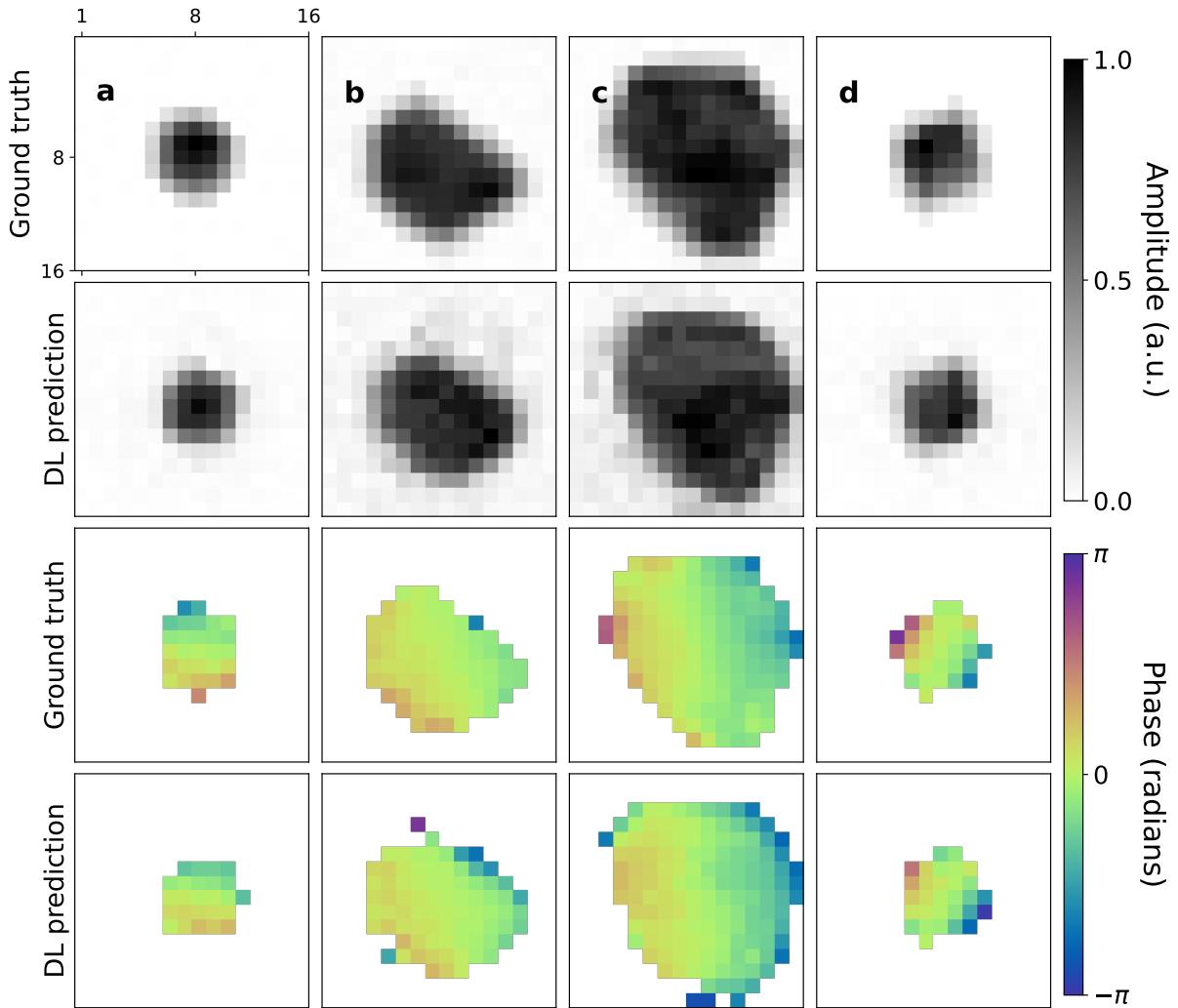
2196 *Central-CNN* has been trained over 50 epochs on 10000 samples of central  $32 \times 32 \times 32$  pixel-size  
 2197 patches cropped out of  $128 \times 128 \times 128$  pixel-size full BCDI simulated pattern using the WCA  
 2198 loss function. Fig.6.23 shows a good learning curve which is supported by the results on test  
 2199 data, illustrated in Figs. 6.24 - 6.25



**Figure 6.23:** 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 6.24:** 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  $\mathbf{b}_0$  and  $\mathbf{b}_1$  makes such that the central portion already contains the full useful signal and shows the diversity of the dataset. Row from  $\mathbf{a}_1$  to  $\mathbf{d}_1$  shows the corresponding ground truth RSP while the last row shows the RSP predicted by the DL model.



**Figure 6.25:** Corresponding objects from the central RSP patches in Fig.6.24. 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.

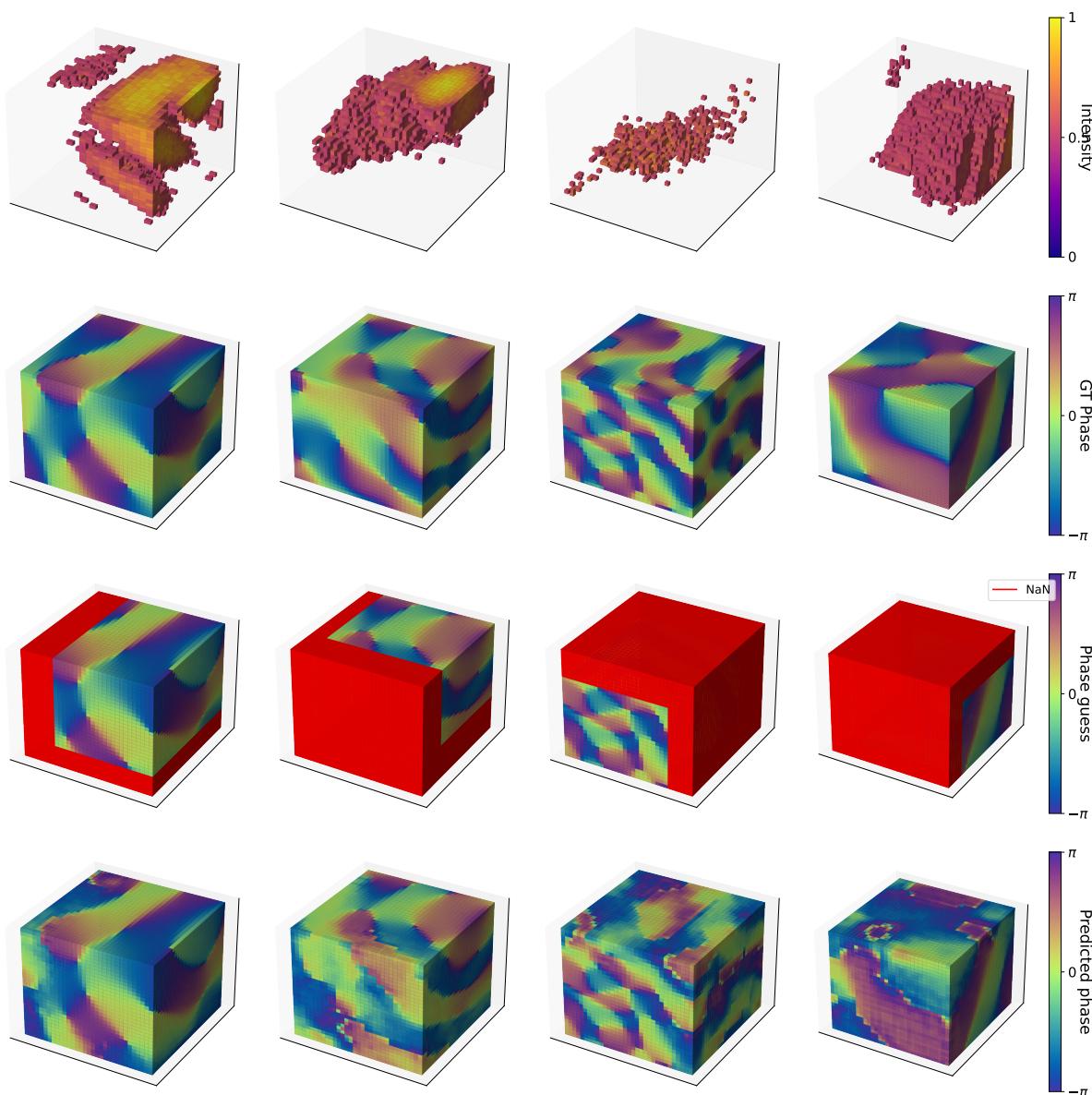
From the results obtained after the training of the model dedicated to the central portion of the simulated BCDI patterns, one can conclude that the model is capable of retrieving the correct RSP for the low strain case, meaning that the leap from the 2D case to the 3D case does not imply unforeseen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

2205

### 2206 6.6.5 Results on outer patches

2207 *Outer-CNN* was trained on patches extracted from outer shells over 50 epochs on a dataset  
 2208 containing 50000 samples. Fig. 6.26 illustrates some relevant results. In particular one can  
 2209 observe that the model can predict the RSP in the missing regions providing a relatively smooth  
 2210 transition between the “known” and “unknown” parts. This result is particularly interesting as

it proves that a CNN trained with the WCA loss function learns 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 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.



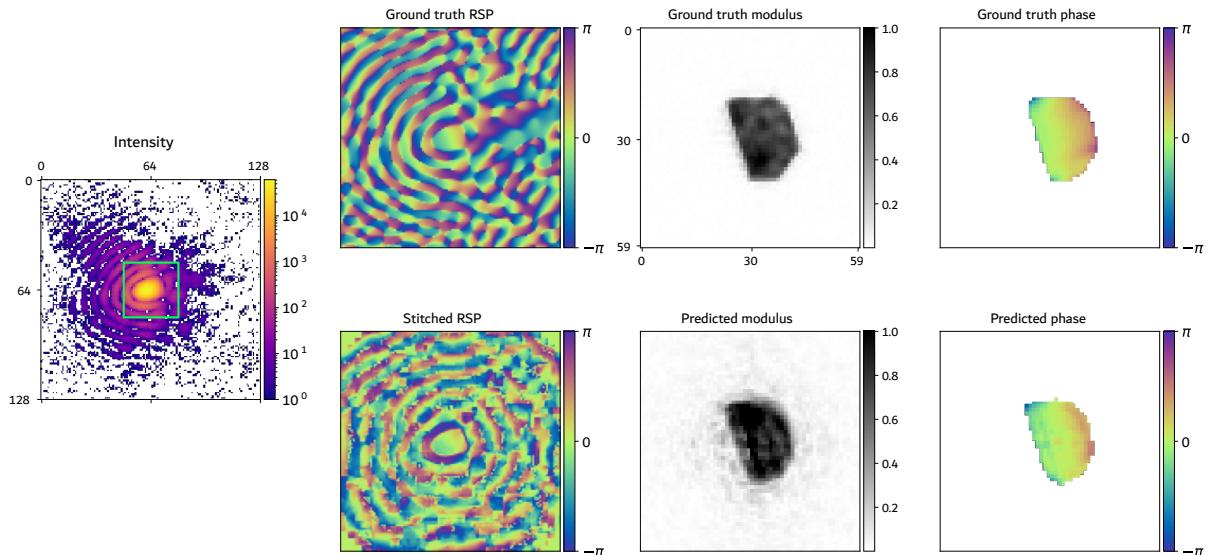
**Figure 6.26:** Examples of RSP prediction for outer patches. The first row shows the central slice of some patches extracted far from the central part of the BCDI peak. The second row shows the ground truth RSP, the third row the RSP initial guess obtained from the overlap with the nearest neighbor of the innermost shell, and the last row shows the DL output. The DL prediction is limited to the missing region (in red).

### 2217 6.6.6 Results on the full RSP - Stitching the patches

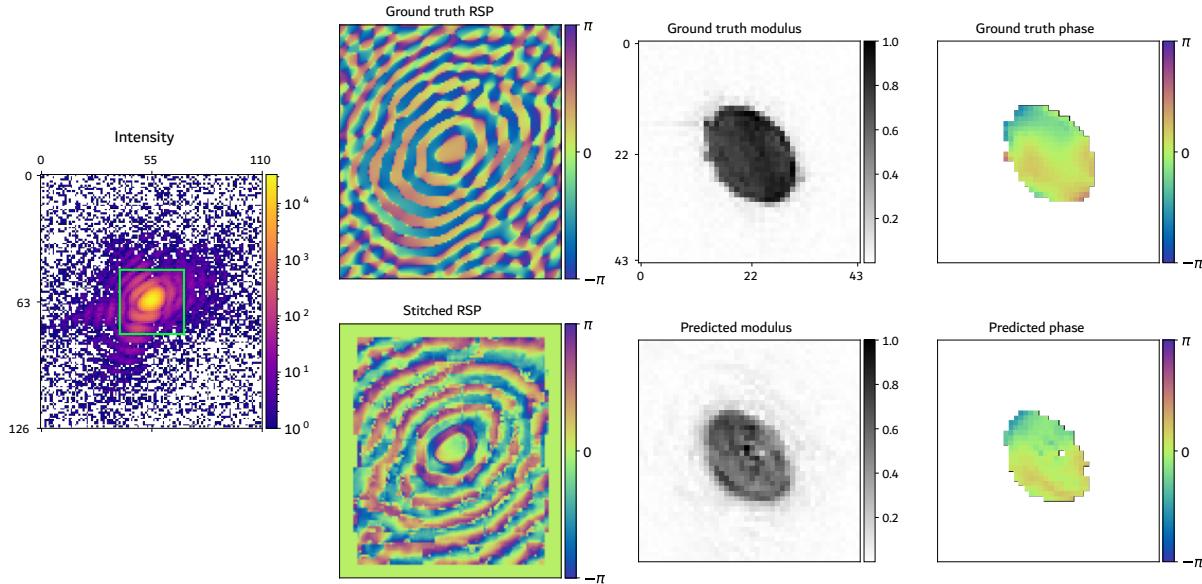
2218 Here the results of the combination of the two CNNs are presented for the low-strain case. Once  
 2219 completed the training the model was tested on full simulated and experimental BCDI pattern.  
 2220 In order to properly retrieve the full RSP corresponding to the diffracted intensity a stitching  
 2221 algorithm for the predicted patches was designed. The stitching takes place progressively start-  
 2222 ing from the central patch and updating the full RSP array shell by shell. Once the prediction  
 2223 of the central patch RSP is obtained from *Central-CNN*, the patches of intensity belonging to  
 2224 the first shell at distance 10 pixels are extracted and the overlapping regions between each  
 2225 patch with the central one are calculated. The predicted central patch RSP for each overlapping  
 2226 region is therefore located in the initial guess RSP array that is given to the model as input  
 2227 paired with the intensity patch. Subsequently, the full batch of pairs  $(I, \varphi_{\text{guess}})_{\text{shell}=1}$  is sent  
 2228 through *Outer-CNN* and the corresponding RSP output is obtained. At this stage two main  
 2229 issues have to be considered, namely: (i) during the training of *Outer-CNN* the initial guess RSP  
 2230 was taken from simulated *ground truth* RSPs while here it is taken from the model's previous  
 2231 prediction itself. It is for this reason that any small unavoidable discrepancies between the  
 2232 predicted and ground truth RSP can lead the model to further errors. (ii) When a round of RSP  
 2233 is predicted there are overlapping regions between patches of the same shell and patches of the  
 2234 previous batch. The most straightforward way to perform the stitching of the patches into the  
 2235 full RSP is to overwrite each time the results. However, although a better approach based on  
 2236 the average of the overlapping prediction was implemented, the issue did not seem to be fully  
 2237 resolved. A first test was conducted with this simpler solutions for both issues, and a more  
 2238 robust approach is briefly discussed at the end of the section.

2239

2240 The crop-predict-stitch method is hence repeated until the last shell. The number of shells  
 2241 and predictions scales with the size of the BCDI data, and it is however always restricted to a  
 2242 spherical region around the Bragg peak. It is therefore less accurate for non-cubic data. Here,  
 2243 Fig.6.27 shows an example of full RSP stitching for a cubic 128 pixel-sided simulated BCDI  
 2244 pattern, while Fig.6.28 reports the analogous result for an experimental data phased with PyNX.



**Figure 6.27: Example 1.** Results of the stitching of RSP predicted patches for 3D simulated data (central slice displayed). The green square on the intensity figure represents the central patch.



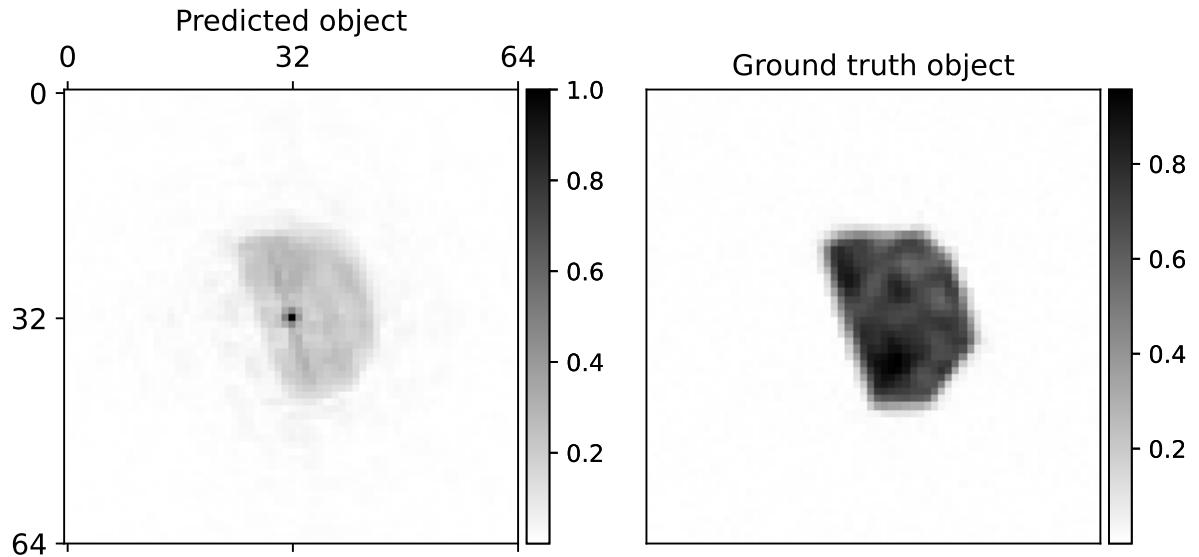
**Figure 6.28: Example 2.** Results of the stitching of RSP predicted patches for 3D experimental data (central slice displayed). Here the ground truth quantities are obtained with PyNX phase retrieval of the diffraction pattern.

2245 What emerges from Figs. 6.27 - 6.28 can be summarized as follows:

- 2246 (i) the accurate prediction of the central RSP patch is fundamental to retrieve the low resolution  
2247 estimate of the object
- 2248 (ii) the stitching is problematic already from the first shell outside the central patch, most  
2249 likely because of the two issues pointed out above (initial guess from model prediction and  
2250 RSP averaging of overlapping regions)
- 2251 (iii) the outer RSP patches correctly infer the oversampling ratio as the “thickness” of the  
2252 RSP oscillations matches the one of the diffracted intensity, nonetheless, the preferred overall  
2253 orientation of the fringes seems to be circular.
- 2254 (iv) from the reconstructed object’s modulus a non-physical more intense spot in the center of  
2255 the array had to be filtered out. The occurrence of this spike indicates the presence of wrong  
2256 zero frequency components in reciprocal space, hence wrong zero RSP values<sup>1</sup>. Fig. 6.29 shows  
2257 the object’s modulus before the removal of the high intensity spike located in the center.

---

<sup>1</sup>For zero values of the RSP in the center of the object we have  $O(\mathbf{r} = 0) = \sum_{\mathbf{q}} \sqrt{I(\mathbf{q})} e^{i\varphi(\mathbf{q})} = \sum_{\mathbf{q}} \sqrt{I(\mathbf{q})}$  hence an incorrect constructive interference of zero-RSP contributions that leads to the central spike.

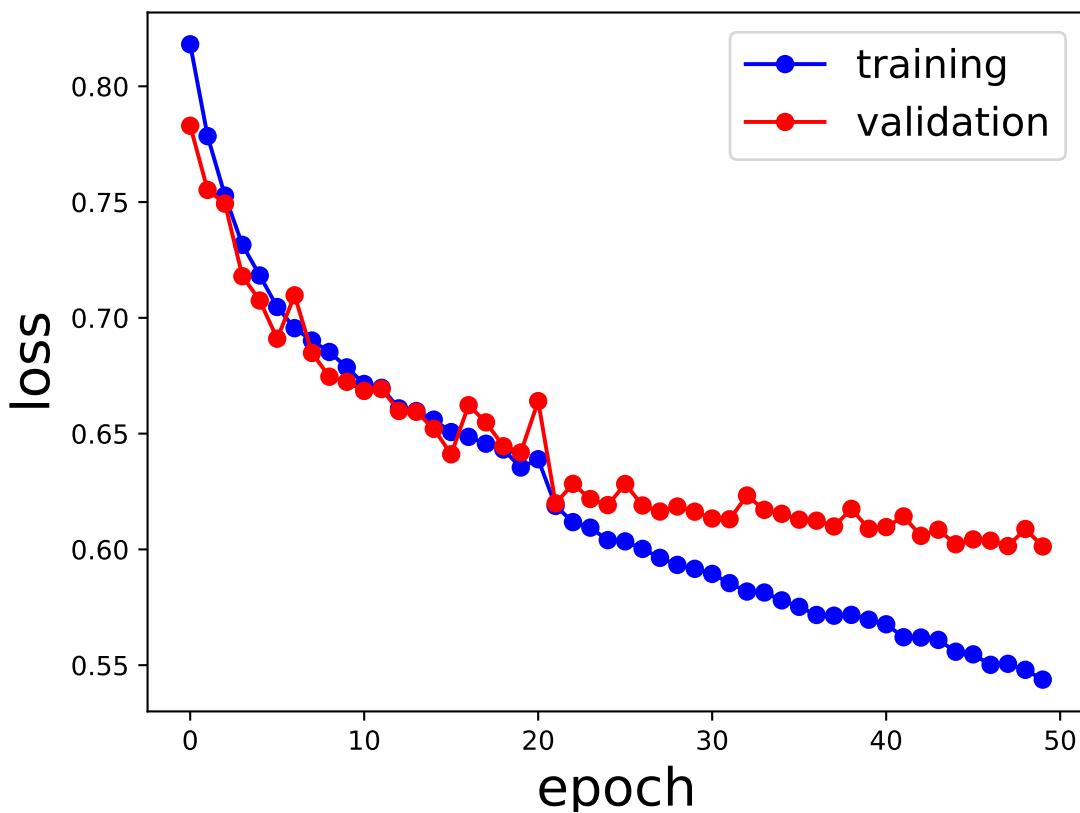


**Figure 6.29:** Predicted object's modulus before the filtering of the zero-frequency component, corresponding to noise in reciprocal space.

## 2258 6.7 Patches: 3D case high strain

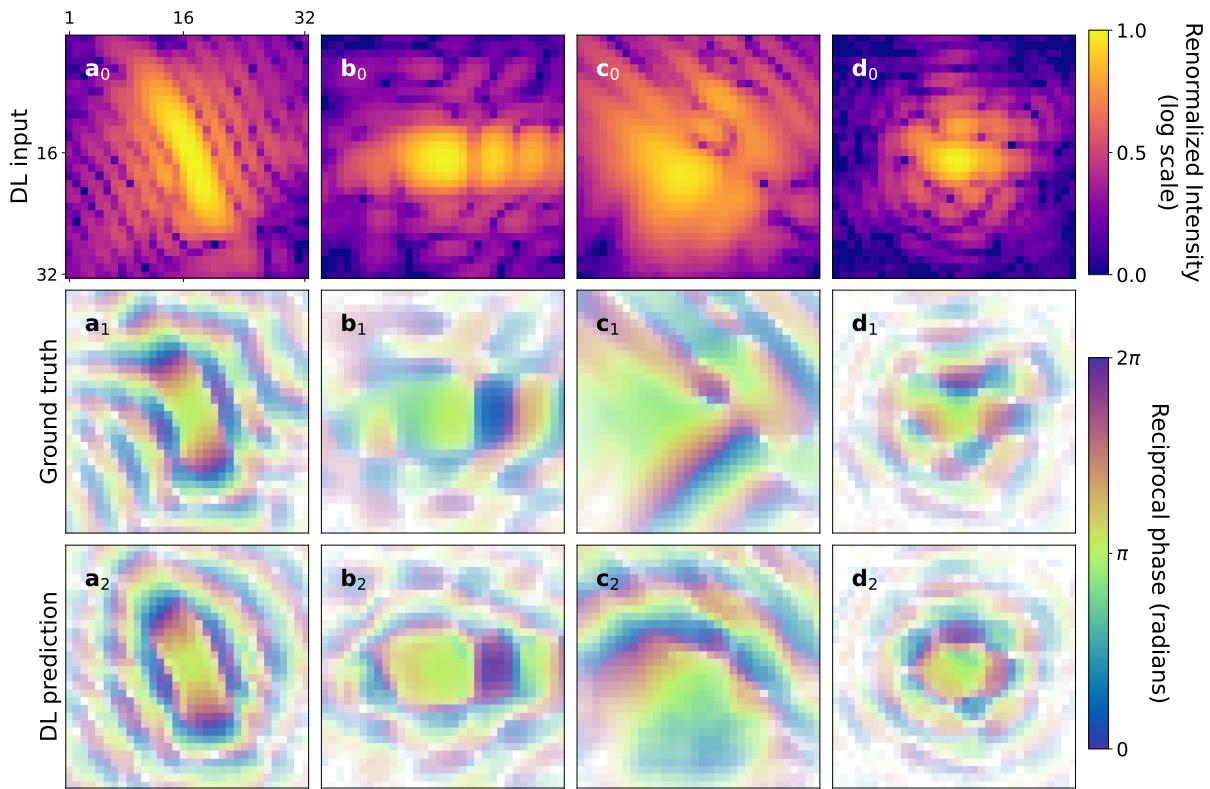
2259 For completeness, the same model has been trained on a dataset containing highly strained  
 2260 simulated diffraction patterns. Each BC DI pattern has been simulated following the procedure  
 2261 explained above in Sec. 5.4.1 and 6.6.2 for a total amount of 50000 for both central and outer  
 2262 patches. The two CNN have been trained for 50 epochs each and then tested on new simulated  
 2263 data.

2264 For what concerns the first CNN trained on central patches it was possible to deduce a  
 2265 more difficult learning from the loss curves. While in Fig. 6.23 the validation loss reaches 0.3 at  
 2266 the end of the training, for Fig. 6.30 it only drops to 0.6 with a significant divergence between  
 2267 training and validation starting from the 20th epoch. A higher loss value is nevertheless  
 2268 expected because of the more complex intensity-RSP mapping for the high strain cases, while  
 2269 the increasing gap between training and validation loss is a signature of poor generalization,  
 2270 early symptom of overfitting.

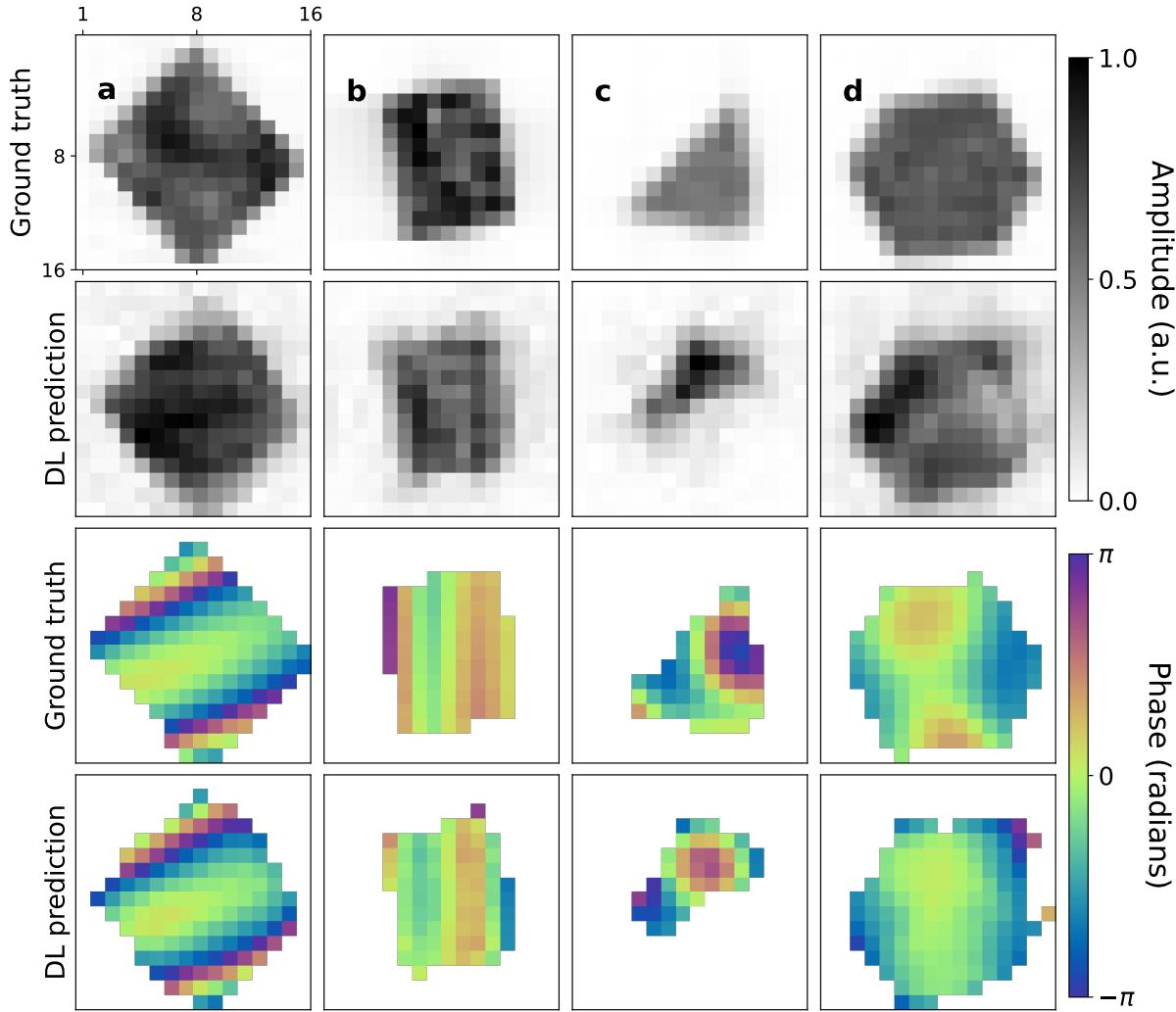


**Figure 6.30:** Training and validation loss trends for the training of *Central-CNN* on central patches of highly strained BCDI patterns. Higher loss values can be observed when comparing with the low strain case (Fig. 6.23) and a beginning of overfitting from the 20th epoch is also visible.

2271 The results on test data show that the model does not always manage to correctly predict  
2272 the RSP, especially when the iso-phase regions are not spherically symmetrical (Fig. 6.31c). It  
2273 however succeeds to retrieve the correct RSP oscillations (Fig. 6.31a) inside the central fringes  
2274 elongated by the high strain. The reconstructions in real space, shown in Fig. 6.32, confirm  
2275 satisfactory results for the first column and poor results for the third column.

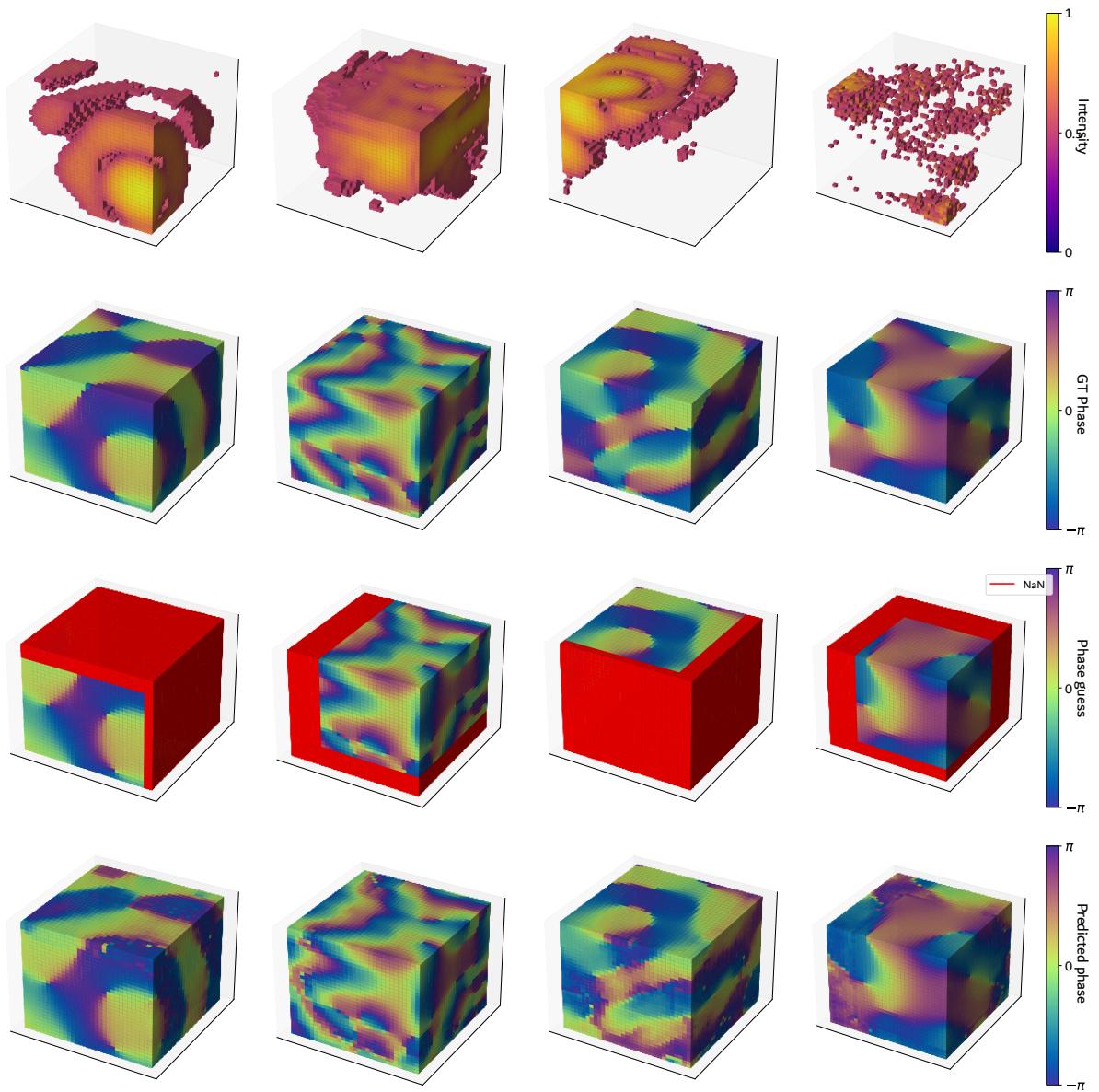


**Figure 6.31:** Slices of the central patches. First row shows four different examples of simulated high-strain BCDI patterns cropped around the center of the peak. Ground truth and predicted RSP are shown in second and third row respectively. The model manages to estimate the correct RSP when this shows spherically symmetric iso-phase regions while struggles more for asymmetries.



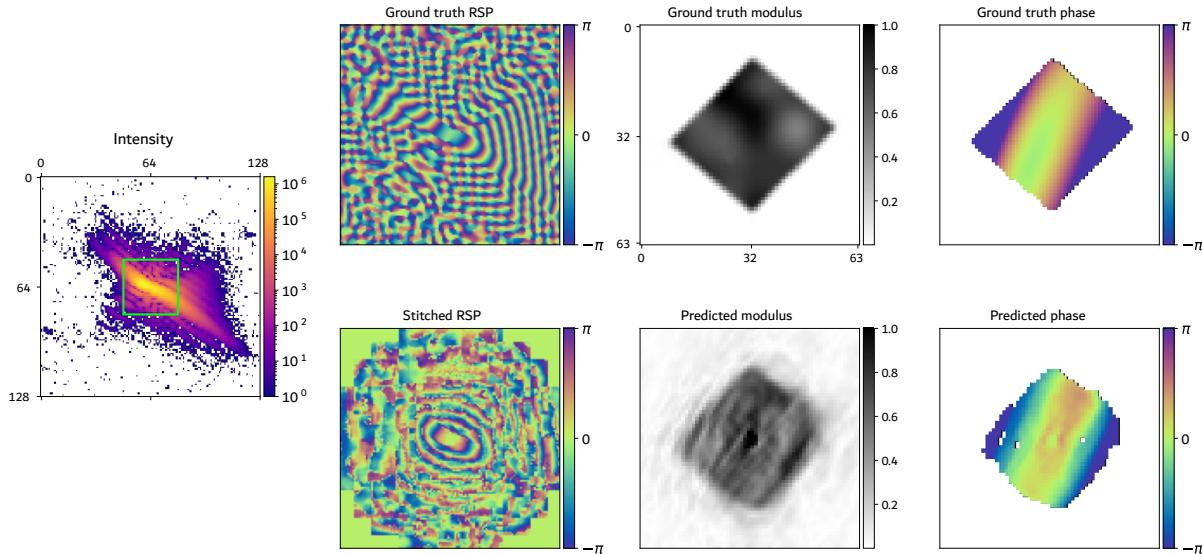
**Figure 6.32:** Corresponding reconstructed objects. Except for case **c** the predicted RSP is good enough to well estimate the size, shape and phase of the real space object.

Regarding the *Outer-CNN* trained on outer patches of highly strained patterns 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.6.33). The good accuracy of the outer patches predictions suggests that the crucial and more challenging map to retrieve is the central patch.



**Figure 6.33:** Examples of RSP prediction for outer patches cropped from simulated high-strain data. Similarly to the examples shown in Fig. 6.26 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.

2281 For completeness, the result of the full RSP stitching and the corresponding reconstructed  
 2282 object for the high-strain case is shown in Fig. 6.34. A simulated test data has been used for  
 2283 the ground truth comparison. It is clear that the stitching algorithm is performing poorly as  
 2284 observed for the low-strain case. However, the central RSP patch is fairly similar to the ground  
 2285 truth and therefore the low resolution reconstructed object shows roughly the correct shape  
 2286 and phase.



**Figure 6.34:** Results of the stitching of RSP predicted outer patches for 3D simulated “high-strain” data (central slice displayed). Compared to Fig.6.28 it is visible how the stitched RSP is more “confused” outside the central patch. The less regular and symmetric fringes’ spatial arrangement is not correctly recovered during the stitching process. However, the discrete accuracy of the central patch allows for a close low-resolution estimate of the object’s modulus and phase.

### 2287 6.7.1 Discussion

2288 Although it failed, this study on the prediction of the RSP in smaller patches has led to a better  
 2289 understanding of the problem and nevertheless unveiled some interesting insights. For instance,  
 2290 it showed that the retrieval of the mapping between patches is possible with a CNN trained  
 2291 with the WCA loss function, untying the relationship with the real space object. Moreover, it  
 2292 emerged that the main difficulty of this approach is given by the stitching of the RSP patches  
 2293 into the full array. As mentioned above, the hypothesized reasons for this problem are (i) the  
 2294 fact that the model “sees” simulated ground truth RSP guesses during the training and predicted  
 2295 ones during inference, and (ii) the averaging of RSP predictions for overlapping voxels. In  
 2296 order to overcome these limitations other approaches were contemplated but never realized  
 2297 for lack of time. In particular, it was imagined a way to extract, analyze and stitch the patches  
 2298 inside the model into a sort of Recurrent Convolutional Neural Network (RCNN) that would  
 2299 keep track of the previous innermost shell thanks to a dedicated convolutional Long-Short  
 2300 Term Memory (LSTM) [190]. By doing this the model would always be exposed to its own  
 2301 RSP predictions as initial guess for outer patches and the RSP average over overlapping voxels  
 2302 could be replaced with a convolutional layer with non-linear activation function. While the  
 2303 attempts of setting up such model are not reported here, it is mentioned the idea as possible  
 2304 inspiration for future works.

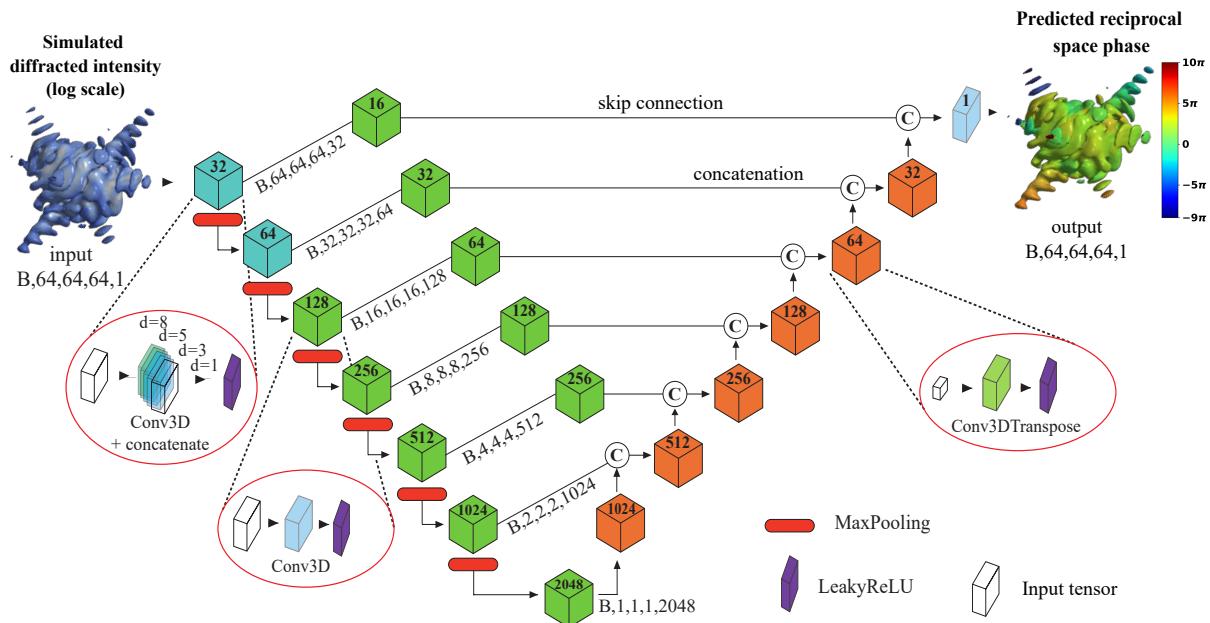
2305

2306 To conclude, main finding of this study on patches is that it is crucial for a good object  
 2307 estimate to accurately predict the RSP in the vicinity of the center of the Bragg peak, and that  
 2308 the CNN model trained with the WCA can accomplish this task for highly strained patterns as  
 2309 well. It was therefore decided to invest the efforts into a regular CNN for the prediction of the  
 2310 RSP of 3D highly strained BCDI patterns with the intermediate size of  $64 \times 64 \times 64$  pixels.

## 2311 6.8 Final model design: 3D case high-strain for central 2312 patch only

2313 Following from the results and considerations relative to the patching approach it was decided  
2314 to focus on the prediction of the central patch only. The size has been increased to  $64 \times 64 \times 64$   
2315 pixels to include more features. Moreover, this is the size employed by most of the recent  
2316 works presented in the beginning of the chapter [167, 182, 183]. Similarly to the previous  
2317 cases, the approach was to create a dataset to train the model with, in a supervised manner  
2318 with the WCA loss function. Only highly strained particles were considered, as it is for those  
2319 that a successful DL model can truly help the PR. The model here described and the results  
2320 obtained on simulated and experimental 3D BCDI patterns can be found entirely in the paper  
2321 "Phase Retrieval of Highly Strained Bragg Coherent Diffraction Patterns using Supervised  
2322 Convolutional Neural Network" [175].

2323 The architecture that was employed is an adaptation of the 3D U-Net employed for smaller  
2324 patches Fig. 6.35 As learned from the preliminary study on the 2D case, to better interpolate the  
2325 diverse distribution of distorted BCDI patterns, the number of trainable parameters and samples  
2326 in the training dataset was increased significantly. Additional encoder and corresponding  
2327 decoder blocks were added for a total of 145 million parameters. Moreover, similarly to the  
2328 model used for the gap inpainting, dilated convolutions were adopted in the first two encoder  
2329 blocks in order to improve the receptive field of the model.



**Figure 6.35:** Schematic of the 3D U-Net model employed for the RSP prediction of highly strained patterns. The model maintains the structure already shown in Fig. 6.22, adapted for  $64 \times 64 \times 64$  pixels data. Moreover additional convolutional blocks have been added between skip connections, to build a deeper layer configuration, with more trainable parameters.

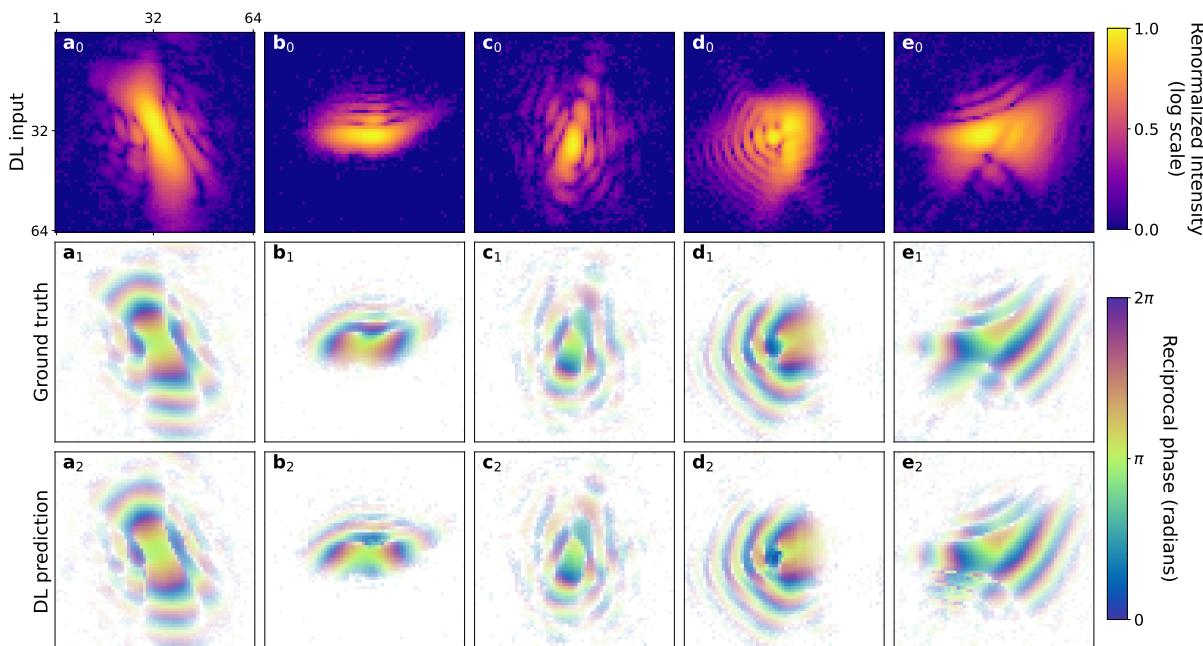
2330 95000 simulated BCDI patterns have been created following the procedure described in  
2331 Sec.5.4.1 on a cubic 64 pixel-sided grid for different particle's shape, strain distribution, over-

2332 sampling conditions and noise levels. Another smaller dataset containing 4000 samples was  
 2333 created instead for testing the model.

2334 The model was trained with the WCA loss function for 60 epochs with a learning rate of  
 2335  $10^{-4}$ . To speed up the process, the training has been conducted using two NVIDIA TeslaV100-  
 2336 SXM2-32GB GPUs using the MirroredStrategy feature for synchronous training across multiple  
 2337 devices provided by the Tensorflow library. This measure allowed to reduce the training time  
 2338 from 2 hours per epoch to 30 mins.

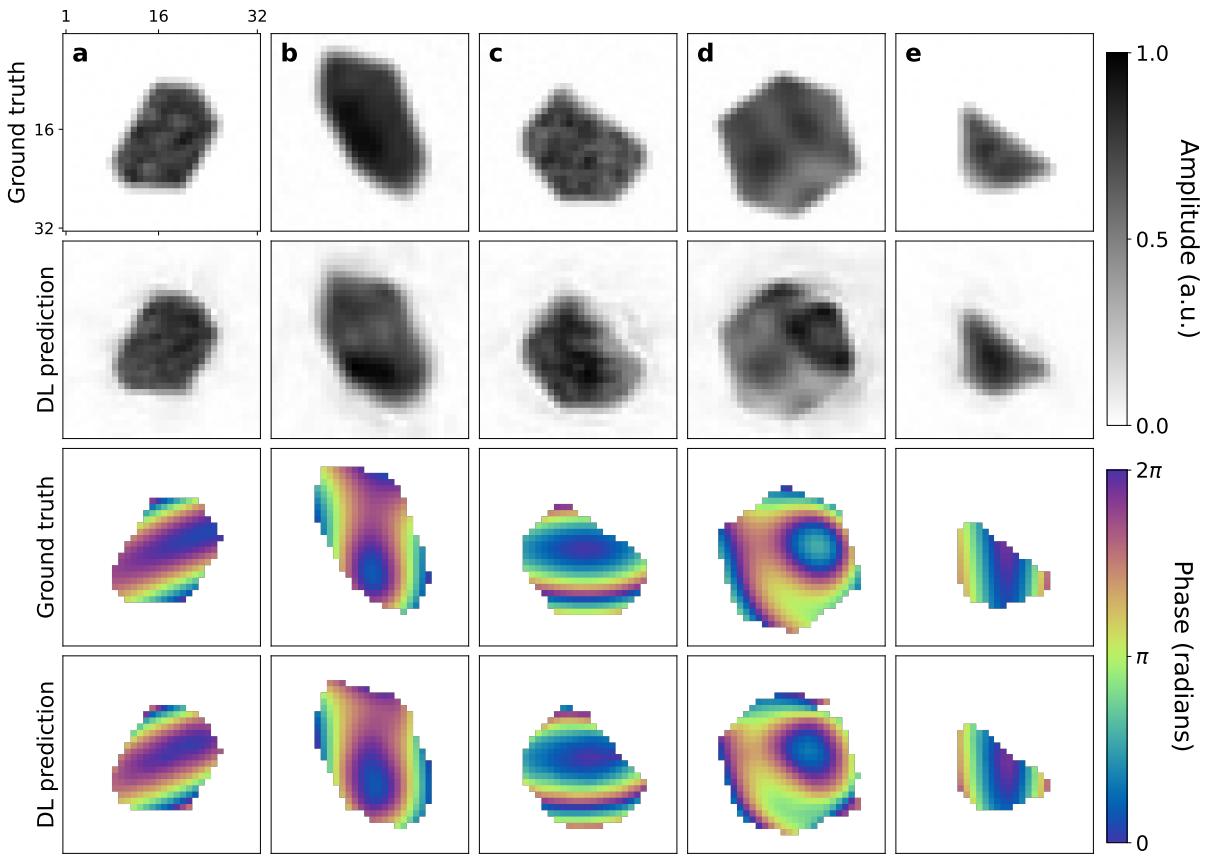
### 2339 6.8.1 Results: simulated data

2340 The model has been tested on simulated data. Figs.6.36 - 6.37 illustrate some examples of  
 2341 comparison between ground truth and predictions for both the RSP and the reconstructed  
 2342 objects respectively.



**Figure 6.36:**  $a_0 - e_0$ ) 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 RSP are displayed with an opacity filter that highlights regions with diffracted intensity.

2343 The model correctly predicts the RSP oscillations inside the fringes even when they have  
 2344 been distorted or merged into a single continuous intensity stripe (Fig. 6.36 a-b) because of the  
 2345 high strain along the corresponding axis in real space.



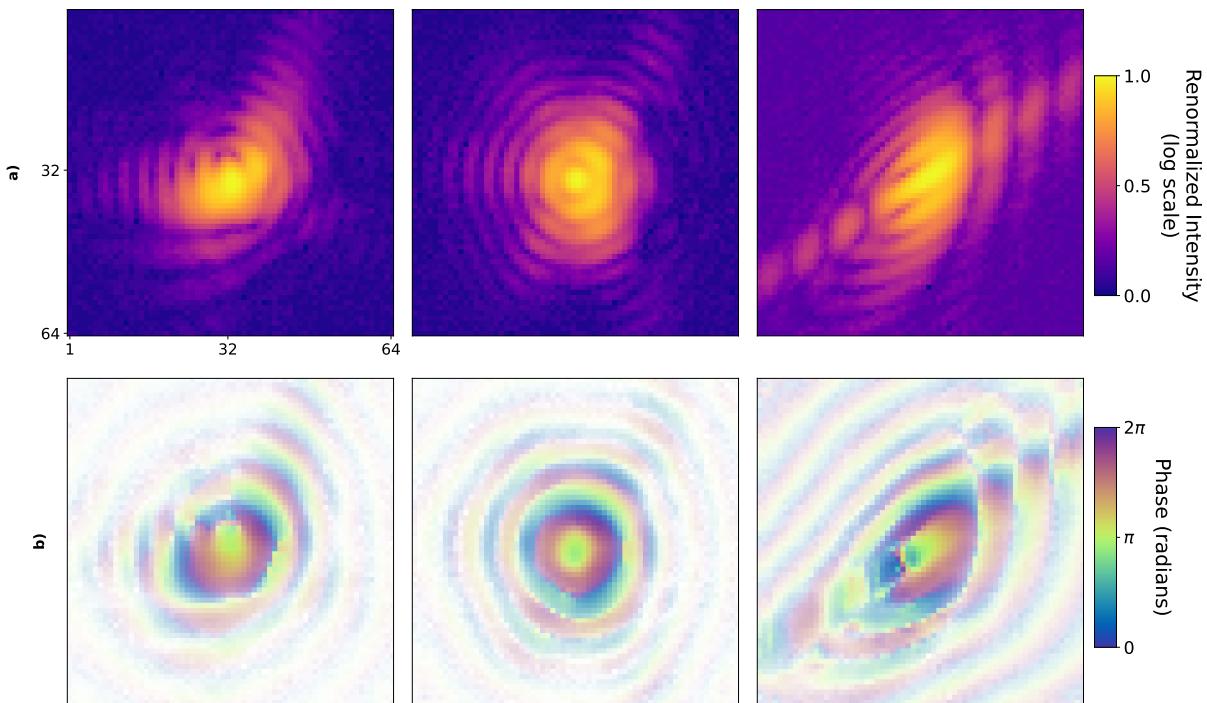
**Figure 6.37:** Reconstructed objects from the diffraction patterns in Fig. 6.36. **a-e)** Central slices of both the modulus and phase for the ground truths and the DL reconstructions.

2346 Except for some noise and inhomogeneities in the objects' moduli, the reconstructions from  
 2347 the predicted RSP achieve good accuracy on simulated data, for different particle shapes and  
 2348 strain distributions. It is worth mentioning that the presence of noise affecting the objects'  
 2349 shapes is an effect of the loss function that is not computed in real space. The model is never  
 2350 directly shown that the real space object are compactly supported, as it comes as consequence  
 2351 of the correct RSP prediction. However, while small discrepancies of the RSP prediction result  
 2352 in noise on the object shape, an overall accurate RSP prediction ensures the retrieval of both the  
 2353 correct shape and phase of the object. This fact is of primary importance when considering the  
 2354 use of the DL prediction as starting point of iterative refinement with conventional algorithms.  
 2355 It is indeed easier to reach convergence from a low resolution noisy but accurate estimate  
 2356 of the object rather than a clean but inaccurate one. Moreover, in experimental conditions  
 2357 the shape may be known via other techniques (Scanning Electron Microscopy) while the  
 2358 strain field, derived from the retrieved object's phase, is the ultimate interesting result of the  
 2359 reconstructions.

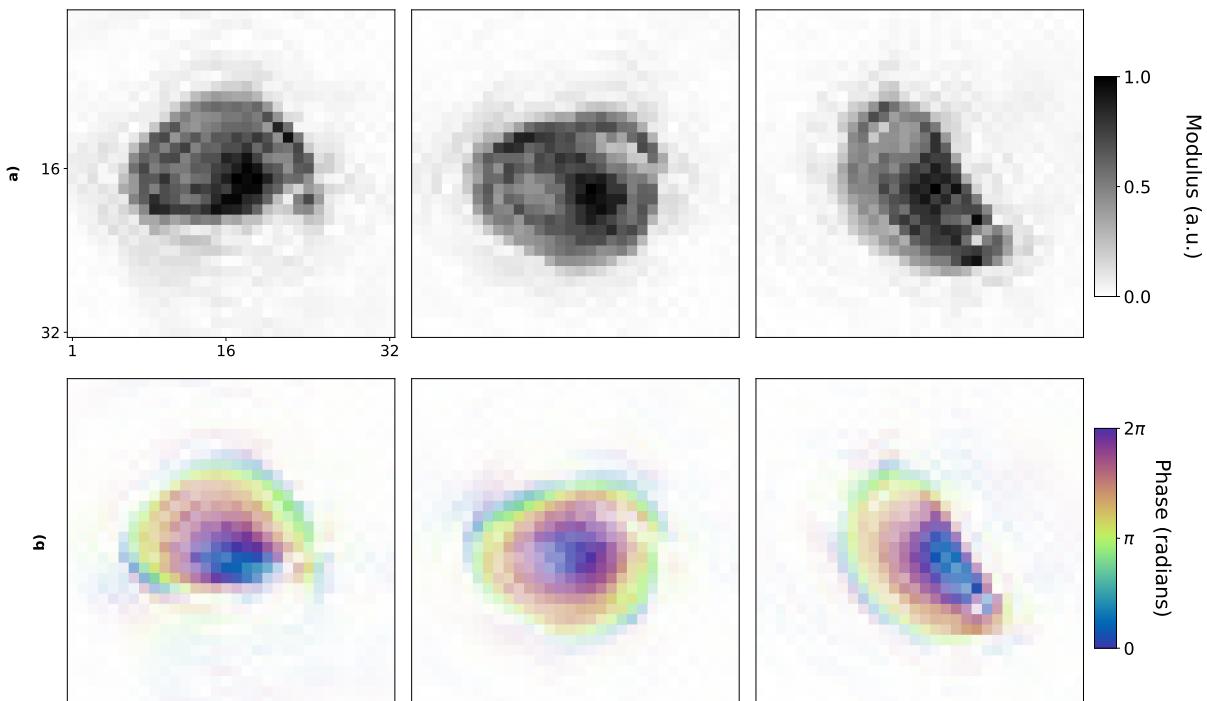
### 2360 6.8.2 Results: experimental data

2361 Given the satisfactory results obtained on simulated data the DL model was also tested on  
 2362 experimental data. Two relevant examples of BCDI patterns collected at the ID01 beamline  
 2363 of the ESRF-EBS are considered here. The first pattern (Fig. 6.38 a) is given by a platinum

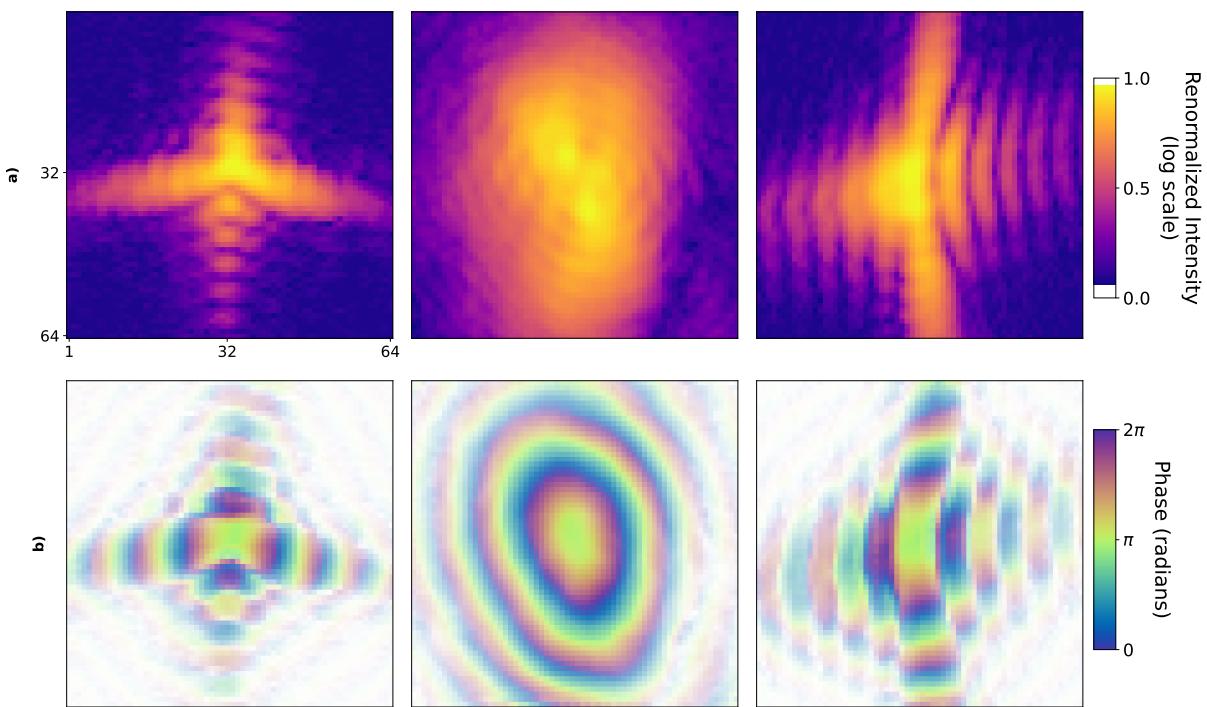
nanoparticle on Yttria-stabilized zirconia (YSZ) (Particle 1) [191] while the second (Fig. 6.40 b) is a dewetted platinum/palladium bilayer on a sapphire substrate (Particle 2) [192].



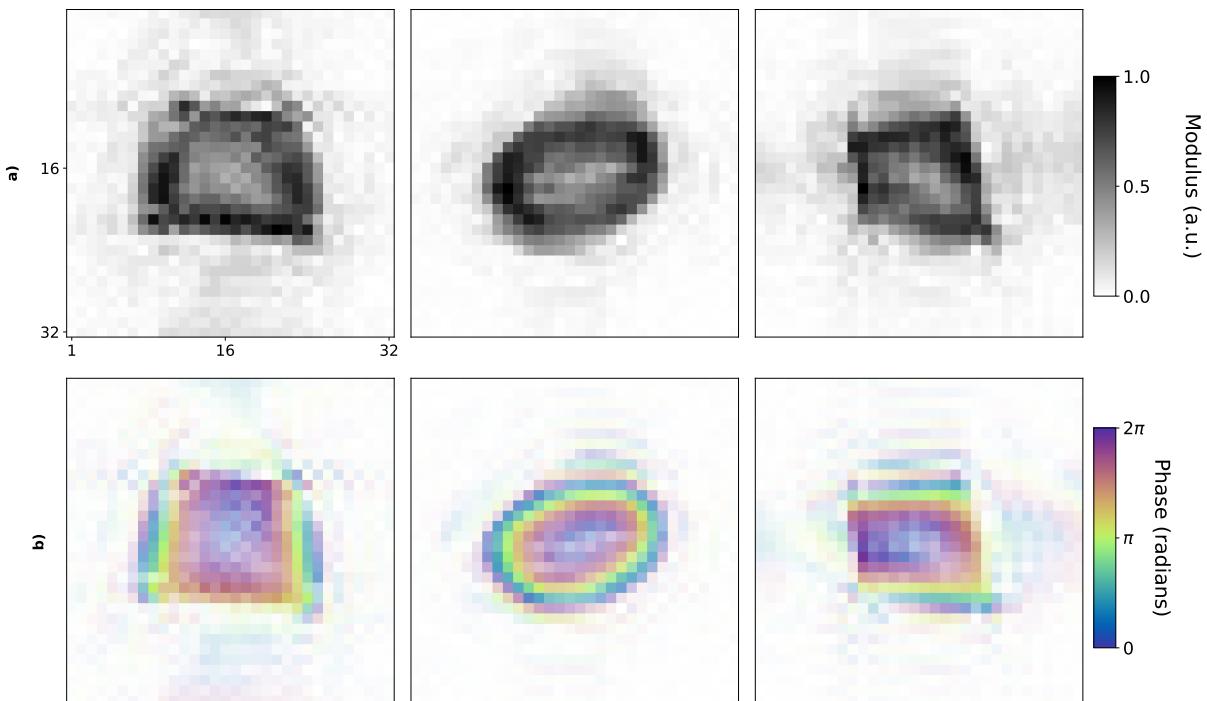
**Figure 6.38:** **a)** The three central slices of the 3D experimental diffraction pattern measured at ID01 from the Pt sample (Particle 1). The images have been transformed in log-scale and renormalized between 0 and 1, ready to be processed by the DL model. **b)** Corresponding slices of the DL model RSP predictions.



**Figure 6.39:** Corresponding three central slices of the reconstructed Particle 1 modulus **(a)** and phase **(b)**. Each slice has been cropped around the particle for better visualization.



**Figure 6.40:** **a)** Central slices of the 3D experimental diffraction pattern measured at ID01 from the Pt/Pd sample (Particle 2). Note the shape of the diffraction pattern with broken centro-symmetry; signature of the high-strain. **b)** Corresponding slices of the DL model RSP predictions.



**Figure 6.41:** Central slices of the reconstructed Particle 2, modulus **(a)** and phase **(b)**

Fig. 6.39 and 6.41 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 (compact support and smooth phases).

2369 In these cases of PR of experimental data it is impossible to establish a comparison with  
2370 a ground truth because it is not available. However, the comparison has been made with the  
2371 results obtained with standard iterative algorithms.

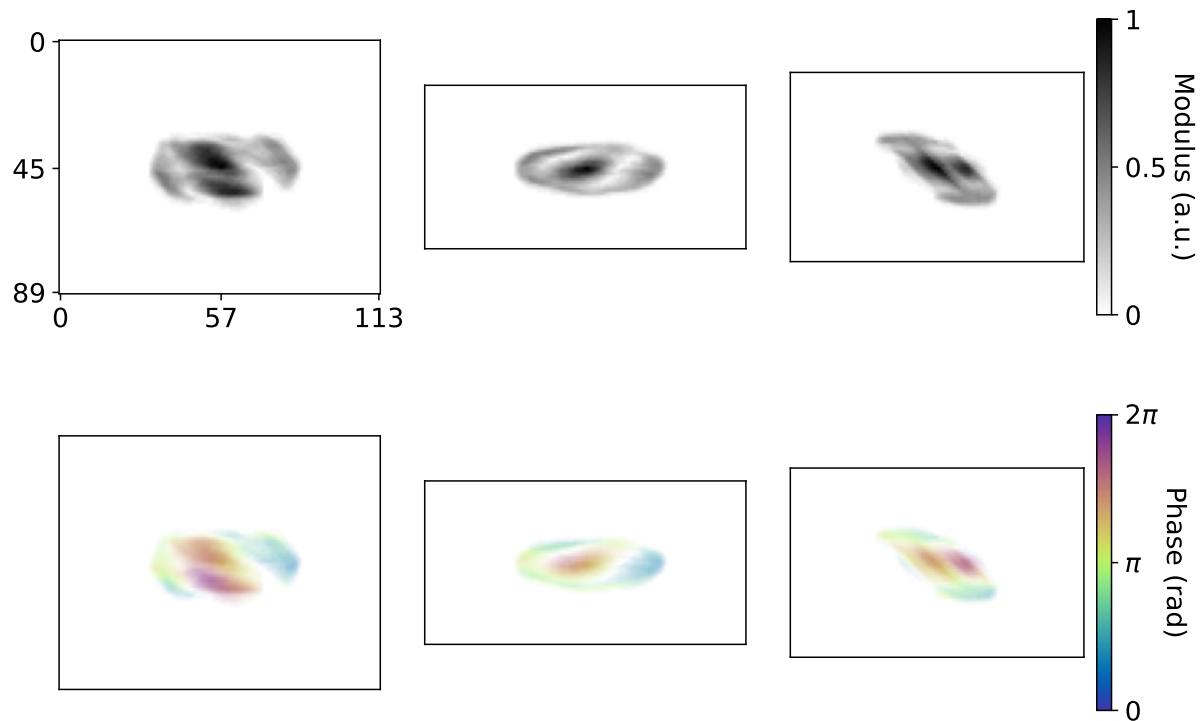
2372 For this, 60 independent and randomly initialized runs have been launched with PyNX in  
2373 order to reconstruct these datasets but no satisfactory results were obtained. Precisely, the  
2374 recipe of 400 HIO + 1000 RAAR + 300 ER and different thresholds for the support estimation  
2375 were used for each run (see Table 6.2). Moreover, the 3 best results according to the Mean-to-  
2376 Max metric [193, 194] were combined with mode decomposition [195] to improve the quality  
2377 of the reconstruction. This last passage, through a Singular Value Decomposition (SVD) of  
2378 the selected reconstructions, allows to estimate the reproducibility of the result by assessing  
2379 the magnitude of the first eigenvalue (first mode). Values above 90% typically imply good  
2380 reproducibility, often associated to good convergence.

2381 However, in both cases the obtained reconstructions were not satisfactory as holes were  
2382 present (Fig.6.42), or the support was excessively shrunk (Fig.6.44).

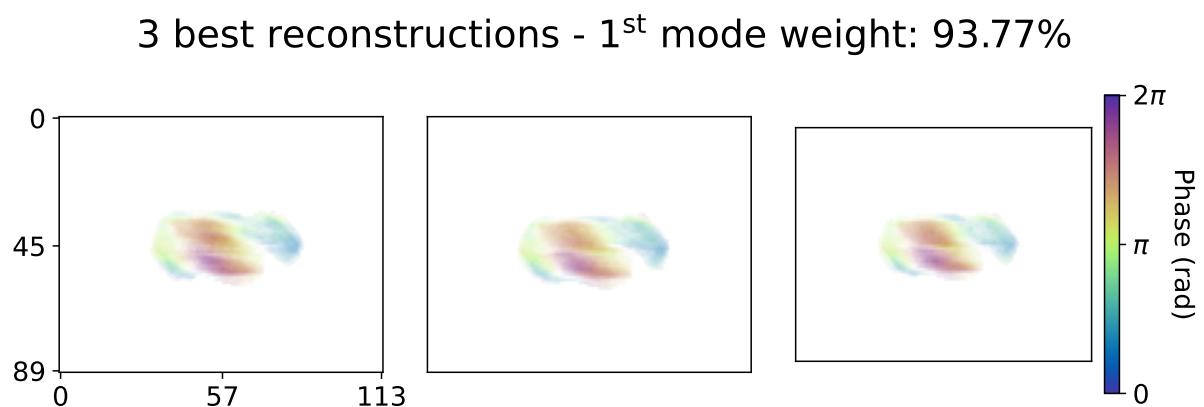
2383

Parameter	Value
obj	autocorrelation
support_autocorrelation_threshold	(0.09, 0.11)
recipe	400 HIO + 1000 RAAR + 300 ER
nb_runs	60
support_update_period	50
support_threshold	(0.15, 0.25)
update_border_n	3
post_expand	(1, -2, 1)

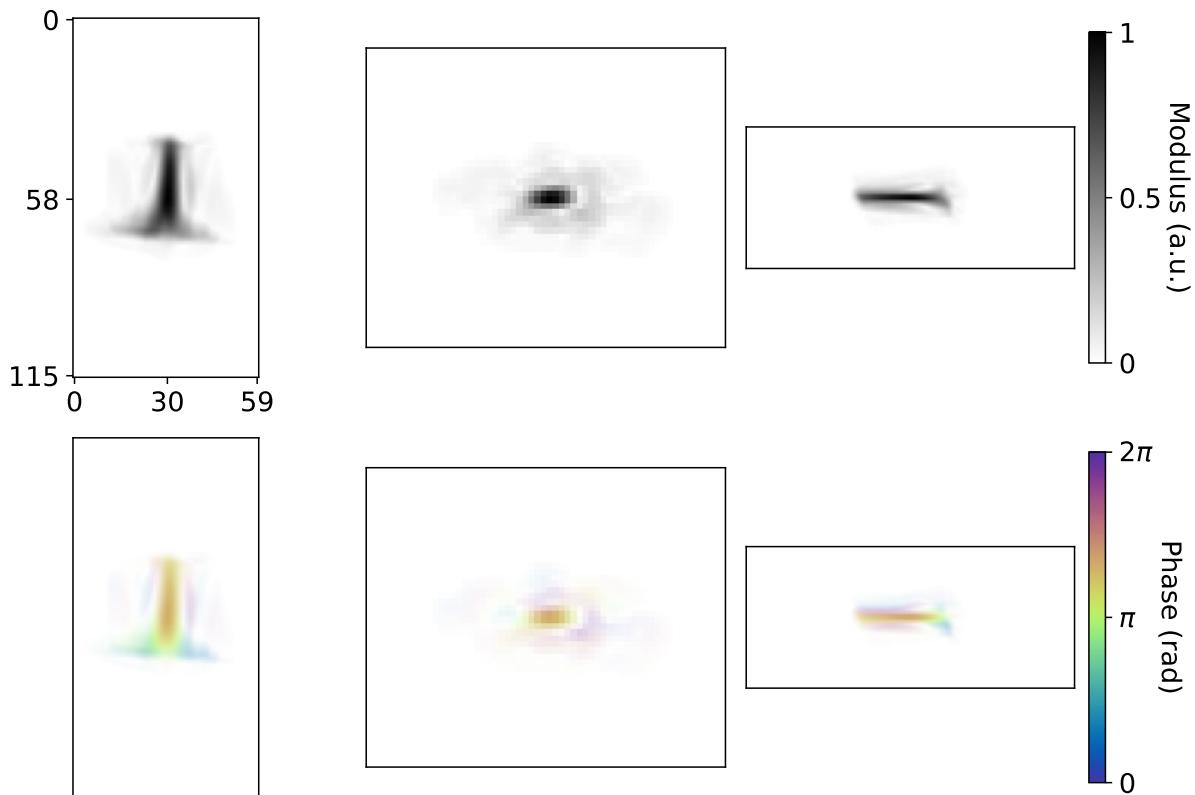
**Table 6.2: PyNX parameter settings for standard PR.** Each of the 60 independent runs starts from an object obtained from the inverse Fourier transform of the diffraction pattern (autocorrelation). Pixels above the autocorrelation's maximum multiplied by a random threshold between (0.09 - 0.2) different for each run, are included in the support. In each run 400 cycles of HIO are followed by 1000 of RAAR and 300 of ER. The support is updated every 50 iterations using the shrink-wrap algorithm [95]. During any of these updates the pixels above the object's modulus maximum multiplied by a random value between (0.15 and 0.25) are included in the support. Moreover, only a shell of +/- 3 pixels around the outer border of the support are updated to avoid too large variations, and a sequence of expansion, shrinking and re-expansion by (1,-2, 1) pixels respectively is evaluated at each update step, in order to make the estimate more robust.



**Figure 6.42:** 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 and domains in the phase denotes a poor quality of the result.

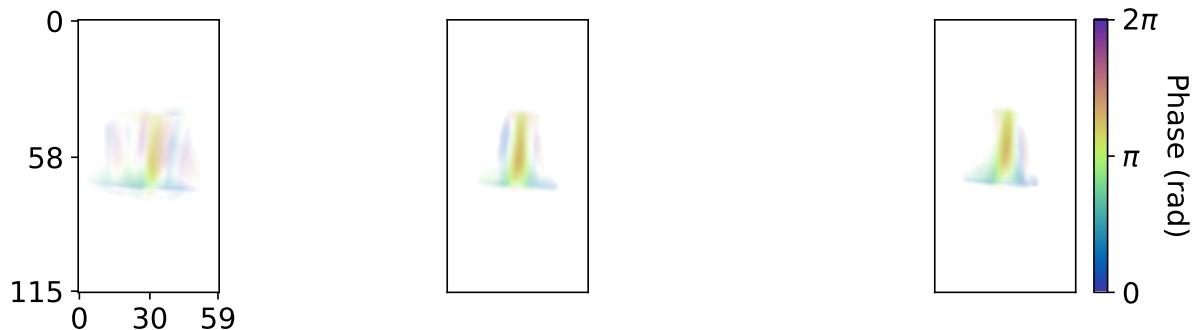


**Figure 6.43:** Phase of the three best reconstructions of Particle 1. Despite the similarity of the solutions, supported by the large weight of the first SVD eigenvalue [195], each of them and their combination presents amplitude dips and phase domains not expected from the studied Pt sample.



**Figure 6.44:** Combination of the 3 best reconstructions of Particle 2 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.

3 best reconstructions - 1<sup>st</sup> mode weight: 72.68%



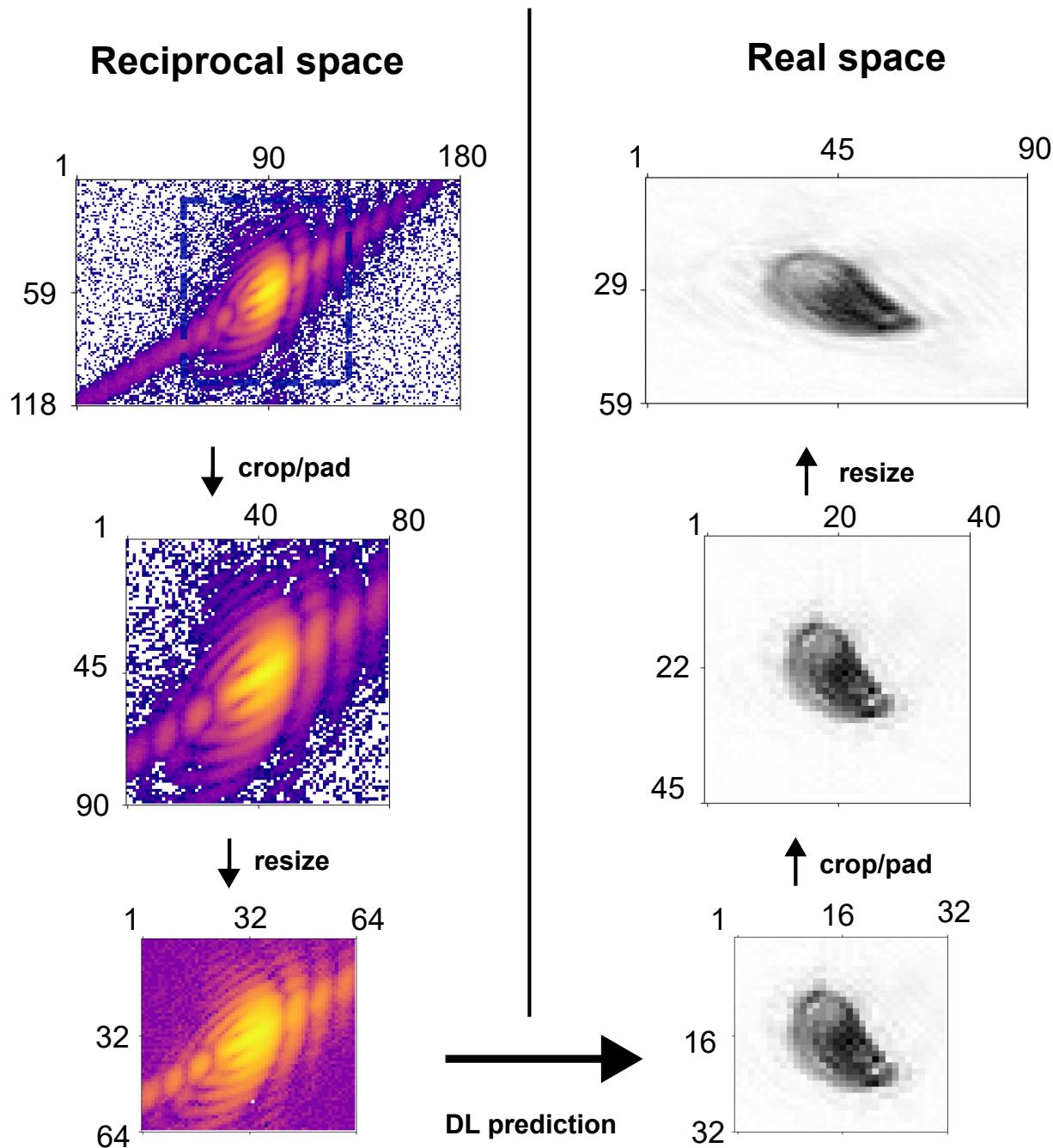
**Figure 6.45:** Phase of the three best reconstructions of Particle 2. The low reproducibility of the result highlighted by the diversity of the reconstructions and supported by the relatively low first SVD eigenvalue, confirms the poorly accurate reconstruction shown in Fig.6.44.

## 6.9 Refinement with iterative algorithms

2384 It has been shown in Figs. 6.42 - 6.44 how the PR of highly-strained particles with conventional  
2386 iterative algorithms and standard routines can be challenging. In order to achieve successful

2387 reconstructions, fine parameter tuning operated by expert users as well as large numbers of  
2388 runs are often required. On the contrary, the DL approach, while fast at inference, is limited  
2389 to low-resolution reconstructions and only generates outputs based on the approximation of  
2390 the statistical distribution of simulated BCDI patterns. For this reason, the DL model for the  
2391 phasing of experimental data needs be used as a preprocessing step that can provide a starting  
2392 point for further iterative refinement. Few iterations of ER steps can be used to polish the  
2393 DL estimate, simplifying the convergence, assumed that the DL estimate is located near the  
2394 solution, in the search space. In this way, the DL model can play a crucial role in guiding the  
2395 search toward the correct solution neighborhood, significantly reducing computational costs  
2396 and overall wall-clock time.

2397 At this stage it is worth mentioning that, since the DL model was trained on 64 pixel-size  
2398 cubic volumes, it was necessary to crop and resize the experimental diffraction patterns. In  
2399 fact, especially for large BCDI data, a resizing or binning step before cropping is often needed,  
2400 such that a sufficient amount of data is contained in the 64 pixel-size volume. This procedure  
2401 requires special attention as for different cropping/resizing parameters the DL prediction  
2402 varies. In particular, it is curious to notice that the cropping/padding operations in reciprocal  
2403 space correspond to interpolations in real space and vice versa. For this reason the procedure  
2404 illustrated in Fig.6.46 had to be adopted to adapt the data to the DL format and then bring back  
2405 the predicted object to the size relative to the original data one. This step is fundamental when  
2406 the DL object is then used as starting point for further refinement with classical algorithms.  
2407 Due to the large diversity of the region of interest (ROI) from one experimental BCDI pattern  
2408 to the other, the manual intervention is for the moment needed to adjust the cropping and  
2409 binning parameters such that the data fed into the DL model resembles the simulated data used  
2410 for training.



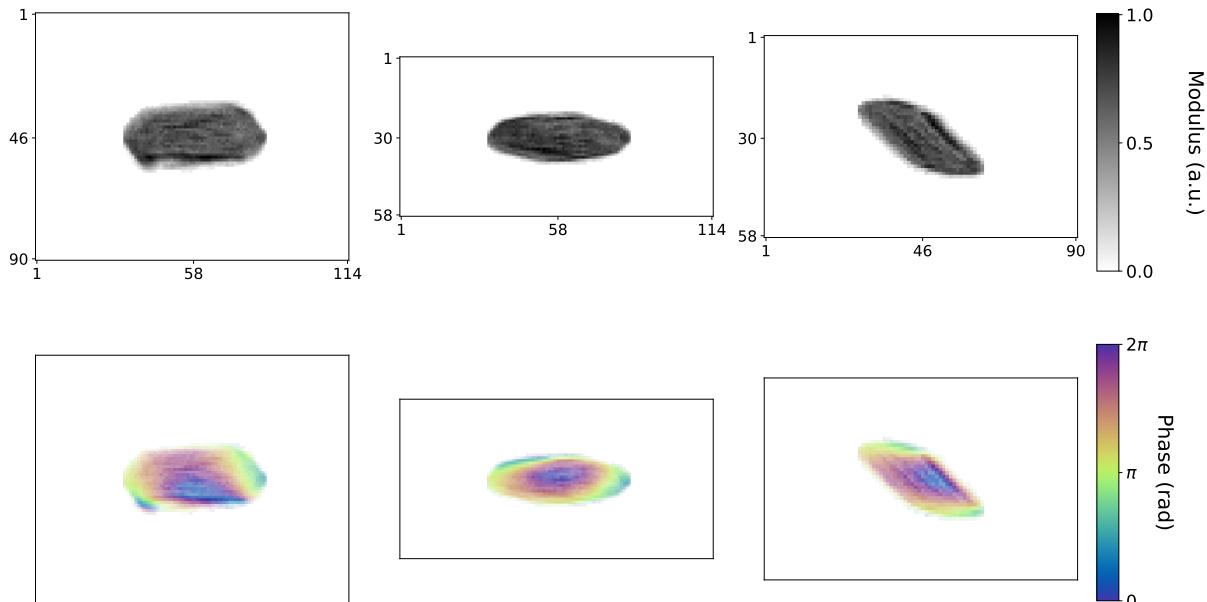
**Figure 6.46:** Manipulation of the datasets. Here the experimental BCDI pattern is firstly cropped around the COM of the Bragg peak, cutting out parts of the signal. The data is then interpolated into a 64 pixel sided cubic grid and transformed in normalized log-scale. The object obtained after the 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. Note that the size of the objects are already shown in boxes of half the size with respect to the corresponding diffraction patterns.

2411 If the DL predicted object is a good low-resolution guess of the solution, few steps of  
 2412 ER are usually enough to reach convergence. In this specific case 300 iterations of ER were  
 2413 performed and, since the estimated DL object is assumed to be close to the solution, only the  
 2414 pixels at the border the support were allowed to be updated (see Table 6.3). Figures 6.47 and  
 2415 6.48 show the results obtained after refinement. Theoretically, one could tune the number of

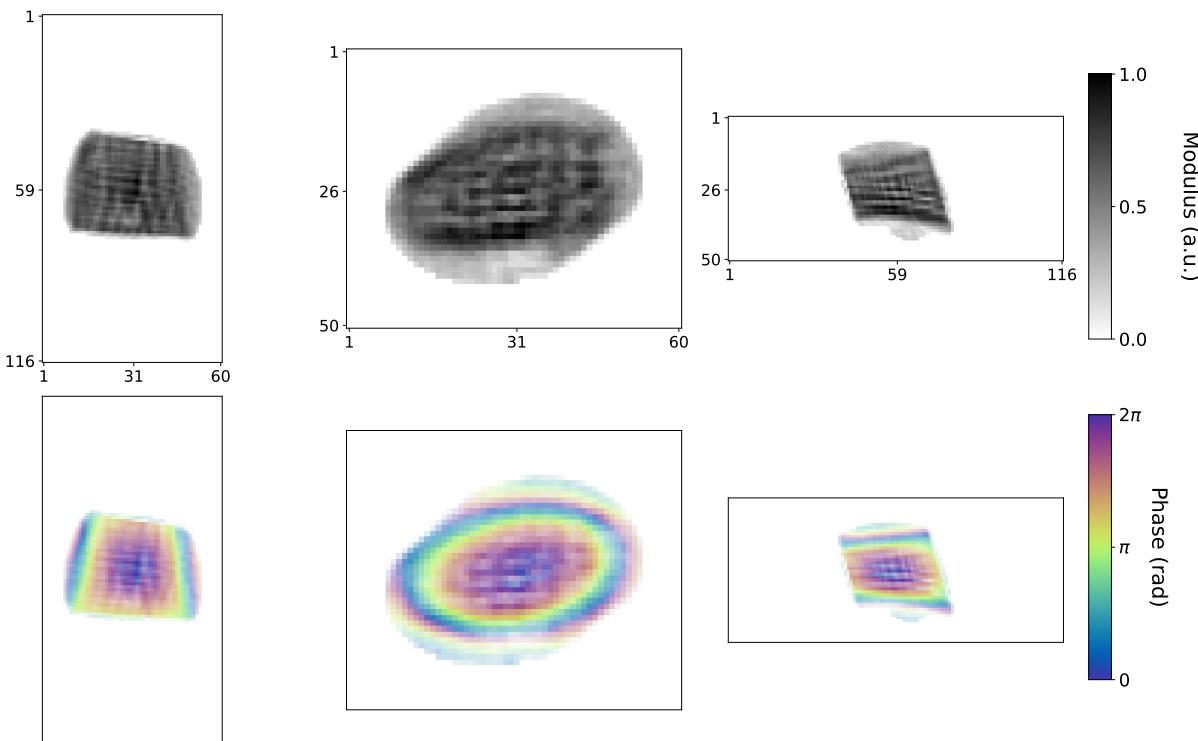
2416 pixels updated at the border of the support depending on the resolution mismatch between  
 2417 the DL data ( $64 \times 64 \times 64$ ) and the full BCDI size. The larger the size difference the more the  
 2418 pixels at the border and vice-versa. However, it was found that a single pixel at the time is  
 2419 often the best choice.

Parameter	Value
obj	DL_obj
recipe	300 ER
nb_runs	1
support_threshold	0.3
support_update_period	50
update_border_n	1
post_expand	(1, -1)

**Table 6.3: PyNX parameter settings for the refinement after the DL prediction** A single run of 300 ER iterations using a support threshold of 0.3 with respect to the maximum of the object's modulus for the support update. This last is performed with the shrink-wrap algorithm, every 30 iterations. At this stage, the only pixels affected by the support updated lie within  $\pm 1$  pixels around the outer border of the support. Moreover, the support is also shrunk and expanded by 1 pixel at each update step.



**Figure 6.47:** Central slices of Particle 1 after 300 cycles of ER for refinement of the DL initial estimate.



**Figure 6.48:** Central slices of Particle 2 after 300 cycles of ER for refinement of the DL initial estimate.

2420     The improved homogeneity of the modulus and the more physical shape of a Winterbottom  
 2421     particle are a clear sign of better reconstructions for both cases. One can observe that the shape  
 2422     and phase of the final reconstruction are not remarkably different from the DL estimates. This  
 2423     proves that the DL model is able to generalize correctly to experimental data as well and that  
 2424     the following ER steps are a good choice for refinement. It is worth noticing that launching  
 2425     several independent runs with the same initial DL guess would not change the outcome as ER  
 2426     converges to the local minimum by projecting each estimate on the constraint sets, therefore  
 2427     never “exploring” other neighborhoods of the solution space.  
 2428

2429     In addition to the higher quality of the results it is worth mentioning the significant reduction  
 2430     of the wall-clock time. Table 6.4 summarizes the time taken for the 3 different methods. A 40X  
 2431     to 50X speed-up is recorded for these dataset sizes and the time saving increases for larger  
 2432     ones.  
 2433

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

**Table 6.4:** 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.

2433 For better comparison, Particle 2 has been reconstructed using a genetic PR approach. Simi-  
 2434 larly to the procedure illustrated in [196], from 15 randomly initialized guessed the following  
 2435 iterative chain was applied:

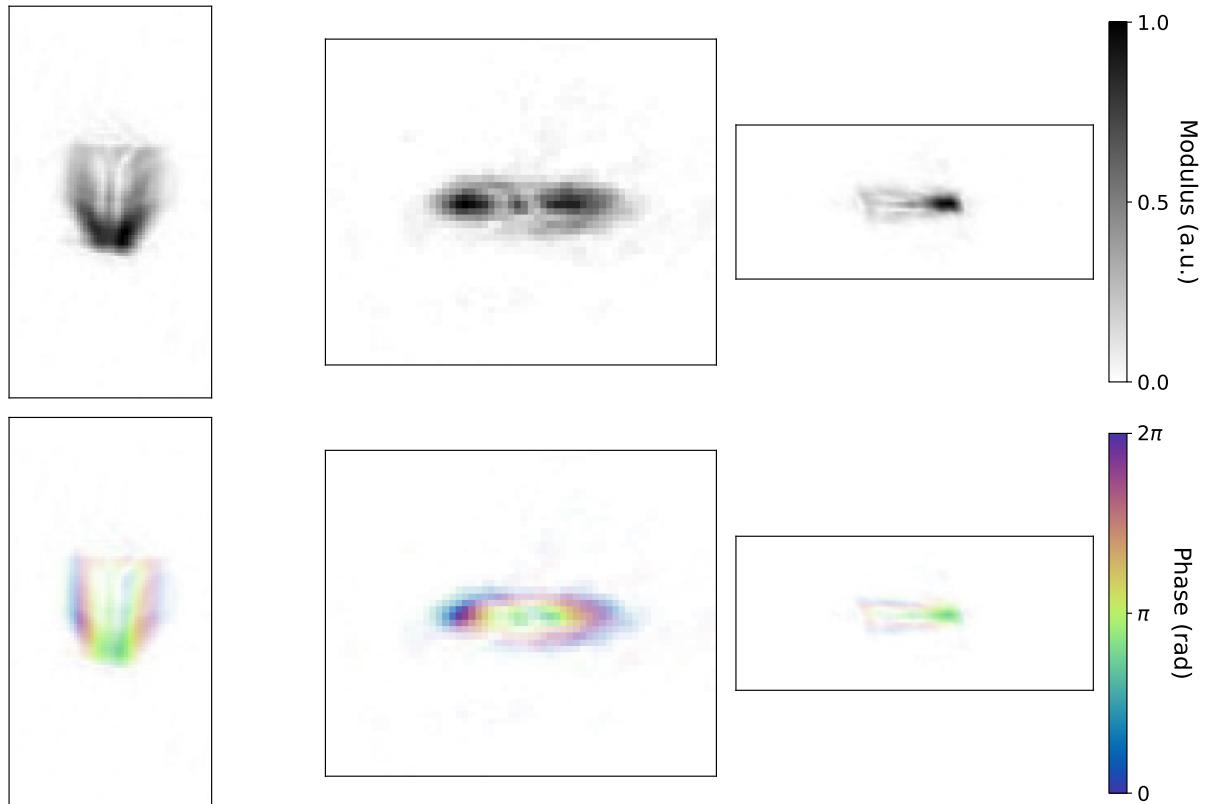
- 2436 • 400 iterations of HIO  
 2437 • 300 iterations of RAAR  
 2438 • 100 iterations of ER

2439 One reconstruction was selected from the population based on a sharpness criterion, defined  
 2440 as the sum over all voxels of the absolute value of the reconstruction raised to the fourth power  
 2441 (see [196]).

2442 The modulus of this selected reconstruction was then used as a seed for the next 10  
 2443 generations of the base chain, with the phase kept unchanged. This update followed Wilkin's  
 2444 approach [104]:

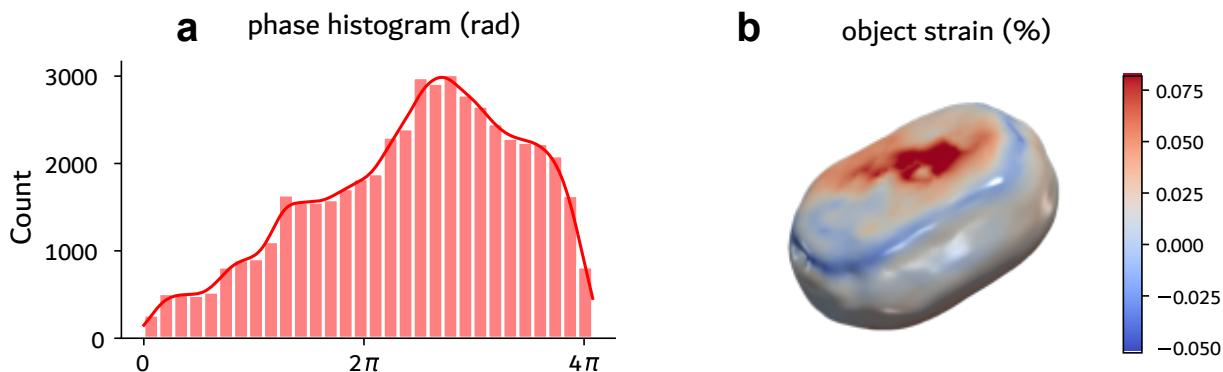
$$\rho = \sqrt{\rho \times \rho_{\text{best}}} \quad (6.6)$$

2445 where  $\rho$  is the modulus of an individual in the population being updated, and  $\rho_{\text{best}}$  is the  
 2446 modulus with the lowest sharpness from the previous generation. The results are shown in Fig.  
 2447 6.49



**Figure 6.49:** Central slices of Particle 2 reconstructed with genetic PR. Although better than Fig. 6.44 the result is far from what obtained with the DL-aided PR.

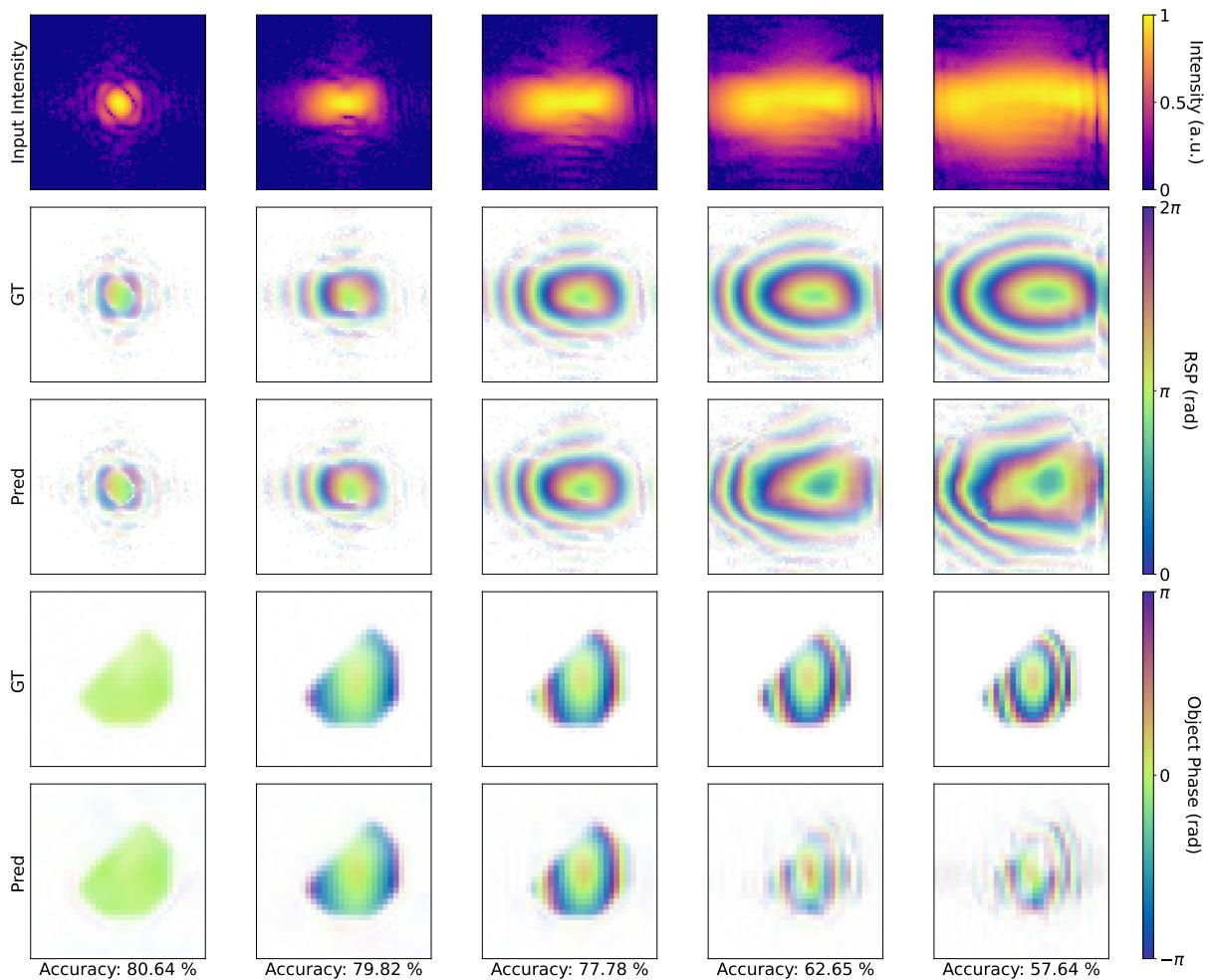
At this point, it is evident that the diffraction pattern of Particle 2 cannot be reliably reconstructed without a DL-based initial estimate. This difficulty is commonly associated with the presence of local minima, which hinder the search for the correct solution and can trap iterative algorithms. An interesting observation is that this behavior can be linked to the phase range of the reconstructed object. Specifically, if the phase values are confined within a single phase wrap, i.e. the interval  $[0, 2\pi]$ , the phase difference between any two voxels is uniquely defined within that domain. In contrast, objects with broader phase ranges typically extend over multiple phase wraps, in which case the phase difference between voxels is not uniquely determined in  $[0, 2\pi]$ . It follows that the larger the range of the unwrapped phase, the higher the “population of local minima”, hence the more challenging the PR. These considerations were already proposed by Minkevich and coauthors in [101]. Fig. 6.50 shows the histogram of the phase range extending over the equivalent of two full wraps of Particle 2 (a) as well as with the strain distribution (b) on the 3D rendering of the orthogonalized reconstruction.



**Figure 6.50: Particle 2.** **a** Histogram representing the range of values of the reconstructed object’s phase. Such broad range is typically referred to as “strong-phase” or “high-strain” and implies challenging PR. **b** 3D rendering of the particle’s strain distribution after orthogonalization. The morphology confirms the Winterbottom shape expected from Pd/Pt on crystalline substrate.

## 2461 6.10 Performance assessment

2462 In this section the results of the DL model presented previously, tested against different  
 2463 strain configurations and magnitudes, are discussed. The scope of the study is to assess the  
 2464 model's performance for these different cases, to evaluate when it works best and ultimately to  
 2465 estimate the gain in accuracy when coupled with ER refinement. The first test was prepared by  
 2466 simulating a Winterbottom shaped particle, similar to the ones used for the training set, with an  
 2467 applied phase built with two Gaussian functions with two different and increasing amplitude  
 2468 ranges. The corresponding BCDI pattern has been simulated for each case, keeping the same  
 2469 oversampling ratio and noise level across the simulations. For each calculated diffraction  
 2470 pattern the RSP has been predicted using the DL model and corresponding objects have been  
 2471 obtained with inverse FT. At this point the accuracy of the prediction was calculated using the  
 2472 formula in 6.5 and the results are shown in Fig. 6.51.

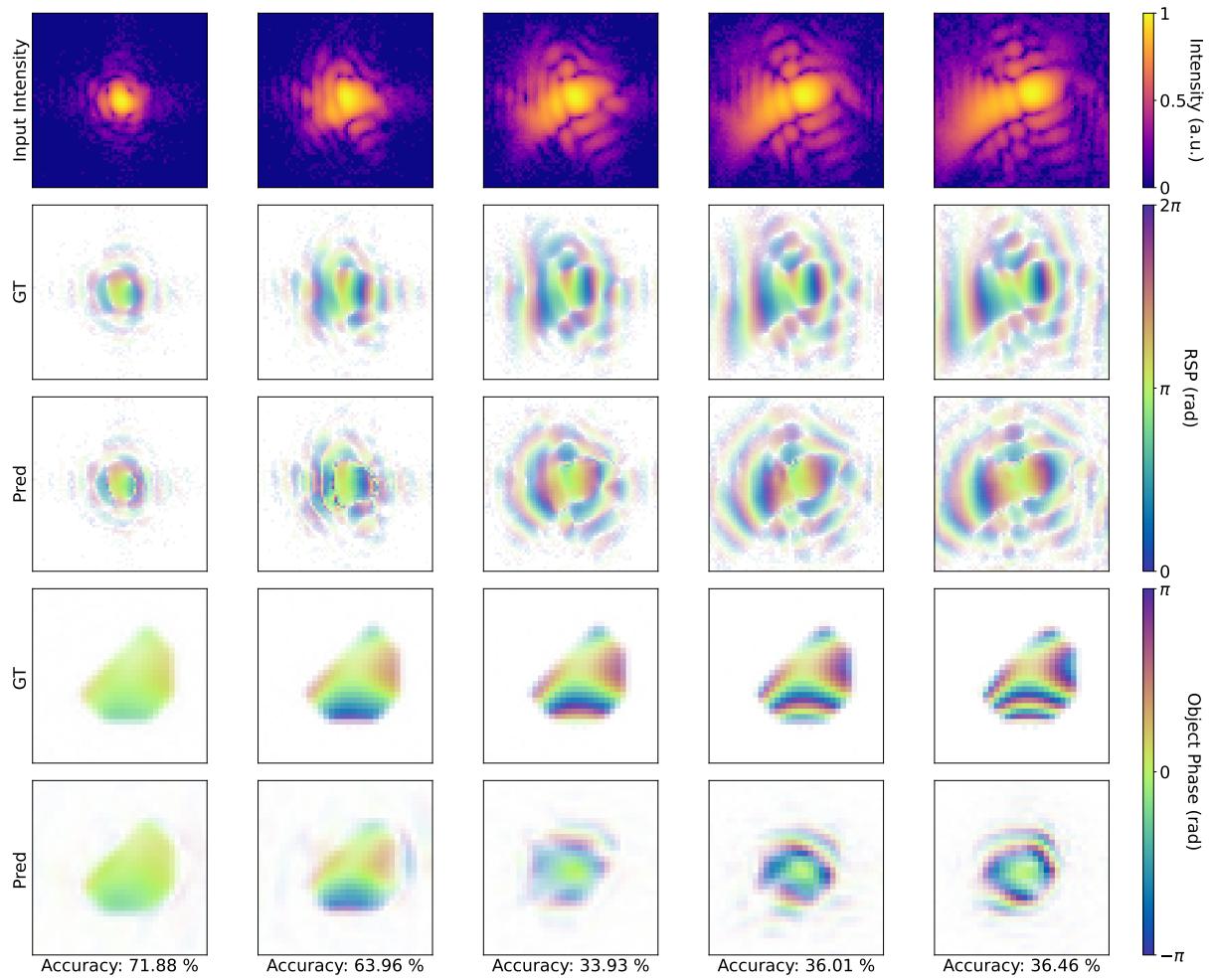


**Figure 6.51:** Evolution of the BCDI pattern and corresponding RSP and object for higher strain calculated with the sum of two Gaussian functions of increasing amplitude. Third and fifth row show the DL predicted RSP and the reconstructed object's phase respectively. As expected the prediction worsens for high phase ranges but it overall maintains a similarity with the ground truth. One could notice the presence of aliasing due to the use of the Fourier transform and large oversampling. Although not observable in experimental conditions, this artifact does not affect severely the analysis and the reconstructions.

2473 As expected the accuracy drops as the phase range increases. It is interesting to notice that  
2474 despite the worsening of the prediction with the increasing strain the object's phase structure  
2475 inside the support resembles the ground truth phase. This detail is fundamental for the ER  
2476 refinement as the initial guess, notwithstanding the inhomogeneous and "shattered" support,  
2477 represents a good estimate of the solution. Objects with similar cleaner supports of incorrect  
2478 shapes and phases are much worse starting points for iterative refinement since they are far  
2479 from the solution.

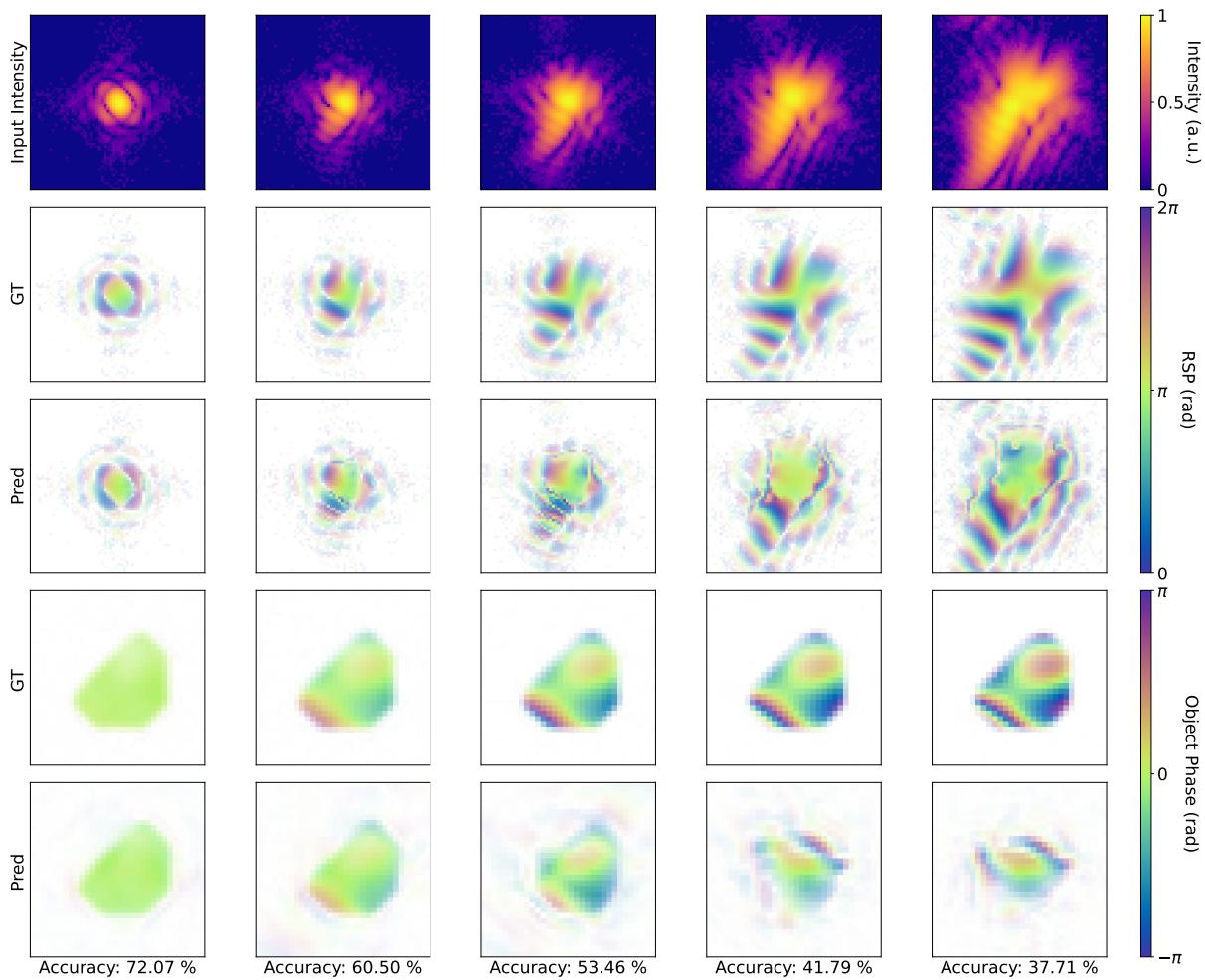
2480

2481 The same procedure has been repeated for a different strain configuration, this time con-  
2482 structed with two cosine functions with increasing amplitude. The phase distribution induced  
2483 by these functions is rather different, more in the spatial structure than the amplitude. Fig.  
2484 6.52 shows that the model accuracy is significantly poorer than the previous case. It is worth  
2485 recalling that the training set was composed of equal amounts of particles simulated with  
2486 Gaussian and cosine phase profiles, meaning that the lower accuracy scores are not due to  
2487 some possible imbalance of the training. The phase range inside the particles of Fig. 6.52, that  
2488 can be estimated visually by counting the number of phase wraps, is also in the same order  
2489 of magnitude as the one shown Fig. 6.51. This further suggests that the poorer performance  
2490 observed in Fig. 6.52 arises from other factors.



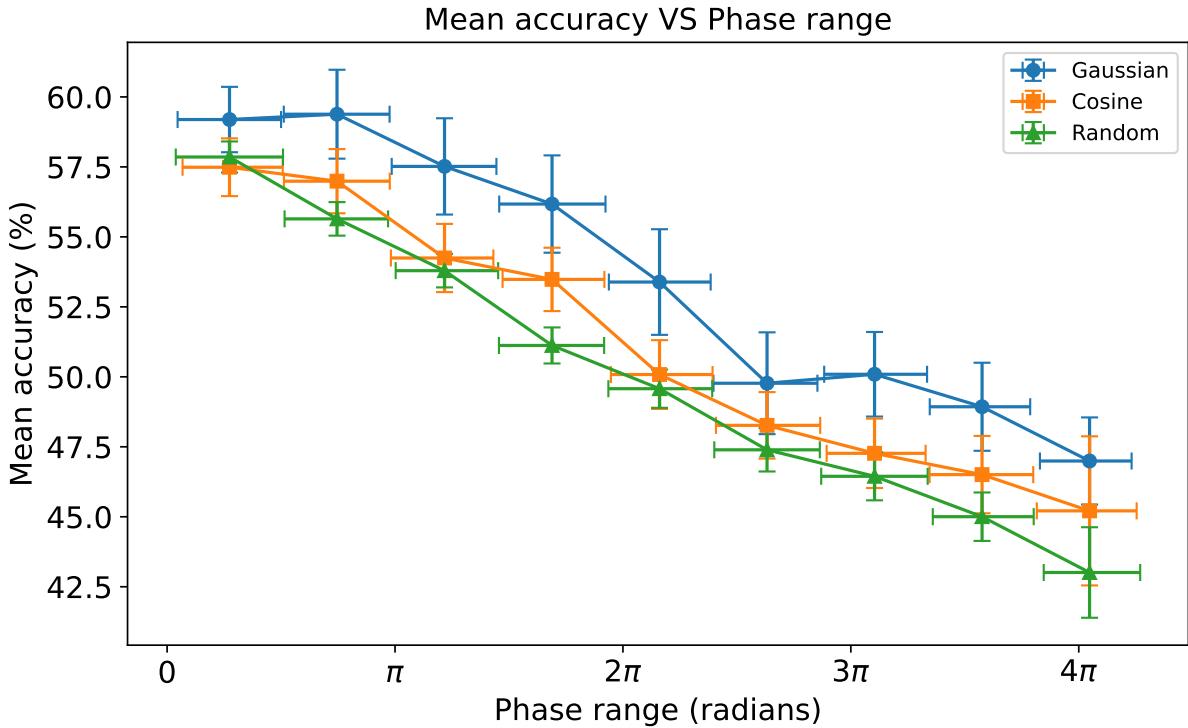
**Figure 6.52:** Evolution of the BCDI pattern and corresponding RSP and object for higher strain calculated with the sum of two cosine functions of increasing amplitude. Third and fifth row show the DL predicted RSP and the reconstructed object's phase respectively. The model rapidly struggles with this type of phase field.

2491 Similarly, the model seems to struggle more for phase fields simulated with using Gaussian  
 2492 correlated random field (same as in 5.4.1) than for the first case. Again the reason doesn't seem  
 2493 to be related to the different phase range nor population imbalance in the training set (Fig. 6.53)



**Figure 6.53:** 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.

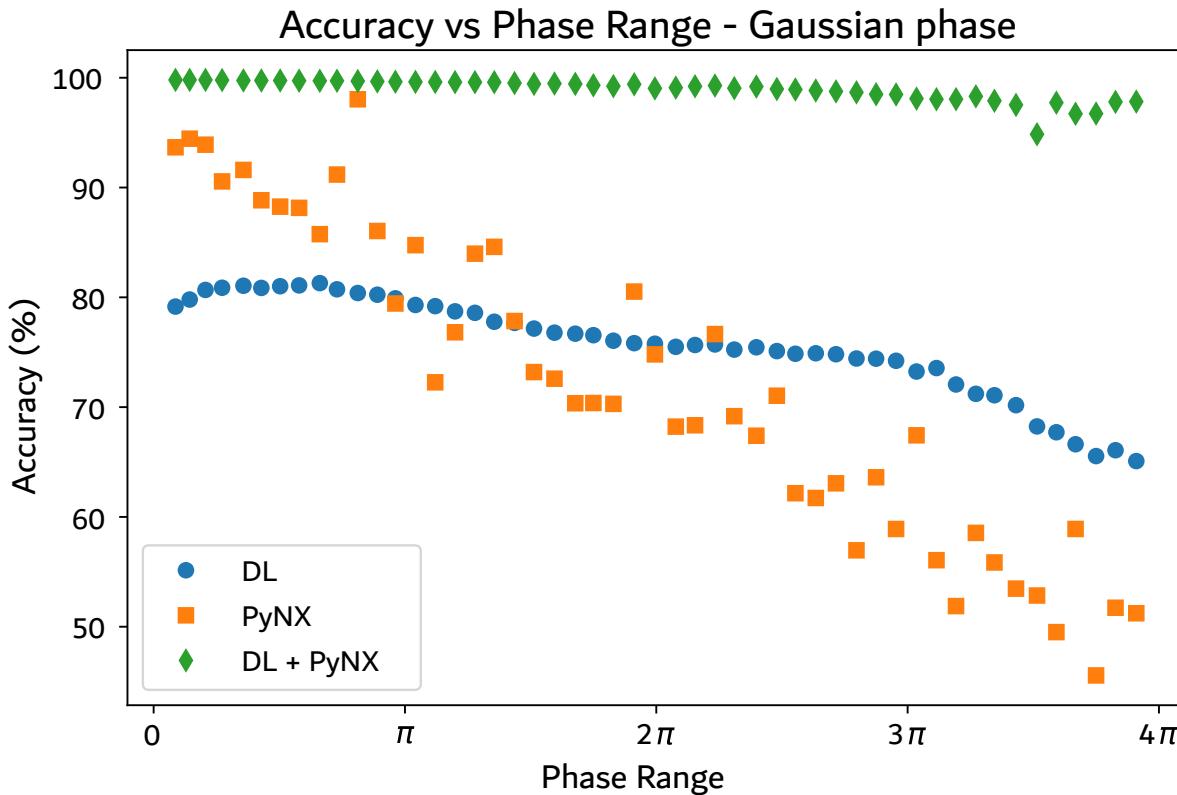
2494 To better assess the performance of the model on the three different cases, the same  
 2495 procedure has been repeated on a bigger and more diverse population. Namely, 50 randomly  
 2496 shaped particles have been simulated and for each of them an increasing strain field has been  
 2497 applied in 10 different steps, for the three types of distributions. The results of the model are  
 2498 shown in Fig. 6.54



**Figure 6.54:** 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. One can see a significant drop of the accuracy with respect to Figs. 6.51 - 6.52 - 6.53 which is attributable to the different random shapes used here instead of the Winterbottom shaped particle that was used for the previous tests. In fact, while the model was trained on equal numbers of Winterbottom-shaped and randomly shaped particles, the diversity among Winterbottom particles is inherently smaller. This lower variability makes them easier for the DL model to learn, which likely explains the higher accuracy obtained previously.

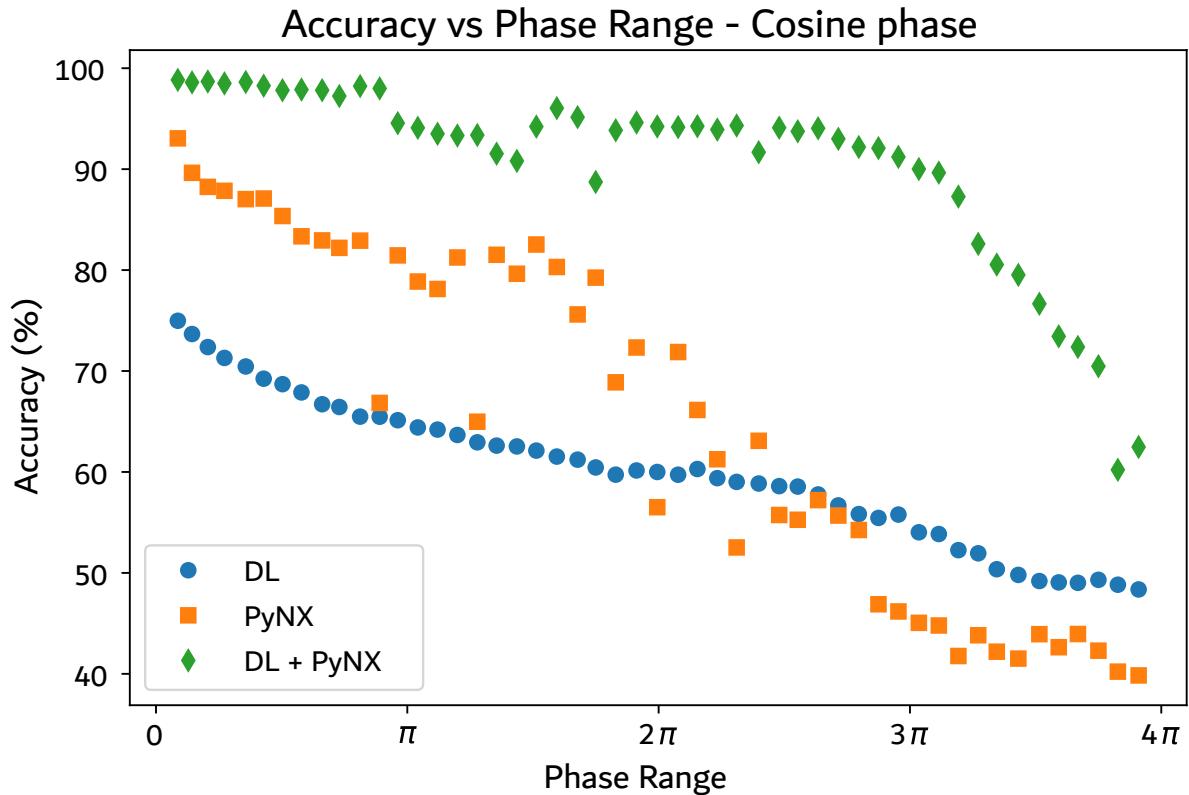
2499 Another study that has been conducted aimed at estimating the accuracy gain of the DL  
 2500 model coupled with ER refinement compared to the sole iterative algorithm. In this case a  
 2501 single Winterbottom particle has been selected for simplicity, and an increasing phase field was  
 2502 applied in 50 steps for the three different types. At this point the accuracy scores have been  
 2503 calculated for the DL model only. Separately, a single run of 400 HIO + 1000 RAAR + 300 ER  
 2504 has been performed using PyNX for each of the 50 diffraction patterns. The parameters are the  
 2505 same reported in Table 6.2, exception made for the autocorrelation and support thresholds fixed  
 2506 at 0.1 and 0.15 respectively. At last, the DL model predicted objects have been used as a starting  
 2507 point for 300 ER refinement, always with PyNX and the parameters in Table 6.3. In all cases, it  
 2508 was made sure that the solution found by either the DL or the iterative algorithm wasn't the  
 2509 twin of the ground truth. If so, the object was flipped before calculating the accuracy.

2510 The analysis has been repeated for the three different phase fields used in the above cases  
 2511 and the results are shown in Figs. 6.55, 6.56, 6.57.



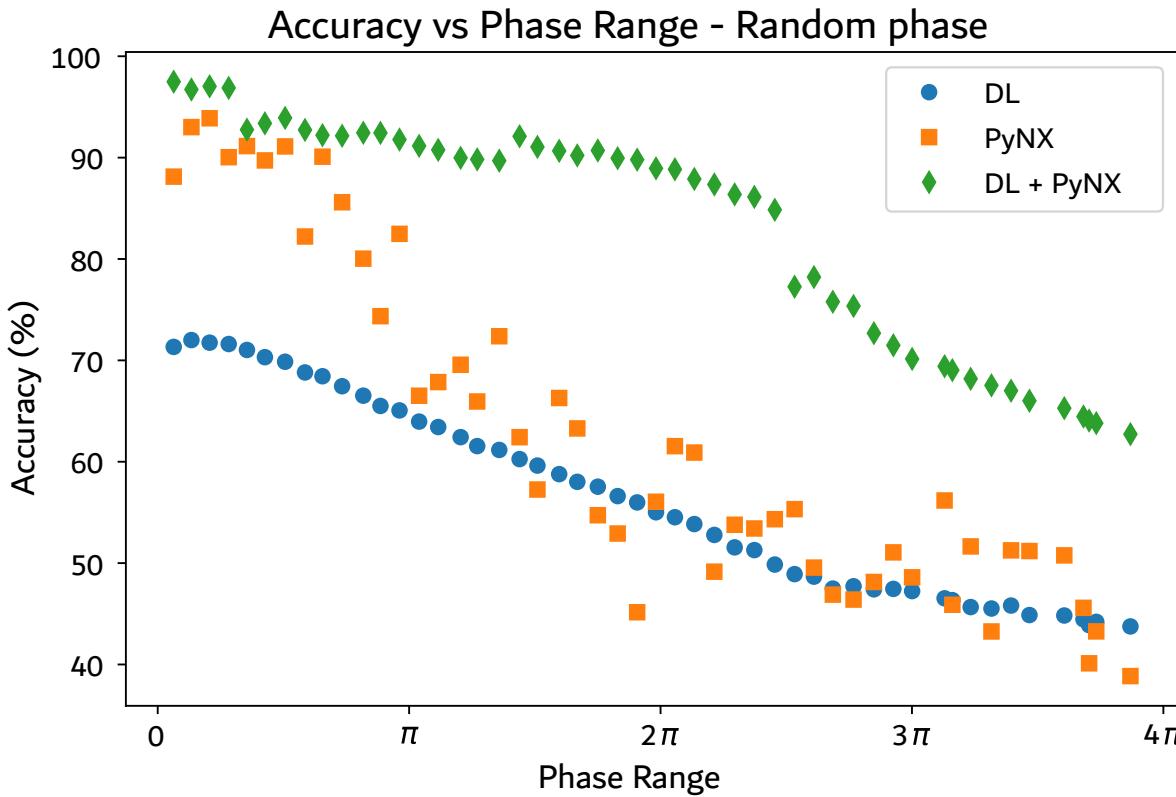
**Figure 6.55: Accuracy vs Gaussian phase range.** Comparison of the accuracy scores of the DL model prediction, iterative PR and DL + ER approaches for objects with phase fields simulated with two Gaussian functions with increasing amplitude. The DL + ER reconstructions always achieve higher accuracies than the other methods, also in those cases in which the DL ones are low.

2512 While for lower phase ranges the DL prediction is worse than the two other methods, the  
 2513 main result of Fig.6.55 is that in all cases the accuracy of the DL + ER method is always superior,  
 2514 meaning that the DL model can significantly aid the PR process also when the first estimate is  
 2515 not very close to the solution (down 50% accuracy in the figure.).



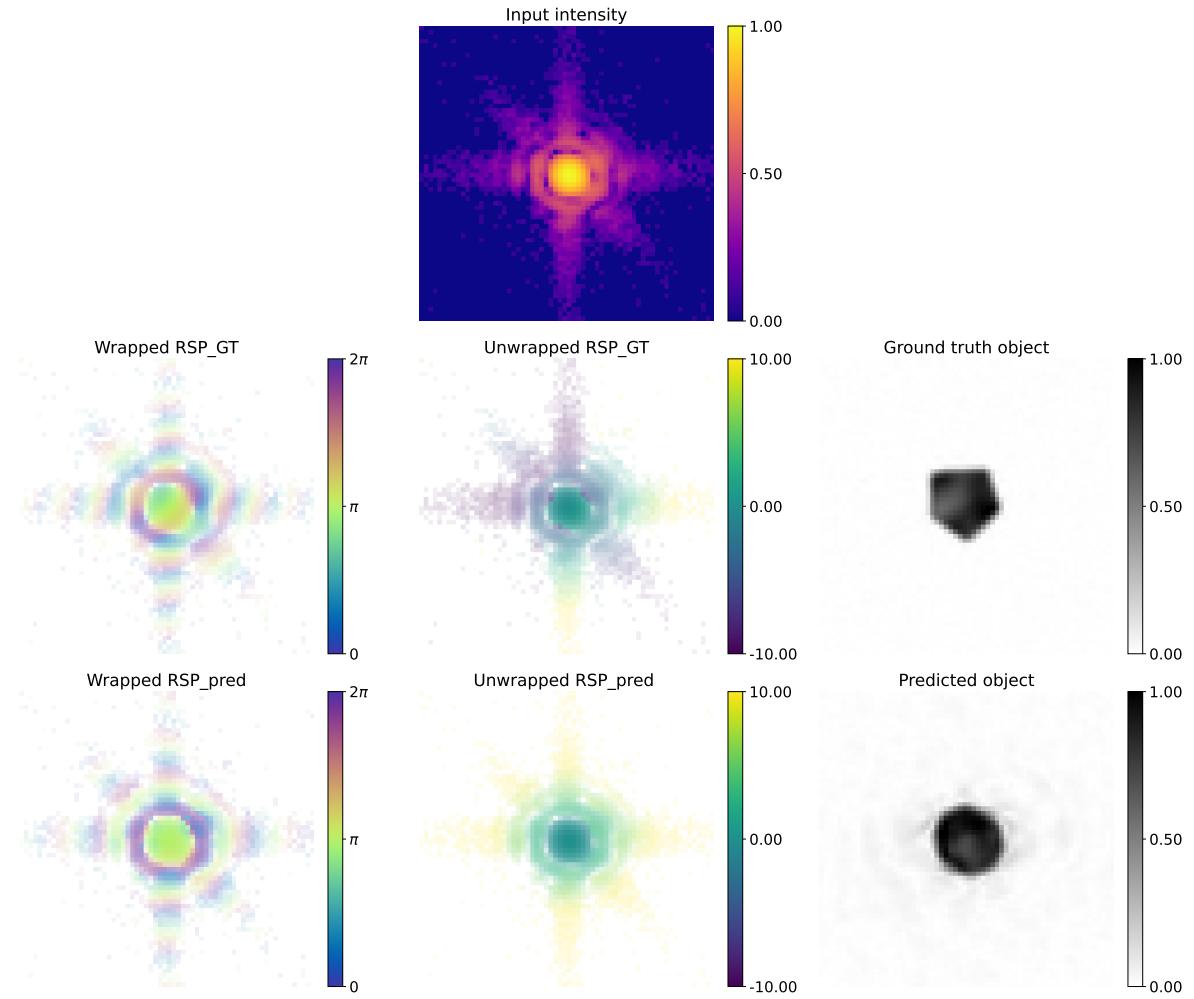
**Figure 6.56: Accuracy vs Cosine phase range.** Comparison of the accuracy scores of the DL model prediction, iterative PR and DL + ER approaches for objects with phase fields simulated with two cosine functions with increasing amplitude. Although PyNX shows better results for most of the phase ranges, the DL + ER combination outperforms PyNX across the whole range.

2516        Similarly, the DL model proves to be significantly helpful at providing a good initial guess  
 2517        for the ER refinement when the object's phase is simulated with cosine functions. In this case as  
 2518        well, the DL model, although when used alone yields relatively low accuracy scores, improves  
 2519        the quality of the solution when coupled with some ER cycles for refinement.



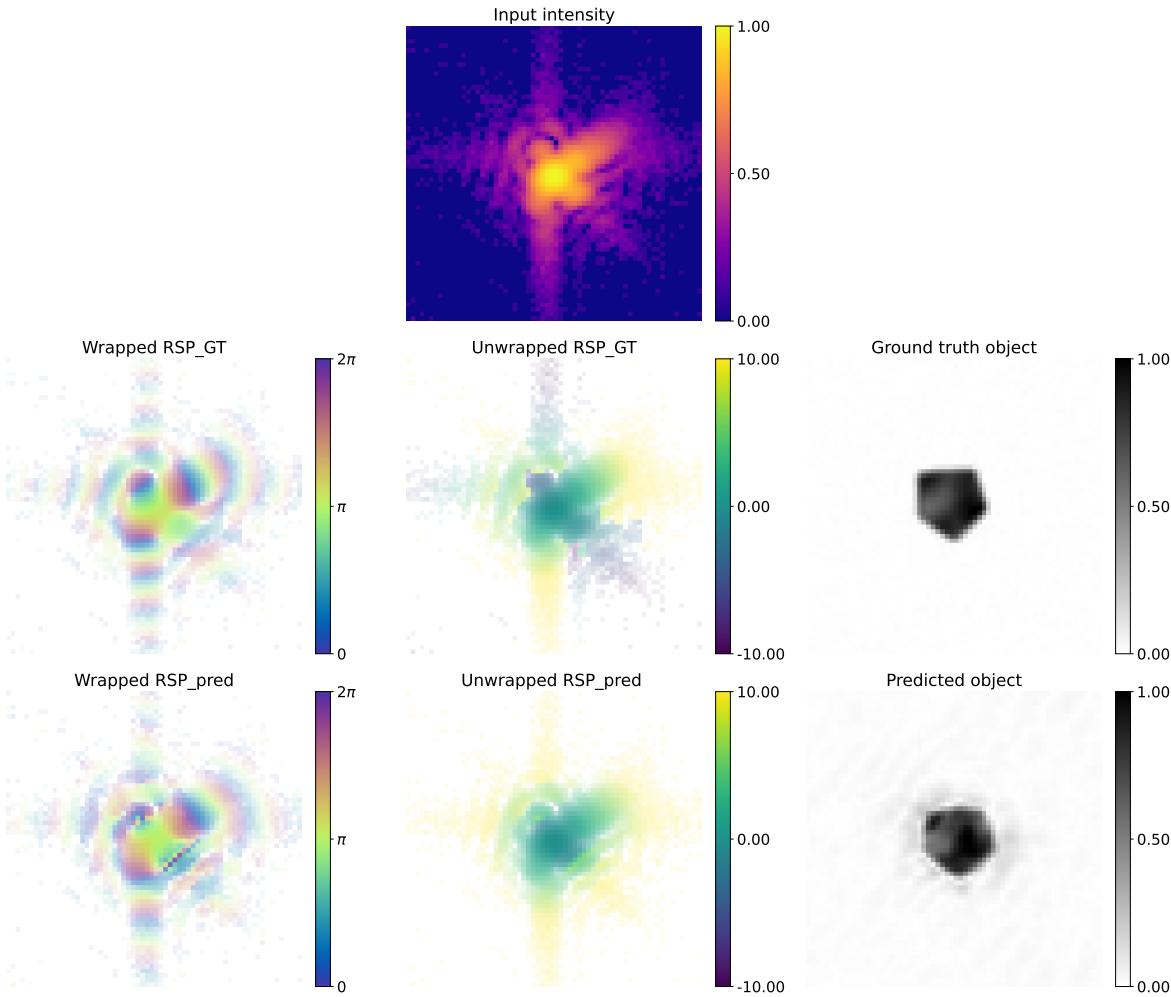
**Figure 6.57: Accuracy vs Random phase range.** Comparison of the accuracy scores of the DL model prediction, iterative PR and DL + ER approaches for objects with phase simulated with a Gaussian correlated random field with increasing amplitude. For small phases PyNX shows better accuracy scores than DL only, however for stronger phases the two methods yield similar results. However, most importantly, the DL + ER combination always outperforms both methods, meaning that the DL result is a close estimate of the solution for which few refinement iterations are enough to converge.

Finally 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 as 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 6.58:** 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 retrieved object as visible in Fig. 6.58. 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. To support this hypothesis, a stronger phase obtained with two Gaussian functions has been applied to the same object and the same test has been conducted. The results illustrated in Fig. 6.59 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. This would explain the initial bump in the accuracy scores present in both Figs. 6.55 - 6.57.

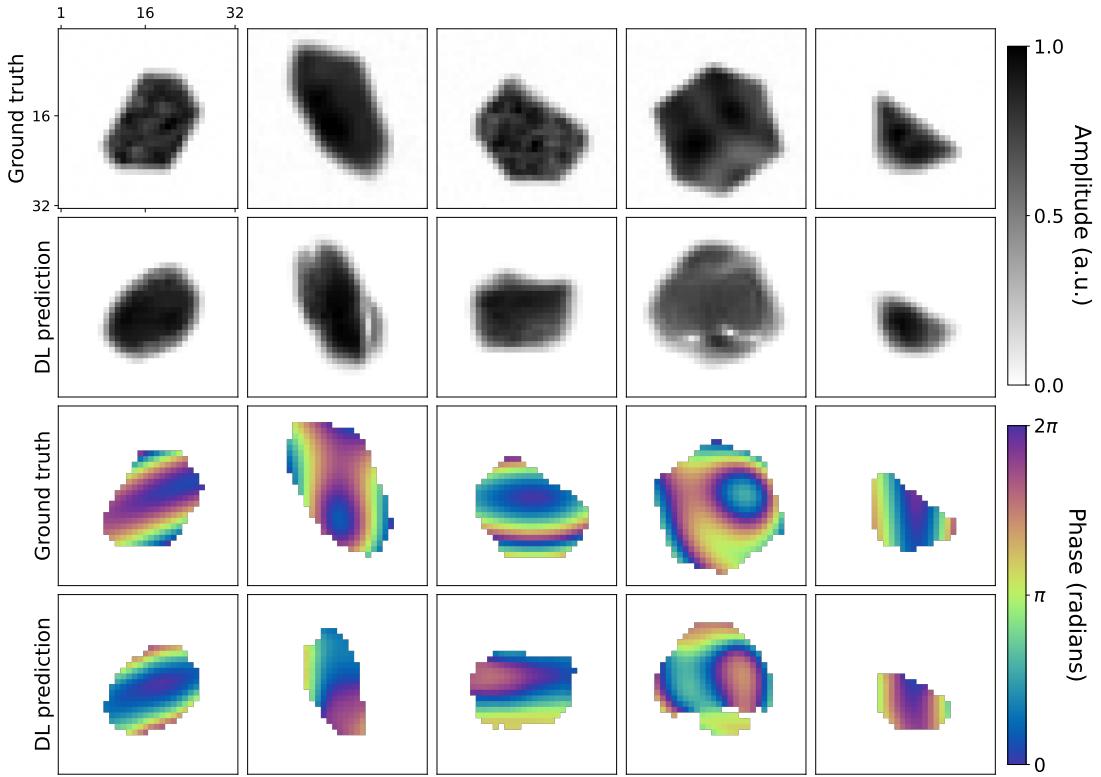


**Figure 6.59:** A stronger phase has been applied to the same object in Fig. 6.58. The strain field produces a more symmetric RSP that is correctly retrieved by the DL model, thus a better object is retrieved despite the higher strain.

## 2542 6.11 Other model test

2543 In this last section I would like to assess the real advantage of predicting the RSP rather than  
2544 the complex real space object. In order to conduct this experiment, the most recent model in  
2545 the literature of Deep Learning for BCDI Phase Retrieval was considered. In particular, the  
2546 model presented by Yu and coauthors in [186], that makes use of complex convolutional layers  
2547 seem to be a good candidate for the testing of our DL model. The 2D model that was presented  
2548 in the paper was transformed into a similar one for 3D data, for a total amount of 36 million  
2549 trainable parameters.

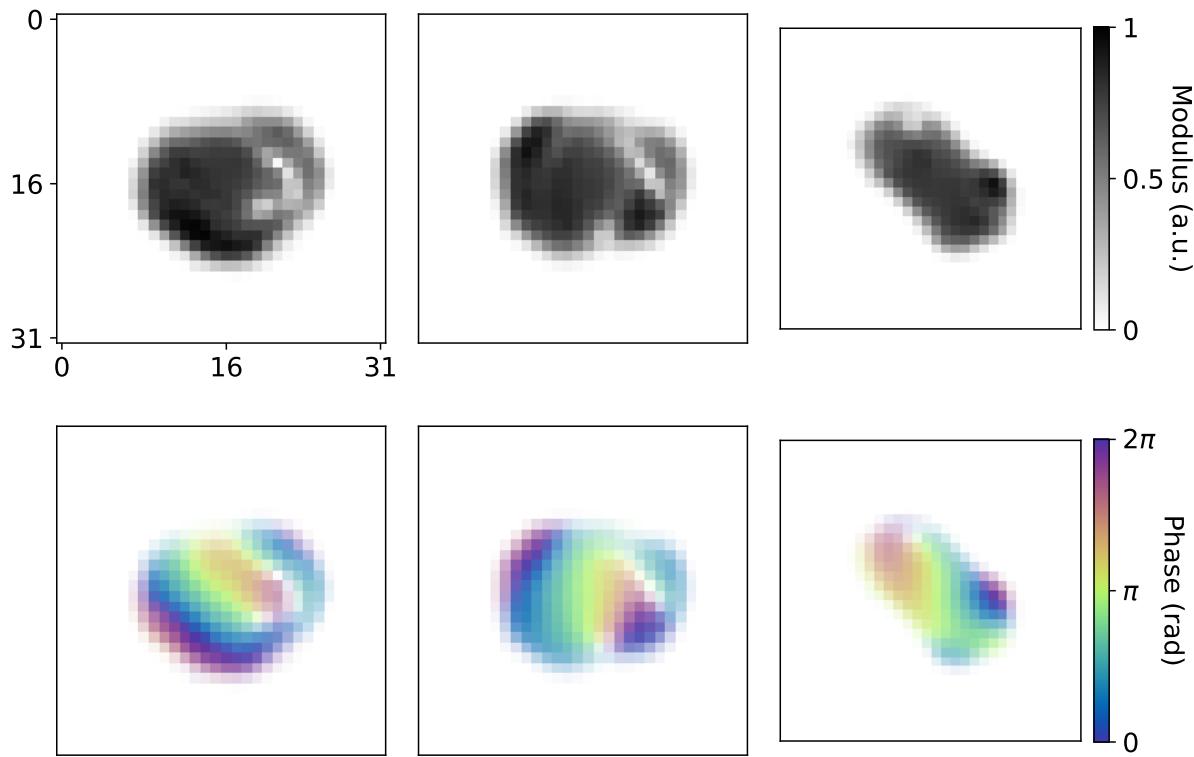
2550 The model has been trained for the same amount of epochs (60) on the same training dataset  
2551 used for our DL model. Model structure and loss function were adopted like those presented  
2552 in the paper, and tests on the same data shown in Fig. 6.37 have been performed with results  
2553 shown in Fig. 6.60.



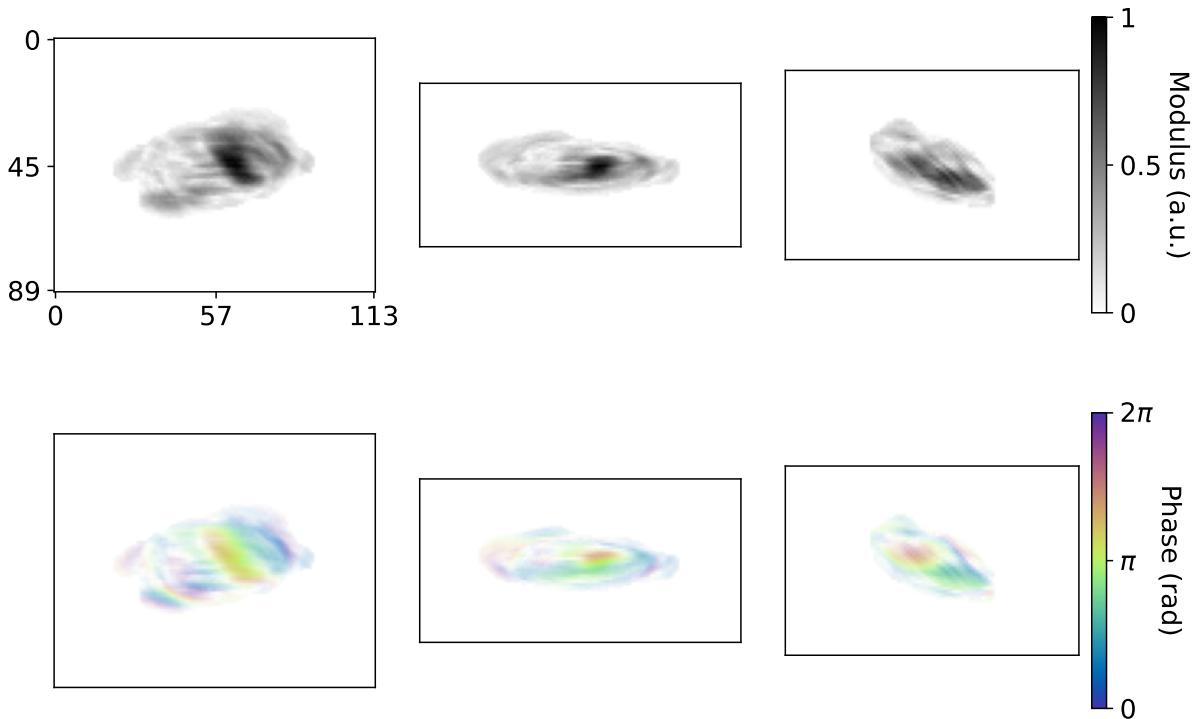
**Figure 6.60:** Predicted objects obtained by the 3D adaptation of the 2D Complex U-Net presented in [186]. The model returns a complex tensor that represents the real space solution. Although much less noisy than our predictions, the reconstructed objects possess incorrect shapes and phases.

2554     The model returns objects that have a better resolution and less noise levels because of the  
 2555 loss function was calculated in real space. However, the shape and phase of the results are not  
 2556 correct, thus unusable for ER refinement because they are misleading initial guesses. In fact,  
 2557 the PR of the diffraction pattern of Particle 1 (Fig. 6.38) has been inferred with this model (see  
 2558 Fig. 6.61) and the result used as a starting point for further refinement using PyNX and the  
 2559 parameters of Table 6.3. The model prediction possesses clean and uniform modulus, and a  
 2560 support which is almost correct. However the phase is quite different from what obtained in  
 2561 6.47.

2562     For this reason, overall, the predicted object does not represent a close enough estimate  
 2563 for ER to converge to the solution. The result instead shows inhomogeneous modulus with  
 2564 amplitude dips and domains in the phase, therefore far from the correct solution.



**Figure 6.61:** Three central slices of the prediction of Particle 1 using the DL model presented in [186] adopted for the 3D case and trained on highly strained BCDI data. The retrieved object is clean and homogeneous but the phase has been revealed to be incorrect.



**Figure 6.62:** PR of Particle 1 obtained with PyNX refinement following Table 6.3 of the DL prediction in Fig. 6.61. The results show that the convergence to the correct solution was not achieved because the DL estimate is too far from it.

2565 This simple example shows that the prediction of the RSP suites better the PR of BCDI  
2566 patterns with convolutional neural networks, especially for highly strained particles.

## 2567 6.12 Conclusion

2568 To conclude this journey through the developments, results and interpretations of the DL model  
2569 for the BCDI RSP prediction the main points can be summarized as follows.

- 2570 • A novel approach for the DL-based PR of BCDI patterns, focused on the prediction of  
2571 the RSP has been investigated. The method proved to be advantageous since (i) it entails  
2572 the inference of a single array rather than two coupled ones, (ii) it exploits the exact  
2573 calculation of the IFT, directly using the diffraction pattern in the transform, rather than  
2574 a learned map from the diffraction pattern and the solution in real space. (iii) it takes  
2575 advantage of the similarity between the input and the output variables, making the best  
2576 use of the skip connections of the U-Net architecture. The superiority of this approach  
2577 over the more conventional one found in literature has been demonstrated as discussed  
2578 in 6.11.
- 2579 • The training of the DL model for the prediction of the RSP can be optimized with the  
2580 use of the custom WCA loss function, designed specifically for the handling of complex  
2581 phases. This loss function naturally resolves the wrap, the sign and the offset symmetries  
2582 inherent to the lost phase problem. Adopting the WCA loss not only improves the results

2583 but also speeds up the training as it takes less time than a loss computed in real space.  
2584 Moreover, the use of the WCA avoids any reference to the real space object during the  
2585 training, opening the door for a patching approach, where the RSP is predicted from a  
2586 sub-volume of diffracted intensity.

- 2587 • Such a tailored model is trained in supervised fashion on simulated data only. This can be  
2588 at first a limiting factor as it requires the simulation of big and diverse datasets. However,  
2589 as the BCDI technique is restricted to single crystals, the population of shapes, though  
2590 being potentially infinite, is physically confined by nature. It is thus easier for the DL  
2591 model to generalize for new particle shapes.
- 2592 • The DL model trained as presented is able to perform on highly-strained particles as  
2593 well, showing promising results in 2D and 3D. It has proven to be successful at inverting  
2594 experimental data that happened to be extremely challenging with conventional iterative  
2595 methods only. This achievement marks a milestone for the BCDI community, upgrading  
2596 the DL studies on BCDI PR from a “*proof of concept*” level to an actual *practical use* in the  
2597 analysis of experimental data.
- 2598 • The DL model exhibits certain shortcomings attributable to the intrinsic symmetry of  
2599 RSP. It in fact “prefers” RSPs with a spherically symmetric distribution and the major  
2600 hypothesis for this fact is that such symmetry is typical of the intensity distribution of  
2601 the average diffraction pattern. Hence, the increased difficulty of the model to escape  
2602 this bias for 3D data as compared to 2D data. Additionally, it is curious to notice that  
2603 this symmetry seems to be the strongest limitation for the DL model, lying above the  
2604 sign symmetry and the high strain, which are both resolved when in many cases. Future  
2605 investigations that elucidate the root causes of this phenomenon may yield improved  
2606 performance on more heterogeneous data sets.
- 2607 • Lastly, the DL model proved to be compatible with iterative refinement using conventional  
2608 algorithms, contributing to a faster and more robust pipeline for PR. The computational  
2609 time and costs typically devoted to the search of the global minimum with regular  
2610 algorithms is now significantly reduced by the DL estimate. This can potentially broaden  
2611 the field of application of BCDI to many more samples in which high-strain is involved  
2612 as cause or consequence of physically relevant mechanism.

2614    **AUTOMATIC DIFFERENTIATION FOR PHASE RE-**

2615    **TRIEVAL**

“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”

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J.R.Fienup [2]

2616    In this chapter a different approach to the BCDI phase retrieval will be presented. It origi-  
 2617    nated from the need to resolve those cases in which neither standard iterative algorithms, nor  
 2618    the DL assisted PR can succeed to converge to a satisfactory reconstruction. The developed  
 2619    approach differs from the alternating projections algorithms classically used for the Fourier PR,  
 2620    as it is formulated as minimization problem solved with gradient descent (GD). The gradients  
 2621    however are computed through the efficient automatic differentiation (AD) enabled by graph-  
 2622    based differentiable programming packages like Tensorflow and PyTorch, accelerated on GPU.  
 2623    For this reason one could see the AD approach as unsupervised machine learning on a single  
 2624    training dataset.  
 2625    Gradient descent-based optimization differs fundamentally from fixed-point alternating pro-  
 2626    jections. In qualitative terms, while the latter alternates between real and reciprocal space,  
 2627    imposing constraints in each, the former begins with an initial complex object and iteratively  
 2628    updates its modulus and phase using the gradients of the mismatch between the measured and  
 2629    calculated diffraction intensities. In this way, the knowledge on the particle can be implemented

2630 by initializing the object with some physical constraints or adding regularization terms that  
2631 will drive the updates towards more reasonable solutions.

2632

## 2633 7.1 State of the Art

2634 AD methods for PR have been investigated already in 2014 by Jurling and Fienup [197] who  
2635 first considered the use of AD for GD-based PR. In this theoretical work the authors proposed  
2636 a pedagogical “manual automatic differentiation” approach for the phase problem, extended to  
2637 complex-valued variables. The authors also denounced the lack of suitable softwares as major  
2638 limitation to the use of AD-based PR. The advent of high-level, GPU oriented, libraries such as  
2639 Tensorflow, PyTorch, JAX and Autograd has opened the opportunity to efficiently exploit AD  
2640 algorithms for the phase problem. The first implementations in the CDI field have considered  
2641 mostly ptychography in forward and Bragg geometries [198, 199] and multi-Bragg CDI [200].  
2642 Chronologically, it was firstly Nashed and coauthors in 2017 [198] who opened the field using  
2643 a Tensorflow AD model for ptychography using the ADAM optimizer. From the same group,  
2644 Kandel *et al.* in 2019, showed the competitive performance of the AD model when compared  
2645 to conventional algorithms and extended the model to multi-angle Bragg ptychography on  
2646 simulated data. In 2023 Maddali and coauthors [200] explored the use of AD methods for multi  
2647 reflection Bragg CDI. The authors leverage the flexibility of the GD approach by designing a  
2648 global optimization function that simultaneously accounts for the geometrical and physical  
2649 constraints related to multi reflection BCDI. More recently Zhou in 2024 [201] and Wu in 2025  
2650 [202] developed AD-based PR algorithms that are able to reconstruct large particles, for which  
2651 a dynamical description of the scattering processes is required. These works again exploit  
2652 the flexibility of AD-based models for the implementation of a forward model tailored to the  
2653 specific problem of mixed kinematic and dynamic x-ray scattering taking place in large crystals.

2654 At the moment of writing, there aren’t any published works that aim at solving the phase  
2655 problem for BCDI datasets considered hard to invert.

## 2656 7.2 Model implementation

2657 In an AD-driven optimization problem some trainable parameters are initialized. In the first  
2658 basic formulation these trainable parameters can be the values of the voxels corresponding  
2659 to the modulus  $\rho$  and the phase  $\phi$  of the complex object that represents the solution of the  
2660 PR problem. All of these voxels contribute to the creation of a simulated diffracted intensity  
2661 pattern via the forward model  $I_{calc} = |\mathcal{F}\{\rho e^{i\phi}\}|^2$ . Subsequently, the gradients of a metric  
2662 (loss function) that estimates the distance between the observed BCDI pattern  $I_{obs}$  and  $I_{calc}$  are  
2663 calculated with respect to each of the trainable variables with automatic differentiation. At  
2664 this point the value of each of these voxels is updated using a chosen optimizer (SGD, ADAM,  
2665 etc.) and a given learning rate. The Tensorflow library allows for an easy implementation of  
2666 the trainable variables, and loss function, and handles gradient operations with predefined  
2667 methods. It is therefore straightforward to run the optimization as it follows the same structure  
2668 of a deep learning model, with less trainable parameters and for a single dataset.

2669 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 as Fourier phase retrieval. In fact, many non-physical modulus-phase configurations could yield an  $I_{calc}$  that is close to  $I_{obs}$  [195]. 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 [85] that steepest GD and even more sophisticated conjugate GD are more prone to get trapped in local minima, which is 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 [122]). 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. The current model design however, does not include any stochastic gradient since the derivatives are calculated for all the data points at each step. The use of ADAM optimizer in this case is therefore advantageous for the first and second order momenta which direct the updates. Future developments of the model could explore the training using structured mini-batches similarly to what Wu and coauthors have done in [202].

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.

### 7.2.1 Object’s shape

The formulation of the object’s shape starts 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, a successful reconstruction shows a *homogeneous* modulus, sometimes quantitatively assessed through the mean-to-max metric<sup>1</sup> [193, 194]. Enforcing a homogeneous modulus by construction limits the search space and helps the convergence to the solution<sup>2</sup>. 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 [179], 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.

---

<sup>1</sup>as in standard BCDI the form factor is approximated uniform across all the scattering sites

<sup>2</sup>This approach of constraining the modulus of the object to be homogeneous was already considered in the literature (see [59, 101, 102])

According to this scheme the relevant parameters to be optimized are the angles  $\theta$  and  $\varphi$  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  $(\theta, \varphi)$  that its orientation in space (Eq. 7.1). 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 as support (Eq. 7.2).

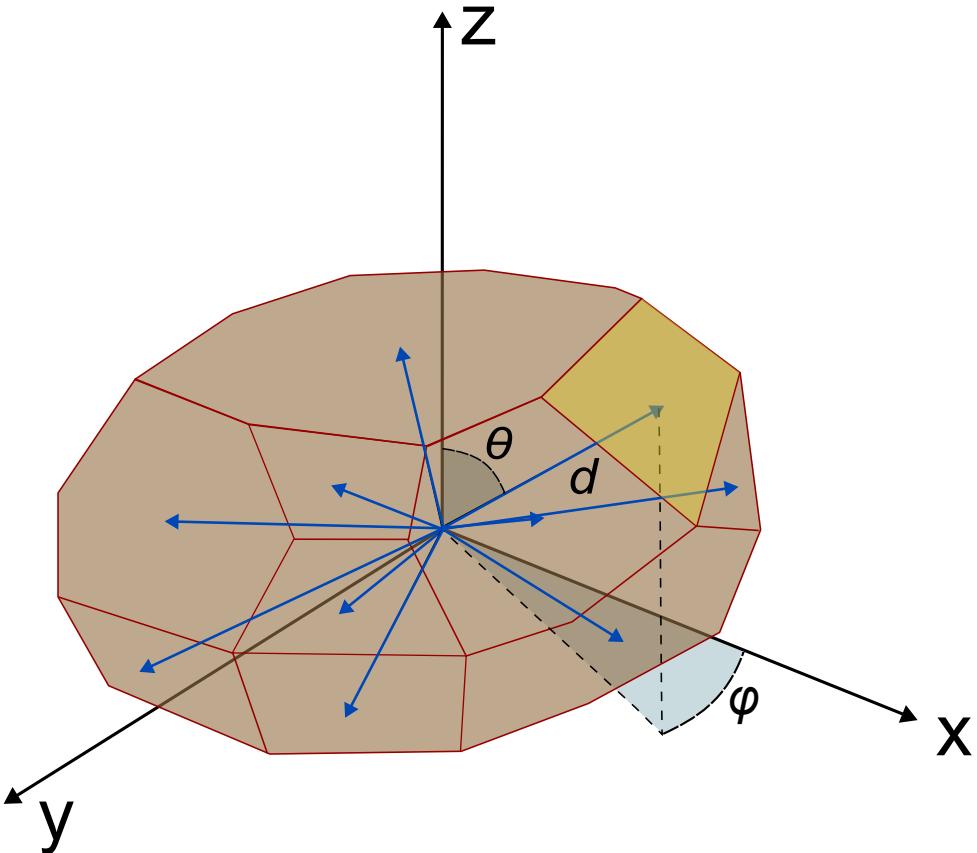
$$\mathbf{n}_i = \begin{pmatrix} \sin \varphi_i \cos \theta_i \\ \sin \varphi_i \sin \theta_i \\ \cos \varphi_i \end{pmatrix}, \quad (7.1)$$

$$\mathcal{S} = \bigcap_{i=1}^N \left\{ \mathbf{x} = (x, y, z) \in \mathbb{R}^3 : \mathbf{n}_i \cdot \mathbf{x} \leq d_i \right\}, \quad (7.2)$$

A schematic representation of this construction is provided by Fig. 7.1.

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. Although these cases are less frequent in typical BCDI experiment, a future development of the models should include the parametrization of concave supports as well. 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 [203]. 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 7.1:** Construction of the convex hull with half-spaces expressed with spherical coordinates. A number  $N$  is given by the user, and a set of  $N$  half-space vectors (blue arrows) are initialized with random length  $d$  and orientation in space  $(\theta, \varphi)$ . The convex-hull obtained with the intersections of the planes built from each half-space vector makes the object support.

2732     The last important consideration of this parametrization is that the support  $\mathcal{S}$  is sharply  
 2733     divided into a binary variable (1 inside and 0 outside) thus leading to differentiability problems.  
 2734     In fact, in such a way the gradients, essential for the support update, are not defined. For  
 2735     this reason  $\mathcal{S}$  is first passed through a sigmoid function controlled by a hyperparameter  $\epsilon$   
 2736     responsible for the smoothening of the support borders. This measure can also be seen as a  
 2737     control of the resolution of the object. Additionally, a mildly steep sigmoid in the early stage of  
 2738     the optimization can help retrieving a low resolution estimate of the support, that can  
 2739     be further refined adjusting the  $\epsilon$  parameter.

2740

### 2741     7.2.2 Object's phase

2742     The parametrization of the object's phase is more challenging. From a qualitative point of  
 2743     view, the prior knowledge that can be exploited for a tailored implementation, is limited to  
 2744     the awareness that a physically meaningful atomic displacement field cannot have "too many"  
 2745     sharp variations. This observation is translated into code by smooth functions parametrization  
 2746     or total variation (TV) regularization [204] of the object's phase. While the former would en-  
 2747     force smoothness by construction the latter would operate adding a penalty to the data-fidelity  
 2748     term of the loss function for non-smooth solutions. Both approaches have been explored and

2749 are reported here.

2750

2751 Forcing a scalar field defined on an  $L \times H \times W$  grid to exhibit smooth behavior is equivalent  
2752 to seeking a sparse representation of that field—that is, to concentrating its essential information  
2753 into far fewer degrees of freedom than the original  $L \times H \times W$  samples. Concretely, one looks  
2754 for a change of basis in which the field can be written as a linear combination of a hierarchy  
2755 of modes or atoms, ordered by “importance”. In a Fourier or wavelet expansion, for instance,  
2756 the expansion coefficients are naturally sorted from largest (low-frequency or coarse-scale  
2757 modes) to smallest (high-frequency or fine-scale modes). Retaining only the largest coefficients  
2758 both compresses the data and removes rapid oscillations, yielding an inherently smoother  
2759 reconstruction. Equivalently, in the matrix case a Singular Value Decomposition (SVD) identifies  
2760 an orthonormal basis in which only a few singular values are nonzero; by truncating to the  
2761 top singular values one obtains a low-rank—and thus smoother—approximation [205]. For  
2762 higher dimensional data, this same principle underlies higher-order generalizations of the  
2763 SVD—Tucker/HOSVD, CP, Tensor-Train, and T-SVD—each of which orders multilinear “modes”  
2764 by their singular-value (or eigenvalue) strength, and truncating to a small subset produces both  
2765 compression and smoothness [206].

2766 In this case the Tucker decomposition was chosen, among the several possible methods,  
2767 for its simplicity of implementation with the Tensorflow library and for the suitability for  
2768 moderately low dimensions [207]. For a 3D tensor the Tucker decomposition is done as follows:

2769 Considering  $\phi \in \mathbb{R}^{L \times H \times W}$  the 3D object’s phase. The Tucker decomposition expresses  $\phi$   
2770 as:

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

2771 where:

- 2772 •  $\mathcal{G} \in \mathbb{R}^{R_1 \times R_2 \times R_3}$  is the **core tensor**,  
2773 •  $U^{(1)} \in \mathbb{R}^{I \times R_1}$ ,  $U^{(2)} \in \mathbb{R}^{J \times R_2}$ , and  $U^{(3)} \in \mathbb{R}^{K \times R_3}$  are the **factor matrices**,  
2774 •  $\times_n$  denotes the mode- $n$  tensor-matrix product.

2775 In index notation, this becomes:

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

2776 With this formulation the parameters  $R_1, R_2, R_3$  are set by the user and define the “storage  
2777 space” in which the information required to represent  $\phi$  has to be condensed. The trainable  
2778 parameters are all the components of  $\mathcal{G}, U^{(1)}, U^{(2)}, U^{(3)}$  therefore scaling from  $\mathcal{O}(L \times H \times W)$   
2779 of a grid-like parametrization to  $\mathcal{O}(R_1 \times R_2 \times R_3 + R_1 \times L + R_2 \times H + R_3 \times W)$ , which is  
2780 also convenient in terms of memory for small core tensors. It is proven that for  $R_i = L, H, W$   
2781 respectively, the tensor  $\phi$  is exactly represented but in practice, for typical BCDI datasets of  
2782  $200 \times 200 \times 200$  pixels a cubic 8 pixels sided  $\mathcal{G}$  is sufficient to represent the phase  $\phi$ . Note,  
2783 that in the general formulation of the Tucker decomposition the  $U^{(1)}, U^{(2)}, U^{(3)}$  are not strictly  
2784 unitary as for the matrices  $U, V$  in the SVD decomposition. The Tensorflow implementation

2785 of the Tucker decomposition is thus rather straightforward as the function `tf.einsum()`  
 2786 takes care of the tensor contraction.

2787

2788 A different approach that has been considered, leverages the TV regularization to push the  
 2789 algorithm towards a smooth object phase. The full  $L \times H \times W$  tensor is therefore optimized  
 2790 and a penalty on the sum of the absolute value of the gradients of the phase is added to the loss  
 2791 function. Precisely, the formula that has been used calculates the sum of the *squared* gradients,  
 2792 since the square root operation, necessary to obtain the correct formula, creates a problem  
 2793 around zero because of the infinite gradient. The final equation is therefore:

$$TV = \alpha \sum_{i=1}^L \sum_{j=1}^H \sum_{k=1}^W \mathcal{S}[(\varphi_i - \varphi_{i-1})^2 + (\varphi_j - \varphi_{j-1})^2 + (\varphi_k - \varphi_{k-1})^2] \quad (7.3)$$

2794 where  $\alpha$  is a hyperparameter that acts as a scaling factor,  $(i, j, k)$  are the indices running  
 2795 over the coordinates of the  $L \times H \times W$  grid and  $\mathcal{S}$  is the object support. The parameter  $\alpha$  in  
 2796 this case was chosen to be assigned relative to a fraction  $\alpha_0$ , chosen by the user, of the TV loss  
 2797 with respect to the data fidelity loss. In such a way, the TV penalty is rescaled with respect to  
 2798 the data loss. Typical values of  $\alpha_0$  range between (0.01, 0.9). Further developments could aim  
 2799 at finding adaptive formulations for the magnitude of  $\alpha$ .

### 2800 7.2.3 Loss function

2801 Another important aspect of the model is the loss function. Typically, for inverse problems there  
 2802 is a *data fidelity* term that in this case measures the distance between  $I_{obs}$  and  $I_{calc}$  according  
 2803 to some metric, and other additional *regularization* terms that guide the optimization process  
 2804 with physical constraints.

2805 **Data fidelity:** The most common and intuitive metrics are the Mean Squared Error (MSE)  
 2806 and the Mean Absolute Error (MAE) that evaluate the Euclidean distance between the observed  
 2807 and calculated intensities. Practically, because of the large dynamic range of typical BCIDI data,  
 2808 the MAE performs better as it doesn't focus on bright pixels only, but manages to correct for  
 2809 lower intensity tails as well. A more faithful metric for BCIDI experimental data is the Poisson  
 2810 Negative Log-Likelihood (P-NLLK). This metric assumes indeed the handling of count data,  
 2811 like the type obtained by photon counting detectors, and that the stochasticity of physical  
 2812 process is Poisson distributed. When summed over the full dataset, the discrepancies between  
 2813 calculated and observed intensities are not intended as Euclidean distances but like divergences  
 2814 between two probability distributions. In other words, the P-NLLK estimates the likelihood  
 2815 that  $I_{calc}$  belongs to the same Poisson distribution of  $I_{obs}$  [208]. Derived from the equation for  
 2816 the probability for Poissonian events, the formula of the averaged P-NLLK, in the form of a  
 2817 Kullback-Liebler divergence is:

$$\langle P - LLK \rangle = \frac{2}{N_{obs}} \left[ \sum_{I_{obs} > 0} \left( I_{calc} - I_{obs} + I_{obs} \ln \frac{I_{obs}}{I_{calc}} \right) + \sum_{I_{obs} = 0} I_{calc} \right]. \quad (7.4)$$

2818 Both the MAE and the P-NLLK have been tested on several simulated and experimental

2819 datasets and the MAE has always shown better convergence. An explanation for this unexpected  
2820 result is yet to be found, but the main suspect is that the gradients calculated during the back-  
2821 propagation can have instabilities because of the logarithm.

2822 **Regularizations:** Beside the TV on the object's phase to ensure smoothness, another term  
2823 that was considered concerns the size of the support. For a given data fidelity value, it is known  
2824 that the object with the smallest support represents the optimal solution [195]. Large supports  
2825 can induce the model to overfit the observed intensity. Intuitively this could be explained with  
2826 the fact that there are many more object phase configurations that would combine constructive  
2827 and destructive interferences to match the observed intensity in reciprocal space. The analogous  
2828 measure is the shrink-wrap algorithm [95] used in alternating projections algorithms. For  
2829 this reason a penalty  $P$  on the size of the support can be added to the loss function with the  
2830 formula:

$$P = \beta \sum_{i,j,k} \mathcal{S} \quad (7.5)$$

2831 where  $\beta$  is a hyperparameter that similarly to  $\alpha$  is calculated from the parameter  $\beta_0$  chosen  
2832 by the user with respect to the data fidelity loss. Typical values of  $\beta_0$  range between (0.01 - 0.5).  
2833 Both the  $\alpha_0, \beta_0$  hyperparameters can be tuned manually during the optimization, to adjust in  
2834 case of need, the strength of the regularization terms. The final formula for the loss function  
2835 can be ultimately expressed as:

$$L = \frac{1}{N_{\text{obs}}} \sum_{i=1}^{N_{\text{obs}}} |I_{\text{calc},i} - I_{\text{obs},i}| + \alpha TV(\varphi) + \beta P(\mathcal{S}) \quad (7.6)$$

2836 For the optimization, a tolerance on the MAE value or a fixed number of steps can be set  
2837 to stop the algorithm. Empirical observations have shown that a MAE value around 0.2 is  
2838 sufficiently low for the result to be considered good. However, an additional refinement with a  
2839 few iterations ( $\sim 300$ ) is recommended for cross-validation.  
2840

2841 Before concluding the section, it is worth highlighting that this AD implementation offers  
2842 the possibility to simultaneously run multiple reconstructions in parallel, efficiently on the GPU.  
2843 One can create a 4D tensor by stacking several copies of the 3D intensity data, creating therefore  
2844 a batch. For each element in the batch a different initial support and phase configuration can  
2845 be chosen, hence increasing the likelihood to converge to the solution.

### 2846 7.3 Results: simulated data

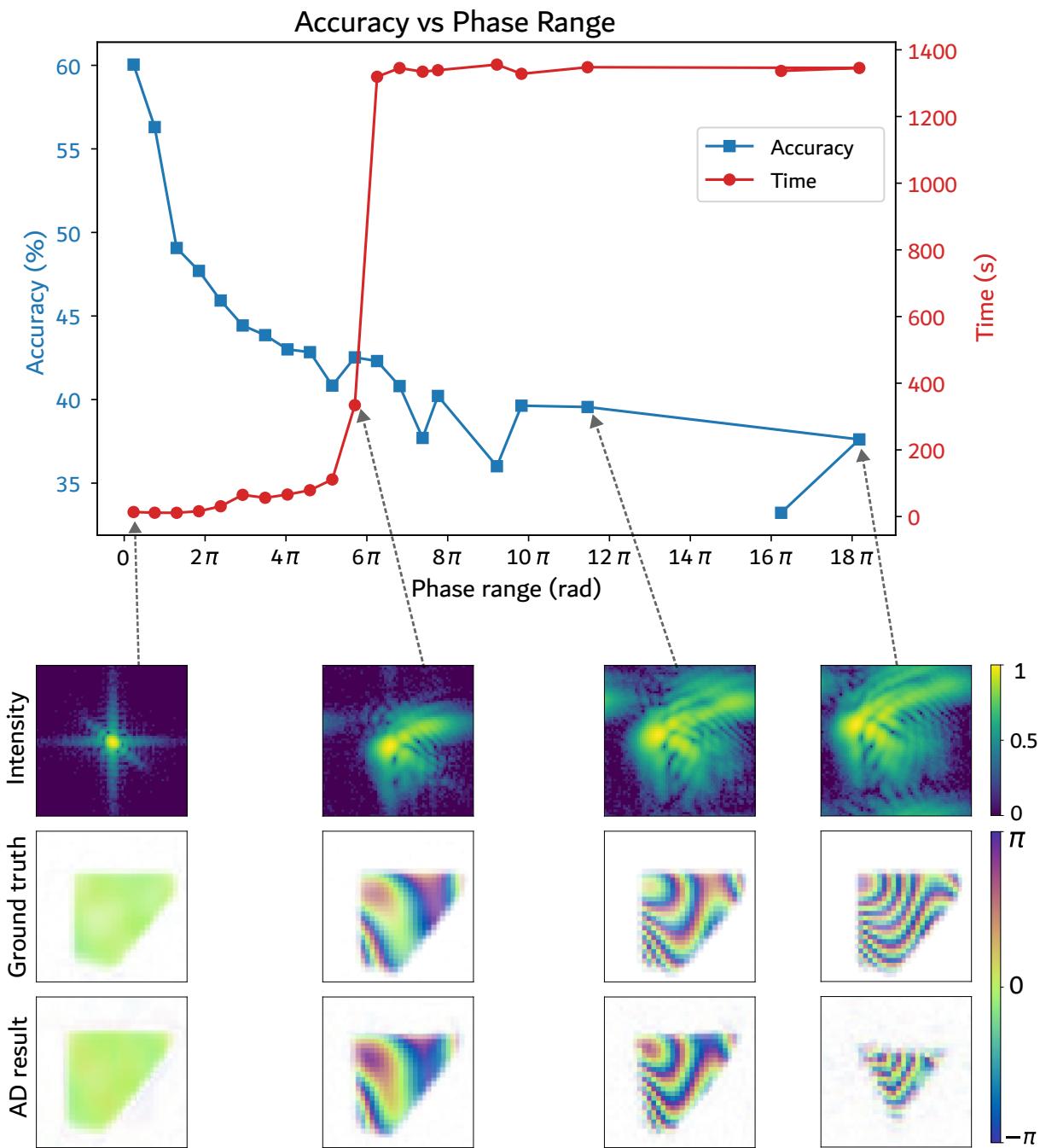
2847 Some tests have been conducted on simulated data. Similar to that described in Section 6.10,  
2848 a 3D randomly shaped faceted particle has been created with an applied Gaussian correlated  
2849 random phase. Subsequently, the diffraction pattern has been simulated on a  $64 \times 64 \times 64$  grid.  
2850 The amplitude of the phase field has been gradually increased from 0 to  $19\pi$  inside the particle  
2851 and for each of the 20 steps the corresponding diffraction pattern was simulated with DFT,  
2852 keeping a constant oversampling ratio and noise level.

2853      The PR has been conducted with the following parameters:

Parameter	Value
batch_size	20
nb_half_spaces	128
sigmoid_eps	0.6
phase_type	'tucker'
ranks	(8, 8, 8)
alpha_TV	0.0
beta_small	0.01
initial_lr	0.05
tol	0.09
max_iter	10000

**Table 7.1:** 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  $\epsilon$  parameter controlling the "spatial resolution" of the support, (iv) the object phase is parametrized with the Tucker decomposition, and (v) is the size of the 3D core tensor (vi) the coefficient multiplying the TV loss on the phase (vii) the coefficient multiplying the penalty on the support size, (viii) the initial learning rate for the ADAM optimizer and (ix) tolerance on the MAE error and (x) the number of maximum iterations.

2854      The results are shown in Fig.7.2. Objects with small phases are reconstructed within a few  
 2855      seconds while for larger phases the computational time rapidly increases to the maximum fixed  
 2856      by the limit of iterations.



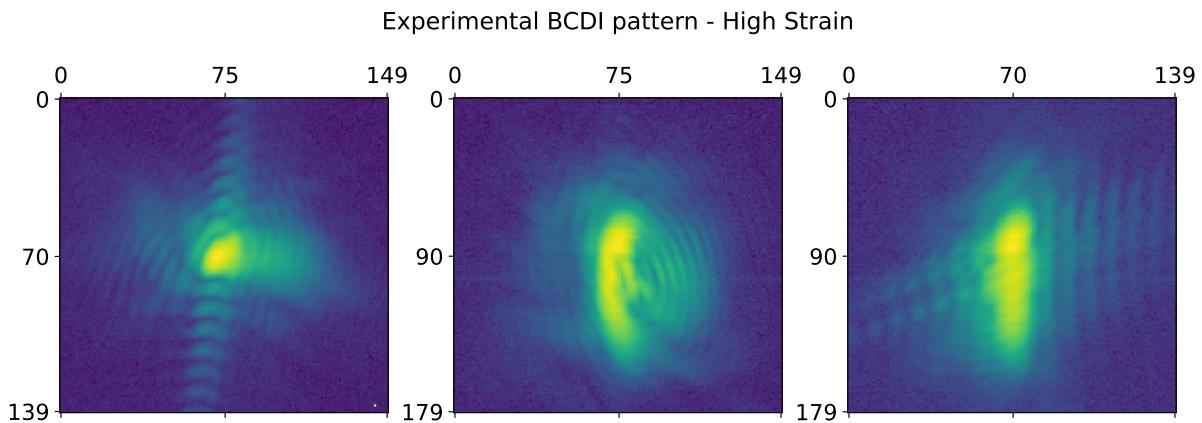
**Figure 7.2:** Results of the PR on simulated data for increasing applied phase magnitude. The accuracy has been calculated with the formula in Eq.6.5 and the time is measured from the start of each run until it ends, either when the tolerance is surpassed or when the maximum number of iterations is reached. For the smallest phase range (0.72 radians) the PR is completed within 11 seconds. Starting from phase ranges exceeding  $6\pi$  the model runs over all the allowed iterations. The PR convergence is however dependent on the initialization of the support (randomly oriented half-spaces) and the phase (random uniform between  $-\pi$  and  $\pi$ ), thus the oscillations of accuracy scores. Notice that the heavily distorted diffraction patterns here depicted show a significant amount of aliasing. Although never occurring in experimental data, this is not a problem to the scope of this analysis.

2858 faceted crystals.

## 2859 7.4 Results: experimental data

2860 In this section two relevant results will be presented. The first example is a highly strained  
 2861 palladium particle on sapphire substrate measured at the ID01 beamline of the ESRF [209]. The  
 2862 large strain inside the particle distorts the BCDI pattern and makes the reconstruction with  
 2863 conventional iterative algorithms overly challenging.

### 2864 7.4.1 Hardly-invertible BCDI patterns with high strain



**Figure 7.3:** Projections along the three axes of a BCDI pattern of the highly strained Pd particle.

2865 Here a comparison between different reconstruction methods will follow. Namely, (i) the results  
 2866 obtained from 60 independent runs of standard PR using PyNX software. Each run consisted of  
 2867 400 HIO iterations followed by 1000 RAAR and 300 ER ones. The parameters are displayed in  
 2868 Table 6.2 already mentioned in the previous chapter. At the end of the process, the two best  
 2869 reconstructions among the 60 runs according to the “mean to max” metric were combined  
 2870 using the mode decomposition technique proposed in [195]. This method is referred in the text  
 2871 to as “PyNX”.

2872 The second method makes use of the DL model presented in the previous chapter. The  
 2873 large original data is firstly binned to a (196, 140, 140) shape and then cropped in a (80,90,110)  
 2874 shaped ROI and resized to a 64 pixel-sided cube for the DL model. The DL predicted object is  
 2875 then interpolated back to the original size and refined with PyNX using a single run of 300 ER,  
 2876 with the parameters listed in Table 6.3. This combined method is referred in the text to as “DL  
 2877 + PyNX”.

2878 The third method employs the AD model presented above with the parameters listed in  
 2879 Table 7.2. Additionally, as last step, the final object is obtained computing the IFFT of the  
 2880 complex diffracted amplitude built using the experimental diffracted measurement as modulus  
 2881 and the RSP extracted from the FFT of the object itself. This last step that can also be seen as a  
 2882 “halfER step”. The method described here is referred in the text to as “AD”.

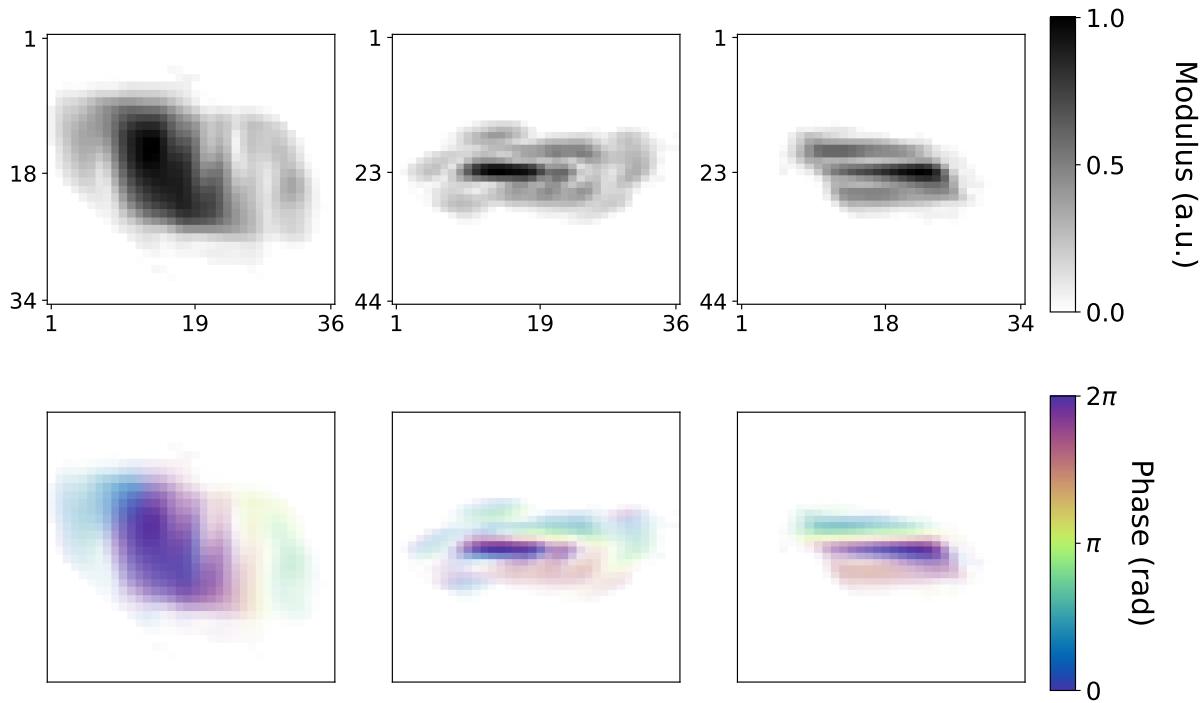
2883

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 6.3 using as initial guess the object found with the AD, is in the text referred to as “AD + PyNX”.

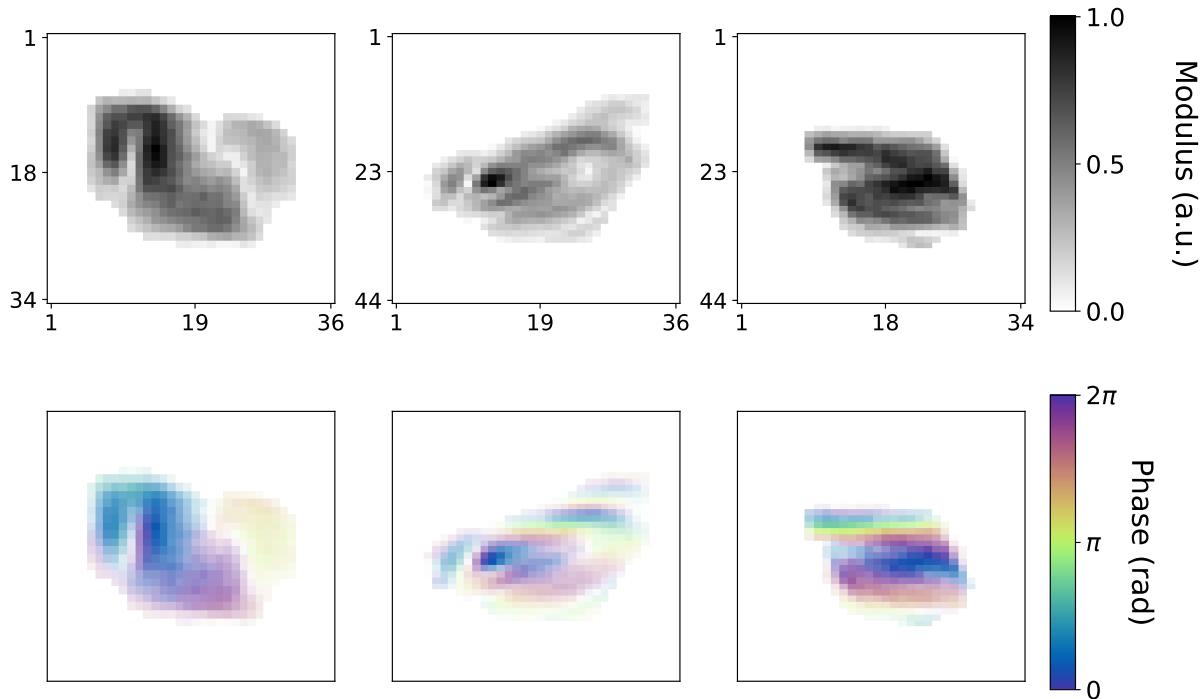
Parameter	Value
batch_size	5
nb_half_spaces	128
sigmoid_eps	0.9
phase_type	‘tucker’
ranks	(16, 16, 16)
alpha_TV	0.0
beta_small	0.01
initial_lr	0.05
tol	0.2

**Table 7.2:** Parameters initialization for the AD model for the phasing of experimental data. Given the larger size of the data as compared to the simulated ones, some parameters have been adjusted with respect to Table 7.1. Namely, the number of copies has been reduced for memory issues, the core tensor for the Tucker decomposition has been increased and the tolerance and maximum iterations constraints have been relaxed.

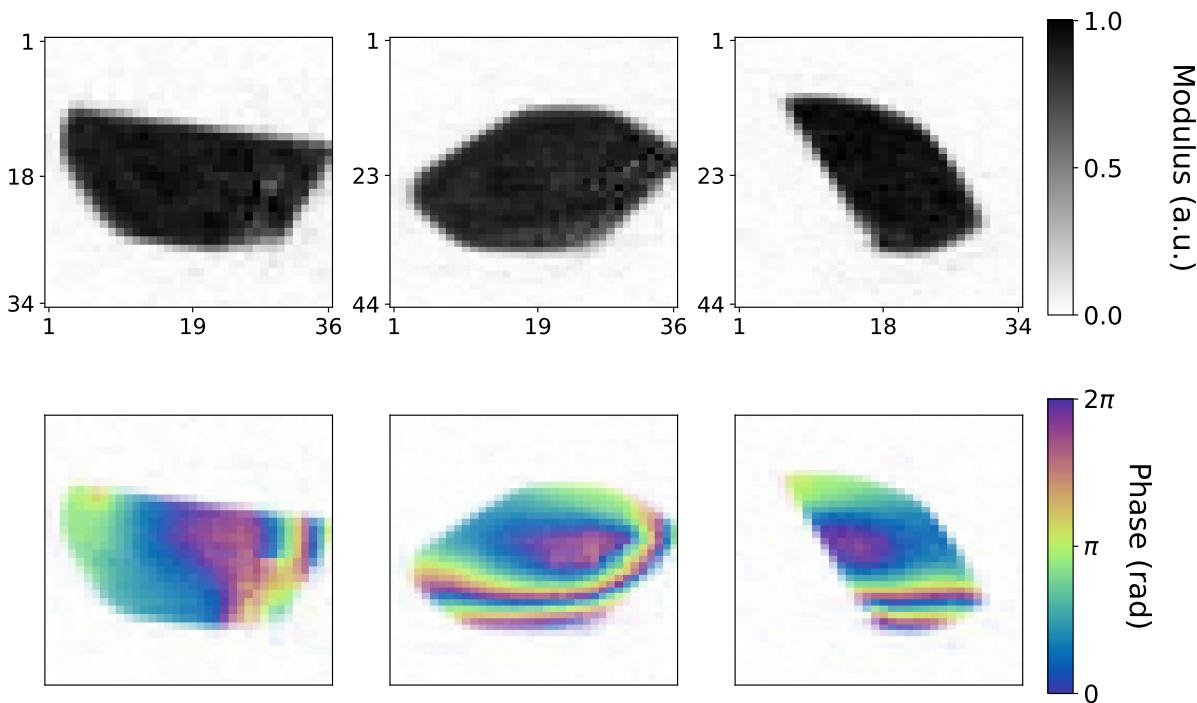
The following Figures 7.4 - 7.5 - 7.6 - 7.7 show the results of the reconstructions obtained with the “PyNX”, “DL + PyNX”, “AD” and “AD + PyNX” methods respectively, while Table 7.3 displays the scores of the Poisson LLk (calculated from Eq.7.4), MAE and MSE metrics evaluated for the  $I_{obs}$  and  $I_{calc}$  in each case.



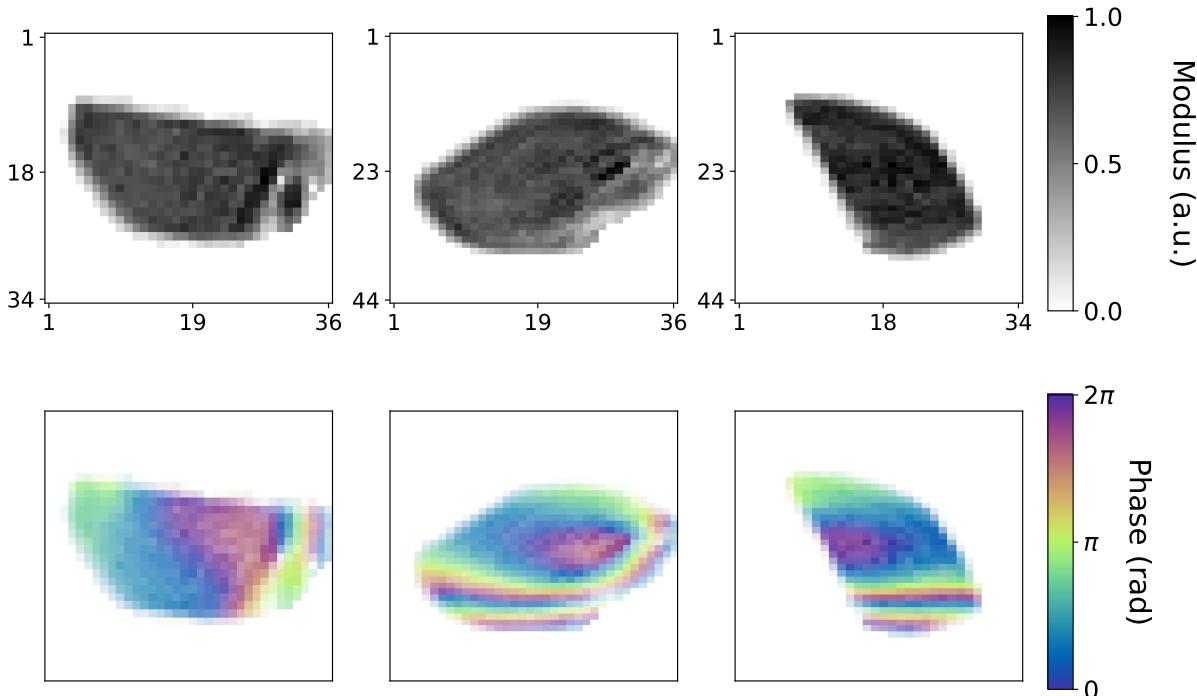
**Figure 7.4:** Central slices for modulus (first row) and phase (second row) of the reconstruction obtained with the PyNX method. The presence of holes and inhomogeneous object's electron density suggests a poor quality reconstruction.



**Figure 7.5:** 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 7.6:** 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. The model overcomes the tolerance level within 1000 steps, for an elapsed time of 325 seconds



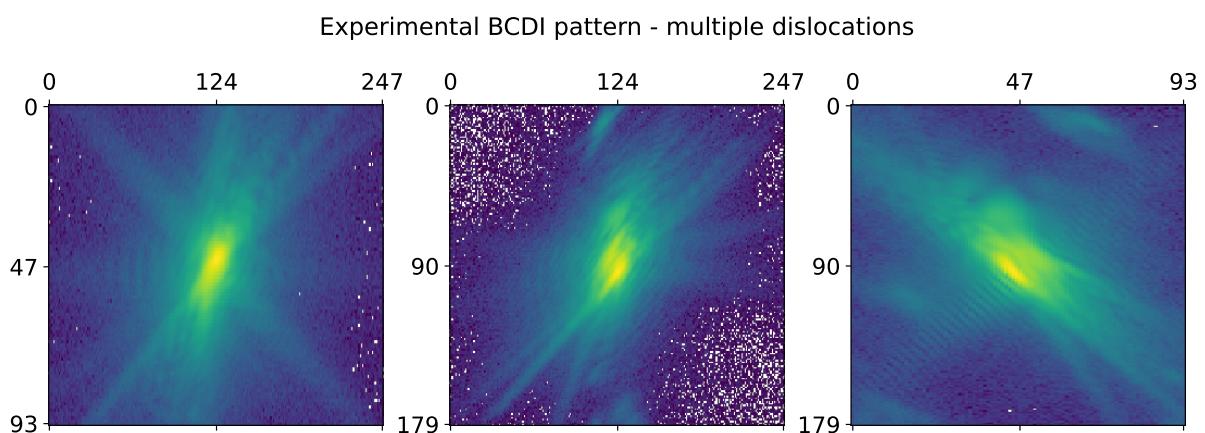
**Figure 7.7:** Central slices for modulus (first row) and phase (second row) of the reconstruction obtained with the AD + PyNX method. The 300 iterations of ER refinement do not alter the shape nor the phase found by the AD model, therefore validating the solution.

	Poisson LLK	MAE	MSE
AD	3.3083e-01	4.1249e-02	7.3738e-03
AD + PyNX	2.9259e-01	1.1893e-01	5.4739e-03
DL + PyNX	3.5195e-01	1.5866e-01	1.5841e-02
PyNX	6.3946e-01	3.1050e-01	3.7996e-02

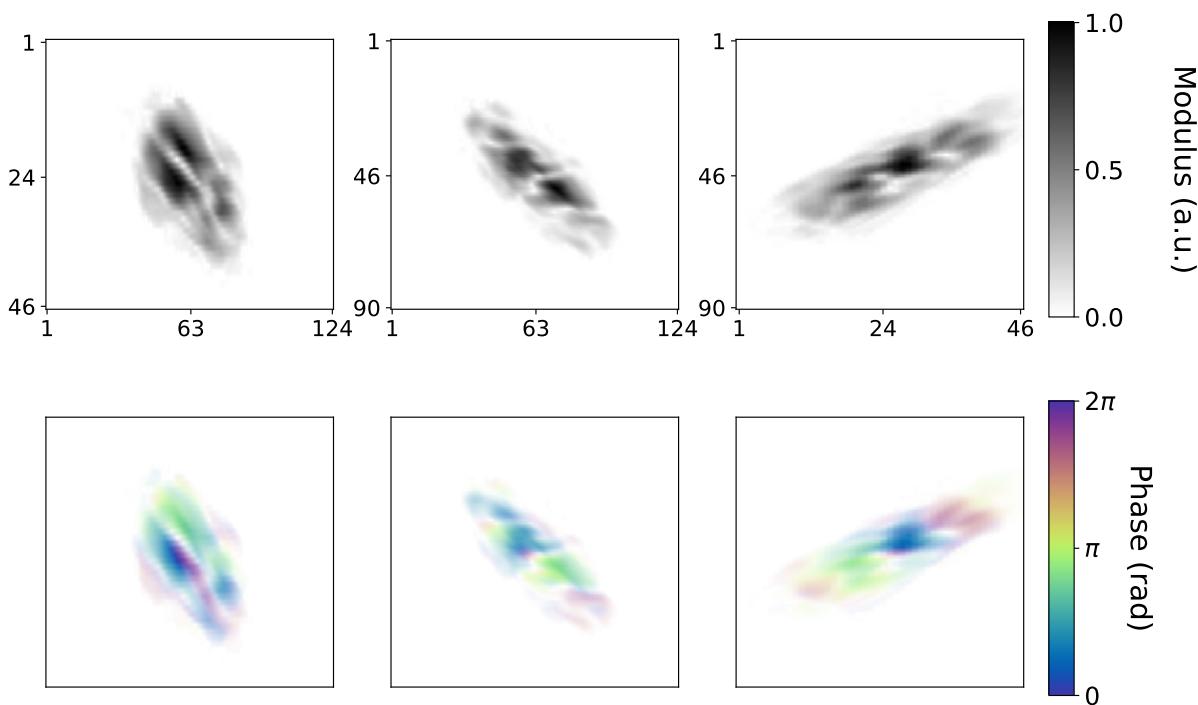
**Table 7.3:** Performance metrics between  $I_{calc}$  and  $I_{obs}$  for the Pd particle calculated for normalized magnitudes. The lowest scores are highlighted

#### 2894 7.4.2 Hardly-invertible BCDI patterns with multiple dislocations

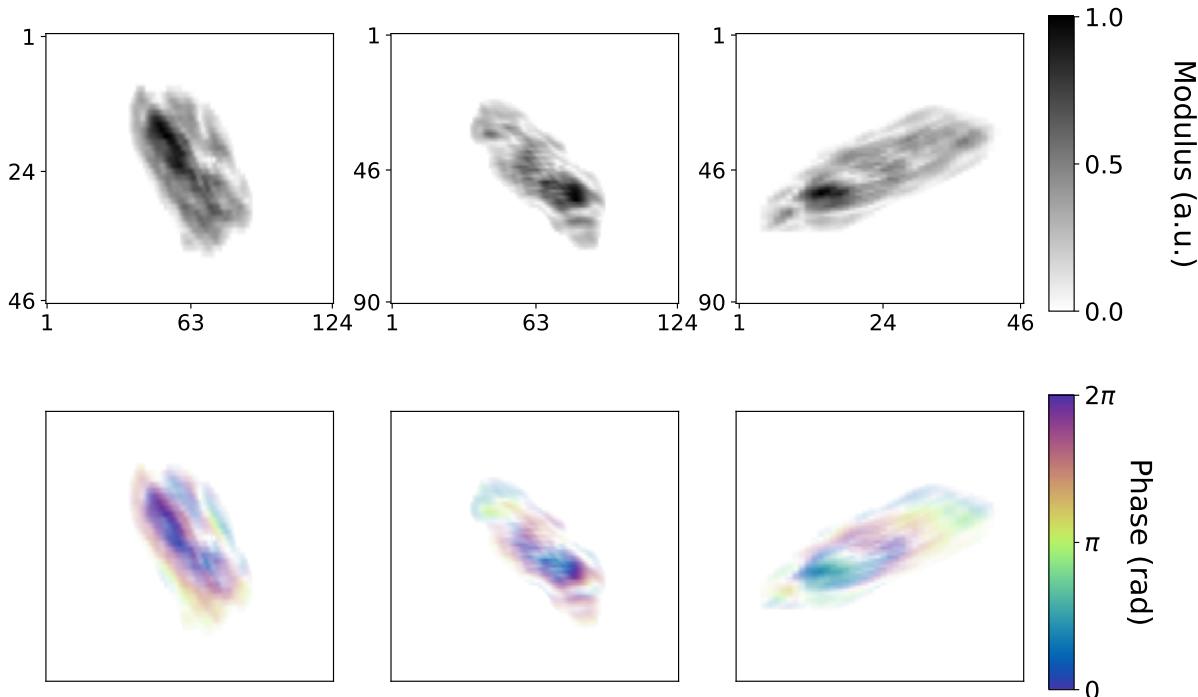
2895 In this paragraph another illustrative example is presented. This time a platinum particle on  
 2896 YSZ substrate. The four different methods presented above are repeated here for this dataset in  
 2897 the same way, except for the AD method, for which the phase has been parametrized with the  
 2898 full grid (phase\_type: grid) and  $\alpha_{TV} = 0.1$  parameter. Although more expensive in  
 2899 terms of memory, this parametrization is more suitable for particles with dislocations, where  
 2900 large phase variations are concentrated in a few voxels, and a built-in sparse formulation can  
 2901 struggle with these high-frequency features. The results are shown in the following Figures 7.9  
 2902 - 7.10 - 7.11 - 7.12 and Table 7.4



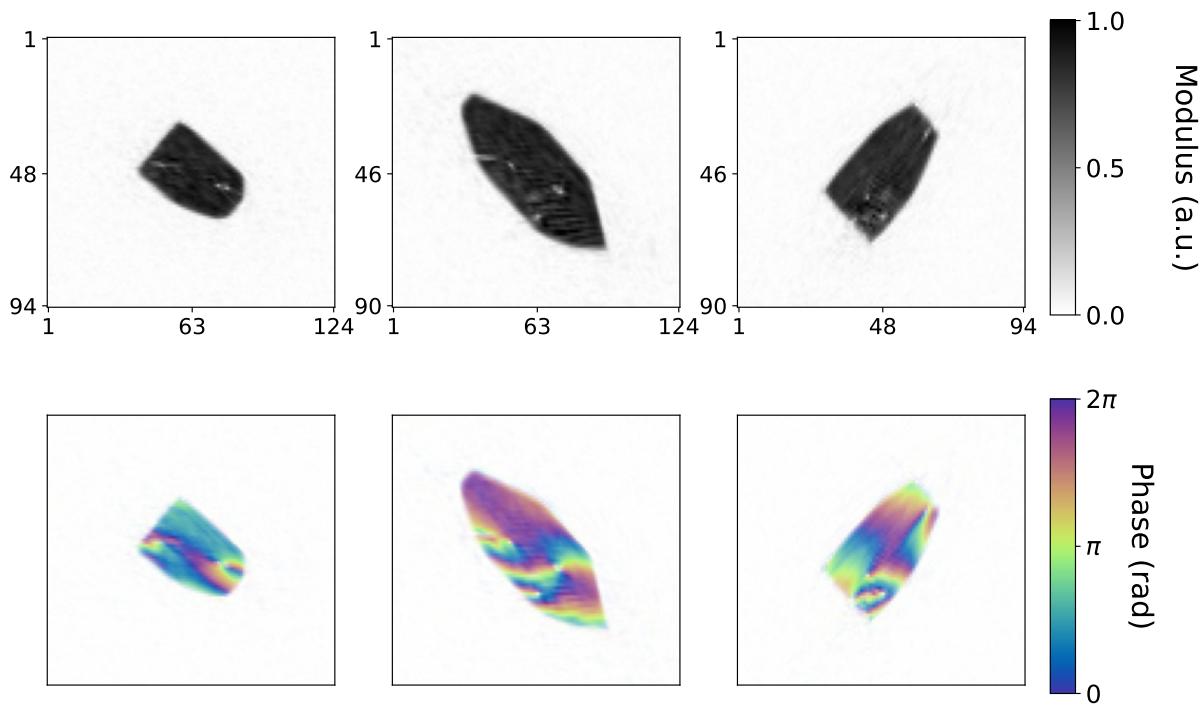
**Figure 7.8:** Projections along the three axes of a diffraction pattern from a Pt particle with multiple dislocations.



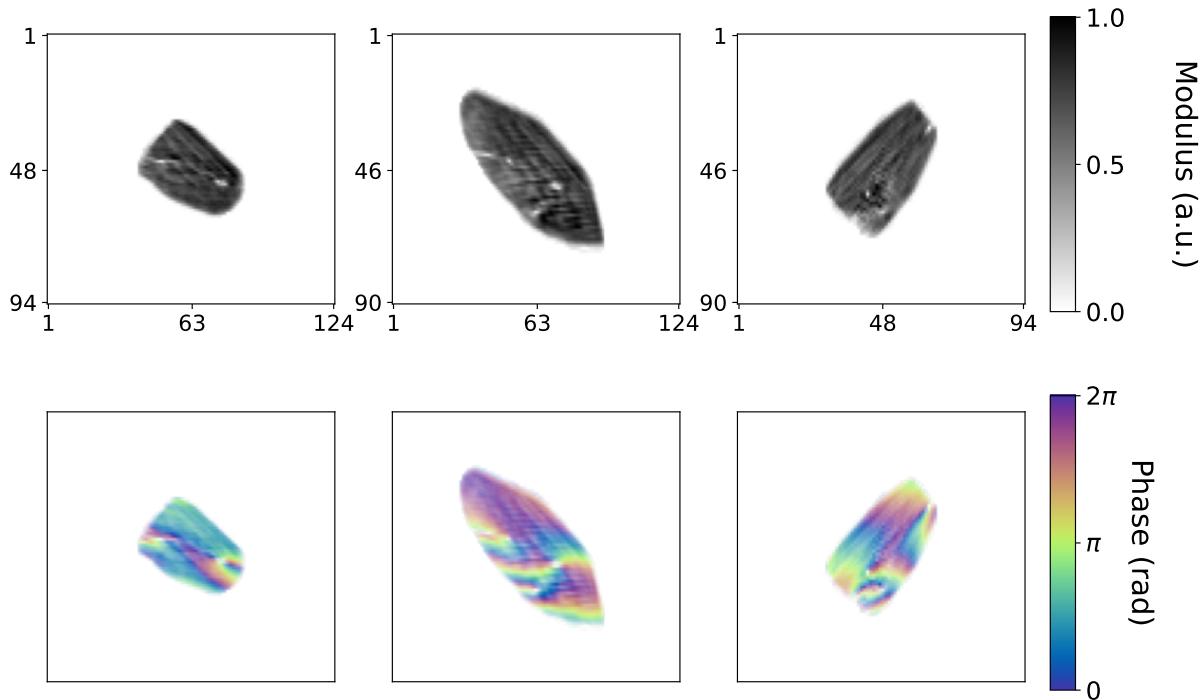
**Figure 7.9:** Central slices for modulus (first row) and phase (second row) of the reconstruction obtained with the PyNX method. The presence of holes and inhomogeneous object's electron density suggests a poor quality reconstruction.



**Figure 7.10:** 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.

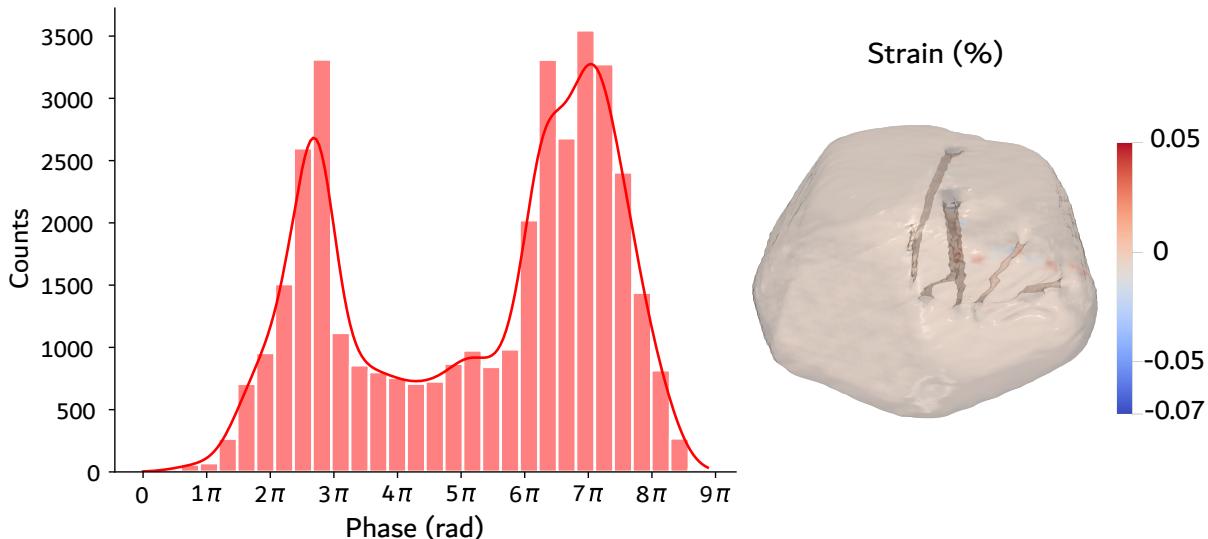


**Figure 7.11:** Central slices for modulus (first row) and phase (second row) of the reconstruction obtained with the AD method. The AD model converges to a reasonably faceted crystal with several dislocations. The model overcomes the tolerance level within 1500 steps, for an elapsed time of 930 seconds



**Figure 7.12:** 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*).

	Poisson LLK	MAE	MSE
AD	2.5650e-01	1.3828e-02	1.0402e-02
AD + PyNX	1.5138e-01	8.5945e-02	3.0269e-03
DL + PyNX	2.9194e-01	1.6544e-01	1.4580e-02
PyNX	3.8114e-01	2.4265e-01	1.7078e-02

**Table 7.4:** Performance metrics between  $I_{calc}$  and  $I_{obs}$  for the Pt particle.**Figure 7.13:** Histogram of the phase range and 3D strain visualization for the Pt particle in Fig. 7.12. The large phase range extending over  $8\pi$  confirms the challenging PR. The 3D rendering with reduced opacity highlights the multiple dislocation cores traversing the particle (orthogonalized).

## 2903 7.5 Conclusions

2904 In this chapter a novel PR method for BCDI based on a physics-informed AD model has been  
 2905 presented. The goal of this section was to explore GD-based methods for PR, aiming at resolving  
 2906 those difficult cases for which both conventional iterative algorithms and DL assisted methods  
 2907 struggle. To sum up, the major factors that made the model successful are here listed briefly:

- 2908 • The efficient exact gradient calculation offered by the automatic differentiation, already  
 2909 identified by Jurling and Fienup as potential alternative to alternating projections for PR, is  
 2910 today easily accessible and GPU accelerated by common machine learning programming  
 2911 libraries like Tensorflow and PyTorch. This ingredient is fundamental for PR of 3D  
 2912 datasets in competitive computational times, comparable to standard PR algorithms  
 2913 optimized for GPUs.
- 2914 • The ADAM optimizer offers adaptive step-size tuning (eliminating the need to set a single  
 2915 learning rate for all parameters) and first and second order moment estimation (thus  
 2916 providing both direction smoothing and per-parameter scaling), leading to more stable

2917 and faster convergence. Additionally, it allows computing gradients on sub-volumes or  
 2918 slices of the diffraction pattern at each step, hence introducing stochasticity that could be  
 2919 beneficial for escaping local minima. This feature, could be developed in future versions  
 2920 of the model, and strategic batches organization (i.e. concentric sub-volumes from the  
 2921 center of the Bragg peak) could be explored as well.

- 2922 • The possibility to easily embed physical constraints in the forward model. In this case  
 2923 the prior knowledge on the homogeneous and compactly supported nature of the crystal  
 2924 electronic density can be easily implemented with the half-spaces method, thus restricting  
 2925 the solution space without need for additional regularization. In the same way, the Tucker  
 2926 decomposition of the object's phase tensor facilitates the finding of physical solutions.
- 2927 • The multidimensional tensor-based computations typical of modern machine learning  
 2928 libraries make it straightforward to extend the optimization to multiple parallel instances  
 2929 of the same phase retrieval problem, with only a modest increase in computational time.  
 2930 The main limitation, however, lies in the substantial GPU memory requirements, as  
 2931 this approach can be highly memory-intensive. To give some useful numbers, with the  
 2932 parameters shown in Table 7.2 a GPU with 32GB of RAM can sustain the optimization of  
 2933 maximum 30 copies of a  $64 \times 64 \times 64$  diffraction pattern.
- 2934 • The loss function definition gives the user high flexibility of implementing the most  
 2935 suitable metric for the specific problem, allowing for parameter tuning during the optimi-  
 2936 zation as well. In this case the MAE metric was found to be the best one for the BCDI  
 2937 problem.

2938 This study has shown that AD-based PR for BCDI is a valuable alternative to conventional or  
 2939 data-driven PR algorithms. It offers an additional tool that can extend the range of applicability  
 2940 of the BCDI technique to highly defective or strained crystals, the reconstruction of which  
 2941 is problematic. The flexibility of the programming enables the implementation of targeted  
 2942 models, and it is suitable for tailored constraints, important for PR of challenging datasets.

2943 The current implementation however still requires significant amounts of time for partic-  
 2944 ularly ill-posed PR, yet it achieved promising results on experimental datasets whose phase  
 2945 retrieval had previously been discarded due to the inability of conventional algorithms to yield  
 2946 a reconstruction. Lastly, for future developments, the model can be extended to non-convex  
 2947 objects or separated ones (e.g. in case of twin boundaries) as well as different forward models  
 2948 beyond the kinematic approximation or further embedding of prior knowledge.



---

## 2950 CONCLUSIONS AND PERSPECTIVES

2951 In this PhD dissertation the use of deep convolutional neural networks and algorithmic dif-  
2952 ferentiation has been explored for the processing of BCDI data. Chapter 2 has presented the  
2953 physics of coherent x-ray diffraction on single crystal, highlighting the effect of internal lattice  
2954 displacement on the data. Moreover, a short practical overview of the typical BCDI experiments  
2955 was given as well. Chapter 3 was dedicated to the fascinating Fourier phase problem. The  
2956 uniqueness conditions, the PR iterative algorithms and some insights on the high-strain case  
2957 were discussed. In particular, the link between the effects of the high-strain on the diffraction  
2958 pattern, illustrated in Chapter 2 and the relative increased difficulty of the PR shown in Chapter  
2959 3 has been emphasized. What emerged in the discussion is that improved performance of the  
2960 PR is obtained when the problem is *regularized* with some prior knowledge that constrains  
2961 or guides the search of the solution. Here, the connection to neural networks introduced in  
2962 Chapter 4 becomes apparent. The strong inductive prior of convolutional neural networks for  
2963 structured images, combined with a targeted data-driven strategy, was explored in Chapters 5  
2964 -6, yielding satisfactory results on two different kinds of inverse problems.

2965 Specifically, in Chapter 5 the preliminary investigations and the development of a novel  
2966 patching-based model for addressing detector gaps in BCDI data were presented. The results  
2967 obtained on new simulated and experimental datasets confirmed the capability of convolutional  
2968 neural networks to extrapolate information from structured images and to accurately predict  
2969 the continuation of patterns within missing data regions. In Chapter 6 the goal of assisting  
2970 standard iterative algorithms during the PR of highly-strained BCDI patterns has been achieved  
2971 with the use of a convolutional neural network trained with the novel WCA loss function  
2972 for the prediction of the *reciprocal space phase*, unlike what is present in the literature. The  
2973 successful results attained by the DL + PyNX refinement method can significantly improve the  
2974 BCDI technique by drastically reducing the computational time needed for the reconstructions  
2975 of experimental data. Over the long term, the computational resources required to train the DL  
2976 model are expected to be minimal relative to the substantial time and energy savings achieved  
2977 through DL-based initialization of the PR.

2978 Chapter 7 instead moves away from the data-driven approach. Here, the computational  
2979 framework for automatic differentiation is leveraged for a physics constrained PR solved with  
2980 gradient descent. The prior knowledge of uniform electron density inside the object support  
2981 and the faceted nature of crystals' surfaces does not come from the data, nor from penalty  
2982 terms but from construction constraints.

Integrating the presented algorithms into the standard BCDI data analysis pipeline should be foreseen to enable more systematic usage. So far, all three main models have been employed by ID01 users for inpainting and phase retrieval tasks, with experimental results soon to be published. However, before this integration some further developments could be considered. I will provide an outlook on potential directions with the following.

• **DL-based Gap Inpainting.** The patching approach has proven to have many benefits, including faster training, larger training datasets and possibility of targeted fine-tuning. The bottleneck of this method is however given by the size of the gaps. Detectors like EIGER, have large gaps (12 - 38 pixels) for which the patching approach is not suitable. One could get around this limitation increasing the patch size, however, in those cases other methods leveraging some type of regularization during the reconstruction [147] can offer a better alternative to a DL approach. Another interesting development could aim to reduce the number of repeated applications of the DL model along the gap. In fact, given the typical structure of BCDI patterns and experimental conditions, the gap only affects a small region of signal, often on long truncation rods streaks. An adaptive algorithm for the inpainting of those regions only, could save time and computations discarding the inpainting of dark regions. More in general, it would be interesting to test the patching-based inpainting on data from other imaging techniques.

• **DL-based Phase Retrieval.** The RSP prediction on patches is at the same time appealing and challenging. The results obtained on independent patches are promising, but the stitching of the patches still represents a challenge. As anticipated in the conclusion Chapter 6, the design of a Recurrent convolutional neural network could better address the problem. In particular, I would suggest an approach similar to what proposed by Pinheiro and Collobert in [210] could be considered. There, instead of stacking many different layers to increase receptive fields, they used *weight-sharing* recurrence, i.e. the same convolutional layer is applied multiple times to its own output. This allows the network to iteratively refine predictions and integrate increasingly larger context without adding new parameters.

This approach could also break the symmetry problem that was evidenced in the end of Chapter 6 (see Fig. 6.58) for which the predicted RSP always tend to show a radial symmetry like the average, over the whole training dataset, of the diffraction intensity that the model receives as inputs.

• **AD-based Phase Retrieval.** Concerning this project, in my opinion the potential is high and several developments, additions, integrations can be foreseen. The high flexibility and efficiency of gradient-based optimizations provided by modern machine learning libraries allows for relatively easy implementation of tailored models. The first important extension to the current formulation should include the modeling of non-convex surfaces as well. An idea that would maintain the half-spaces approach would be to define two or more convex volumes built with the half-spaces method and find the final support with union, subtraction and intersection operations. Additionally, other upgrades to the current model could make use of the observed intensity projection (the projection on the modulus constraint set presented in Chapter 3), combined with the gradient descent, as well as the use of mini-batches to enable stochastic gradient descent, for faster and more robust convergence.

3027 More in general, it is clear that the utilization of GPU-accelerated AD will become  
3028 a pivotal tool for efficient gradient-based optimization in various scientific disciplines  
3029 characterized by intensive computational demands. This approach is anticipated to gain  
3030 prominence in the coming years, potentially surpassing data-driven methodologies in  
3031 certain applications [128, 211, 212].



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