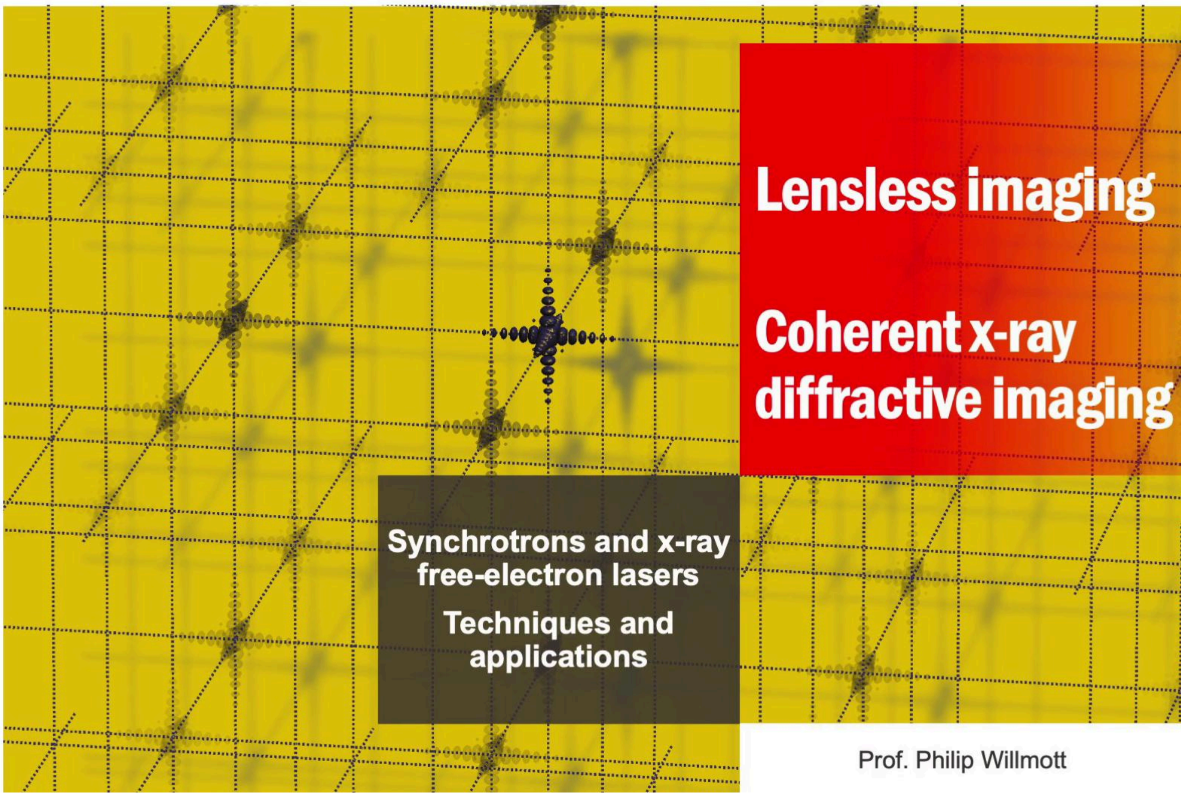
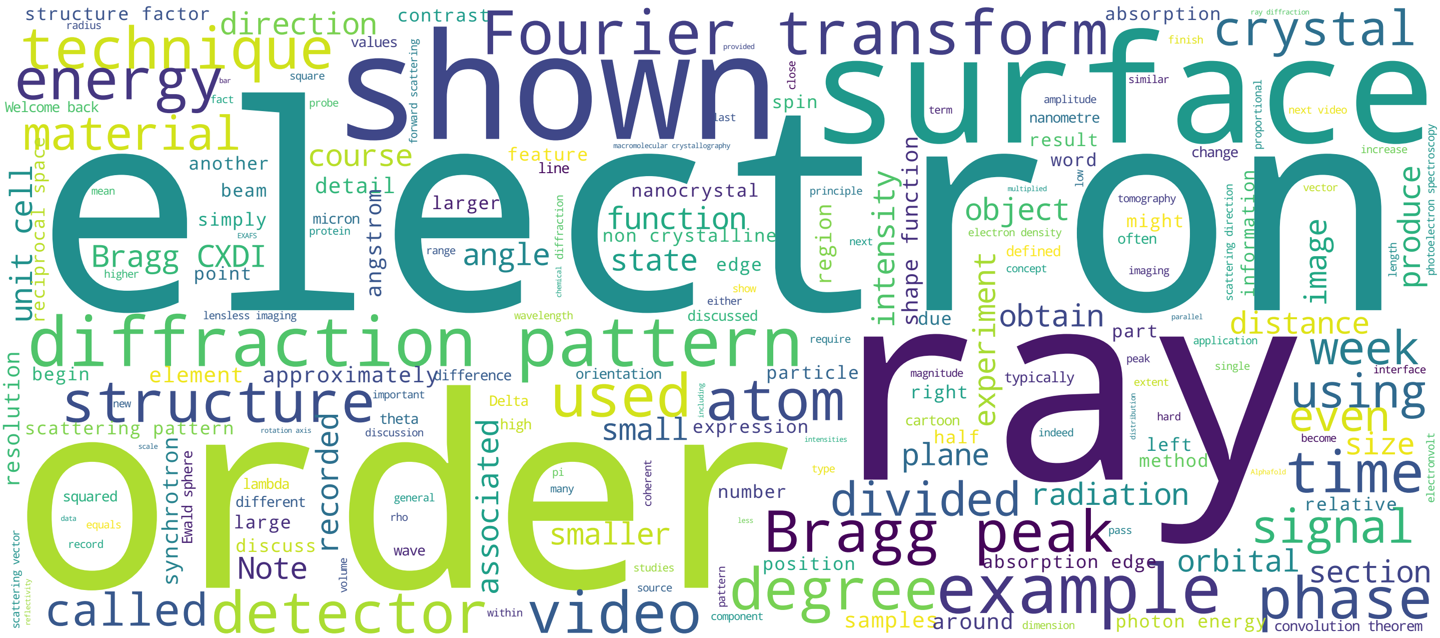
École polytechnique fédérale de Lausanne



Prof. Philip Willmott



Search MOOC

Video

EPFL

Contents and objectives of this video

- CXDI = lensless imaging
  - Setup and procedure
- Bragg CXDI
  - Convolution explanation
  - Setup and procedure
  - Applications

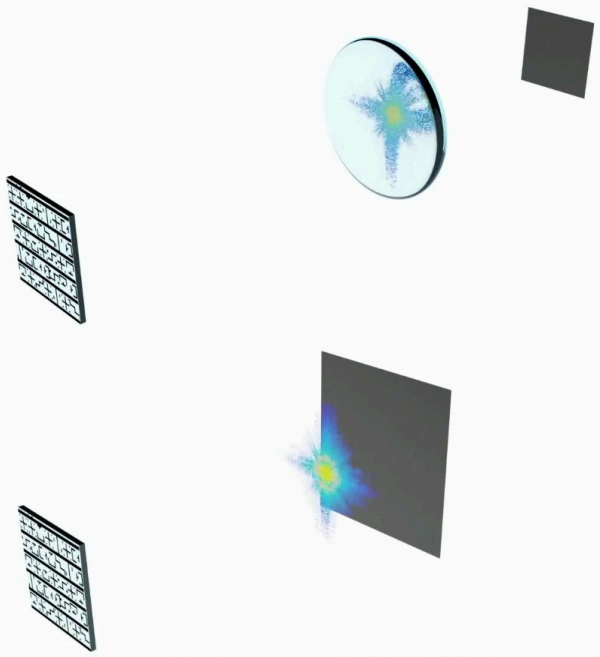
Welcome back to the last of the three videos of this section, in which we cover lensless imaging in more detail, both for non-crystalline samples, but also for nanocrystalline samples in the variant called Bragg coherent X-ray diffractive imaging. This is explained via the convolution theorem.

Notes

Summary

0m 05s

Coherent x-ray diffractive imaging



- CXDI (or CDI)
- Also ‘lensless imaging’
- Diffraction pattern
  - Noncrystalline sample
    - In forward-scattering direction only
  - Crystalline objects
    - Regular array of replicas of same pattern
    - Bragg-CXDI
  - Oversampling determined by size of coherently illuminated sample
    - Smaller samples ⇒ larger features
- Phase problem resolved typically via phase-retrieval algorithms
  - Gerchberg-Saxton (error reduction)
  - Hybrid input-output (Fienup)
  - Difference-map
  - …

Animations courtesy I. Mochi, Swiss Light Source

See V. Elser https://opg.optica.org/josaa/abstract.cfm?URI=josaa-20-1-40

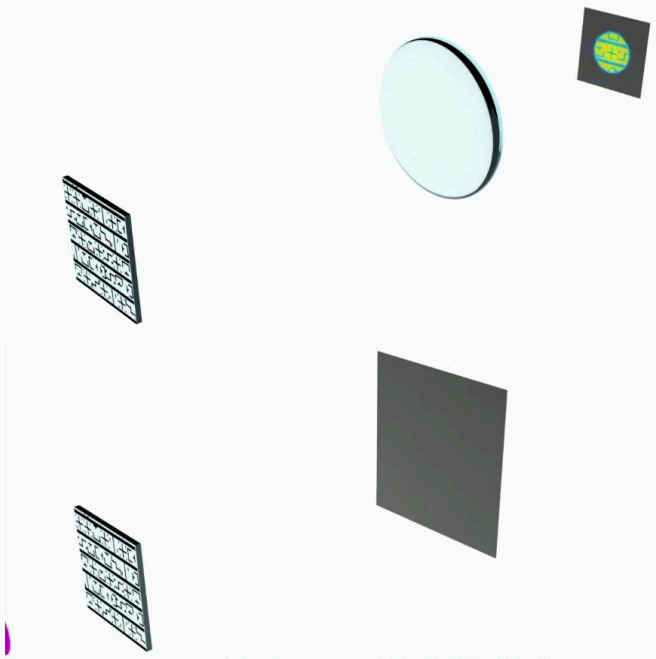
In coherent X-ray diffractive imaging or CXDI or simply just CDI, instead of collecting the radiation or scattered radiation with a lens and forming an image of the scattered object or scattering object, one simply records the far field scattering pattern, which hopefully by now we recognise as being the absolute square of the Fourier transform of the coherently illuminated object. In the case of CXDI of non-crystalline or polycrystalline samples, the scattering pattern is limited to the forward-scattering direction, covering angles of the order of lambda divided by d, where lambda is the X-ray wavelength and d is the lateral extent of the scattering centres within the sample. As these are rarely much smaller than 10 nanometres, the maximum scattering angles involved of the order of about a hundredth of a radian or about a degree for 1 angstrom radiation. For studies of crystalline objects, the scattering pattern is repeated in a regular array in reciprocal space. This added feature is exploited in Bragg CXDI, as we shall shortly see. The degree of oversampling for a given sample detector distance and pixel size is given either by the sample extent or by the lateral coherence length, if this is smaller than the sample size.

Notes

Summary

0m 28s

Coherent x-ray diffractive imaging



Animations courtesy I. Mochi, Swiss Light Source

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- Also ‘lensless imaging’
- Diffraction pattern
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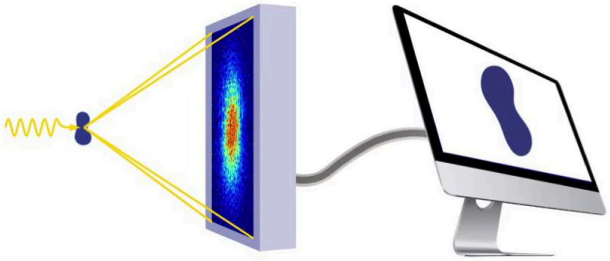
The approach to retrieving the phases from the scattering patterns is typically achieved via iterative algorithms of differing complexity and sophistication. The evolution of these techniques has been nicely summarised in Veit Elser's 2003 review, the link to which is provided here.

Notes

Summary

2m 05s

Coherent x-ray diffractive imaging



- XRD
  - Samples have translational symmetry
  - Record far-field scattering (diffraction) pattern
  - Regain real-space structure through IFT
    - Phase problem
  - Unit cells $\lesssim$ 200 Å
  - Resolution $\lesssim$ Å
- CXDI
  - Same principle as XRD
  - Samples can be crystalline or noncrystalline
    - Scattering pattern: “speckle”
    - Sizes up to ~ μm
    - Requires sample < coherence volume of SR
    - Big improvements with DLSRs!!
    - Resolution down to ~ 10 nm

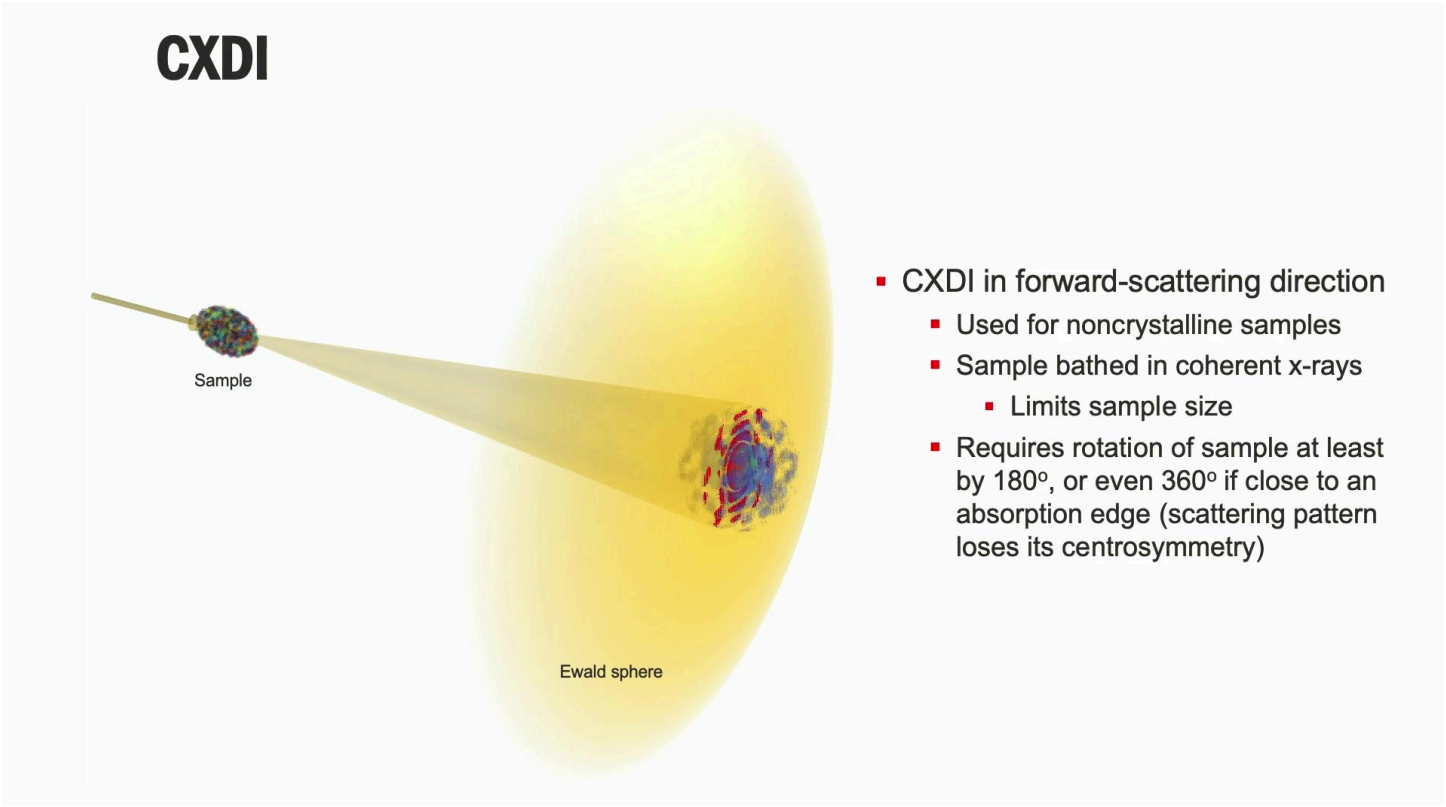
See also H.N. Chapman and K.A. Nugent
https://www.nature.com/articles/nphoton.2010.240

How does CXDI differ from conventional X-ray diffraction? The latter is used for samples with translational symmetry. The phase problem is resolved using many different approaches, depending on the unit cell size, which only rarely exceeds 200 angstroms, and then only in macromolecular crystallography, which is anyway being superseded by cryogenic electron microscopy for such large systems, and the internal complexity of the atomic basis. The real space resolution is of the order of an angstrom or even a small fraction of an angstrom. Now, in CXDI, the principle is much the same. Those samples can be, as we've already said, both crystalline and non-crystalline. CXDI requires that the sample can be contained within the coherence volume of the radiation. Hence, big improvements in both speed and resolution can be expected with the advent of diffraction-limited storage rings. Resolutions down to around 10 nanometres or even smaller is nowadays possible.

Notes

Summary

2m 30s



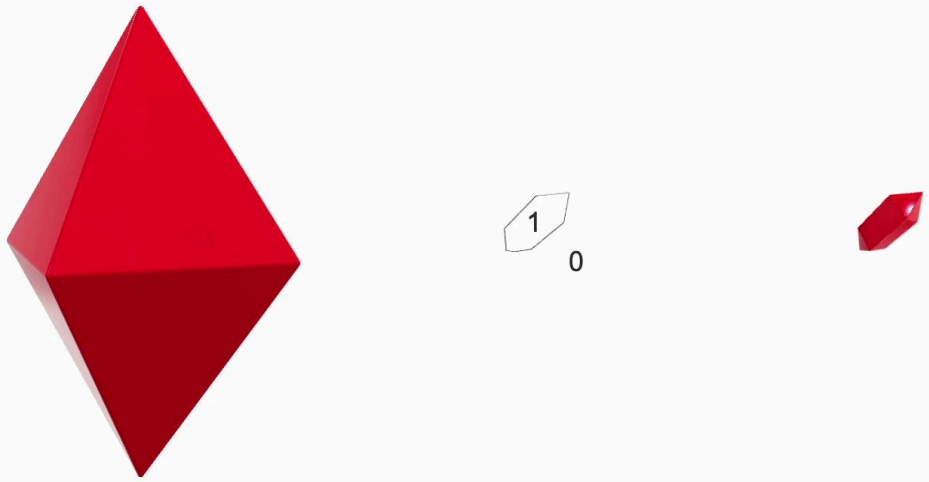
Shown here is a cartoon of a CXDI experiment of a non-crystalline sample. The cartoon isn't to scale, and in fact, the coherent part of the beam incident on the sample must flood it entirely in order to obtain its entire structure. The 3D sample produces a 3D Fourier transform, and hence it must be rotated by at least 180 degrees in order for all the structure factors to pass through the Ewald sphere. If the experiment is performed close to an absorption edge of one of the elements making up the sample, the diffraction pattern will lose its centrosymmetry, as discussed in the first week of this course, and one must therefore record the entire diffraction pattern by rotating the sample through a complete circle of 360 degrees. This set stringent specifications on the performance of the sample manipulator, which shouldn't exhibit a rotation axis drift or wobble that lead to uncertainties in the position of any elements of the sample greater than the desired resolution.

Notes

Summary

3m 46s

Bragg CXDI – a perfectly regular starry firmament



Perfect large crystal x Nanosized volume = Nanocrystal

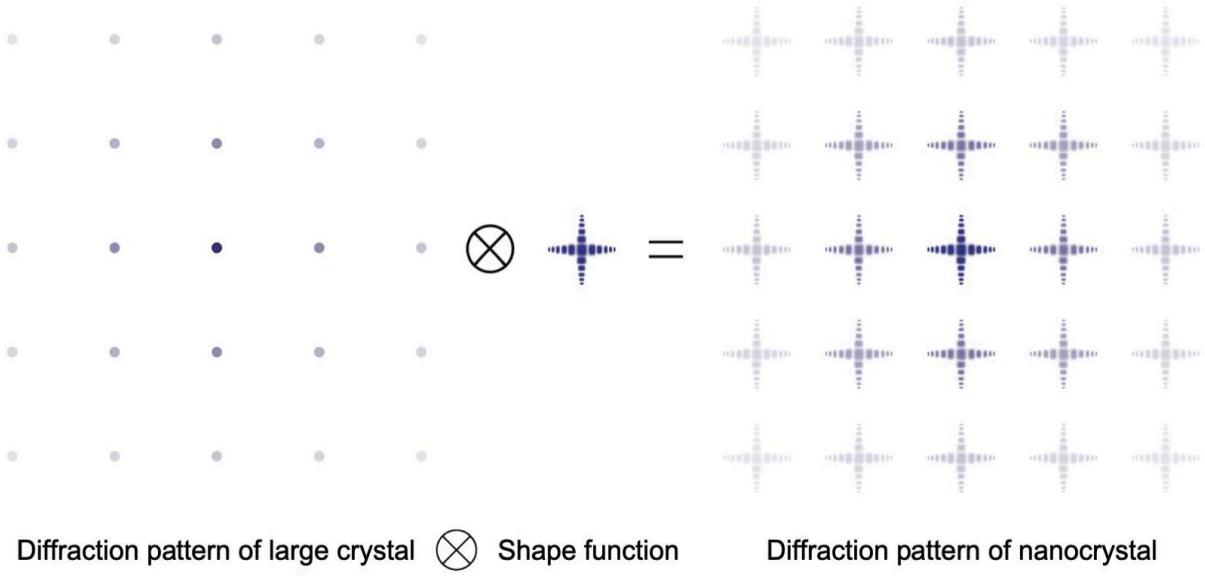
This strict condition is substantially relaxed in Bragg CXDI. Now, in order to understand why, we resort once more to the convolution theorem to explain the appearance of diffraction data in Bragg CXDI. Consider a perfect large scale crystal here on the left. Now, ignoring crystallographic errors, a nanocrystal is simply this large crystal multiplied by a spatial map in which the values are one, inside a region equating to that of the nanocrystal, and zero, outside the region's boundaries.

Notes

Summary

5m 00s

Bragg CXDI – a perfectly regular starry firmament



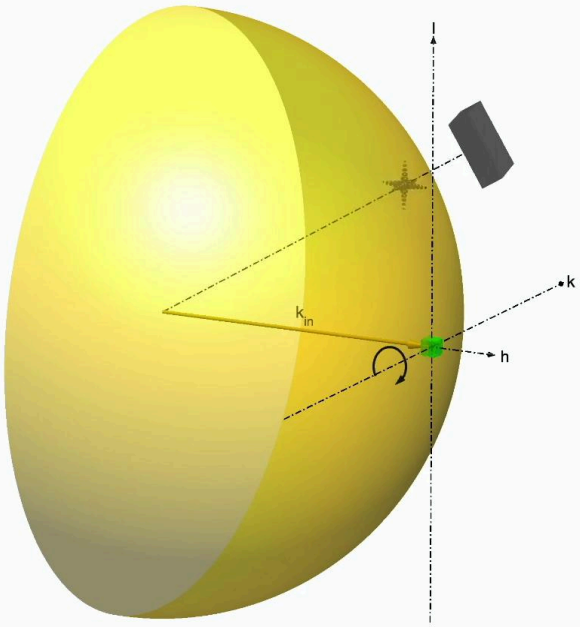
Now, remember that the convolution theorem states that the Fourier transform of the product of two functions, A and B, is equal to the Fourier transform of one convoluted by the Fourier transform of the other. Hence, by applying the convolution theorem to the two functions of the large crystal and the nanocrystal's physical boundaries, the former generates a Fourier transform equal to a perfect diffraction pattern of point-like diffraction peaks, while the latter produces some kind of so-called shape function, i.e., the Fourier transform of the boundary defining the shape of the nanocrystal. For the case of a cubic nanocrystal, the shape function is the Fourier transform of a cube or a so-called sinc-squared function in three dimensions, shown schematically here in two dimensions. Convoluting the ideal diffraction pattern with this shape function generates duplicates of that shape function with intensities determined by those of the Bragg peaks in the ideal diffraction pattern. This is the diffraction pattern of a nanocrystal.

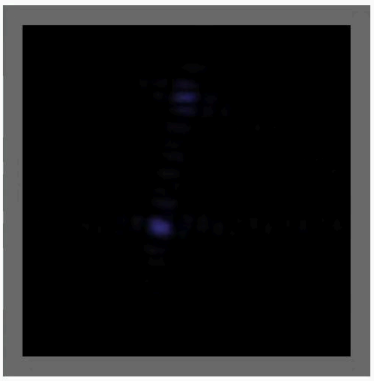
Notes

Summary

5m 37s

Bragg CXDI





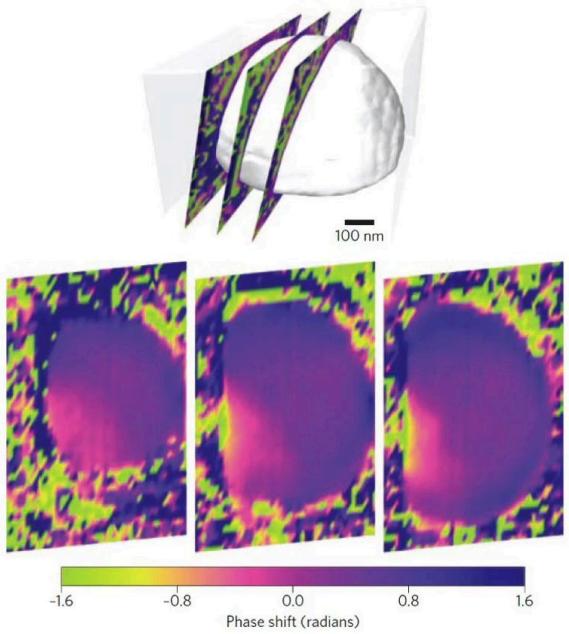
How can we now exploit this to our gain in Bragg CXDI? In this cartoon, we show the front half of the Ewald sphere and one Bragg peak, which for the dimensions we have chosen, happens to be the 1-bar-1-2 peak. This Bragg peak has a non-zero width in reciprocal space because of the nanocrystal's shape function, which we have chosen to be that of a cube. If we rotate the nanocrystal around the horizontal axis perpendicular to the instant radiation, the chosen Bragg peak will pass through the Ewald sphere and the structure factors can be recorded on a detector. Depending on the extent of the features of the shape function, the entire diffraction pattern can be captured after only a few degrees rotation, far less than in the forward-scattering direction. This means the specifications on the wobble and drift of the rotation axis can be substantially relaxed.

Notes

Summary

6m 55s

Applications of Bragg CXDI



M.A. Pfeifer *et al.*, Nature **442**, 63–66 (2006) https://www.nature.com/articles/nature04867

- Phase information arises from strains within the crystal
  - ⇒ Bragg CDI yields high resolution 3-D images of strain from within a nanocrystal in direction of Q
- Bragg diffraction away from (000) direction
  - ⇒ scattering object does not need to be physically isolated
  - Nonperiodic substrates or those with different lattice constants will be invisible to the diffraction process
  - ⇒ use Bragg CXDI to study the impact of an interface with the nanocrystal
- Several different Bragg spots (different Qs)
  - ⇒ 3-D strain tensor within nanocrystal

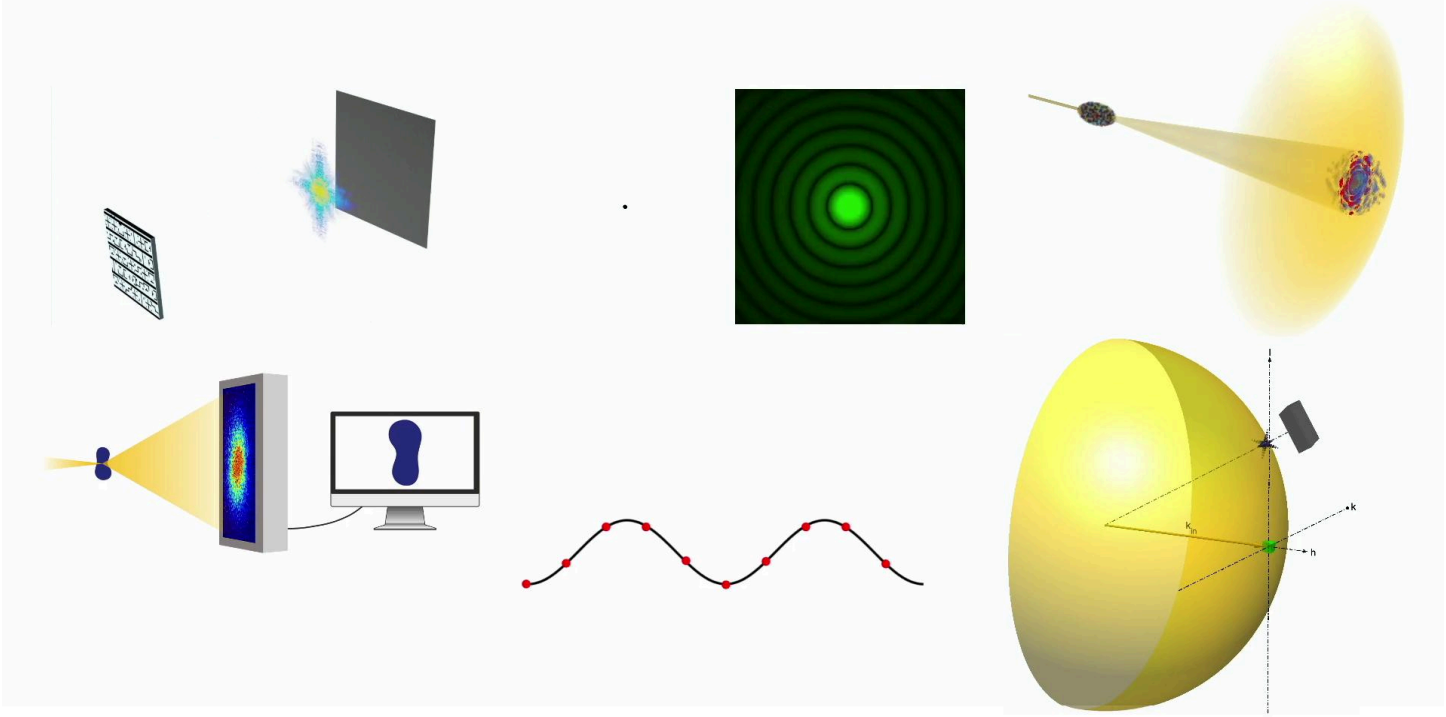
Phase information can be gained through strain fields within crystals, meaning that Bragg CXDI can provide high-resolution 3D images of these strain fields within nanocrystals in the direction of the scattering vector Q. Note that because the diffraction pattern is not in the forward-scattering direction, the scattering nanocrystal doesn't need to be physically isolated and can, for example, be embedded in a matrix of any material, amorphous or crystalline, as long as that material doesn't generate diffraction intensity in the same region of reciprocal space as the nanocrystal. Hence, the interface of nanoscale crystalline objects with other materials can also be studied through Bragg CXDI. By recording several different Bragg spots with different Miller indices, a 3D strain tensor within the nanocrystal can be generated, as shown here in a pioneering work on Bragg CXDI of lead nanocrystals grown on silica substrates and detailed in the provided reference.

Notes

Summary

8m 02s

Summary of this section



In summary, we have discussed the principles of CXDI and the important concept of speckle. We then consider the concepts of oversampling and redundancy before considering in more detail both conventional CXDI for non-crystalline samples and Bragg CXDI used for nanocrystalline samples.

Notes

Summary

9m 10s

In the next section...

In the next and final section of this course, we will cover ptychography, ptychographic and laminographic tomography, higher dimensional lensless imaging, and X-ray photon correlation spectroscopy.

Notes

Summary

9m 34s