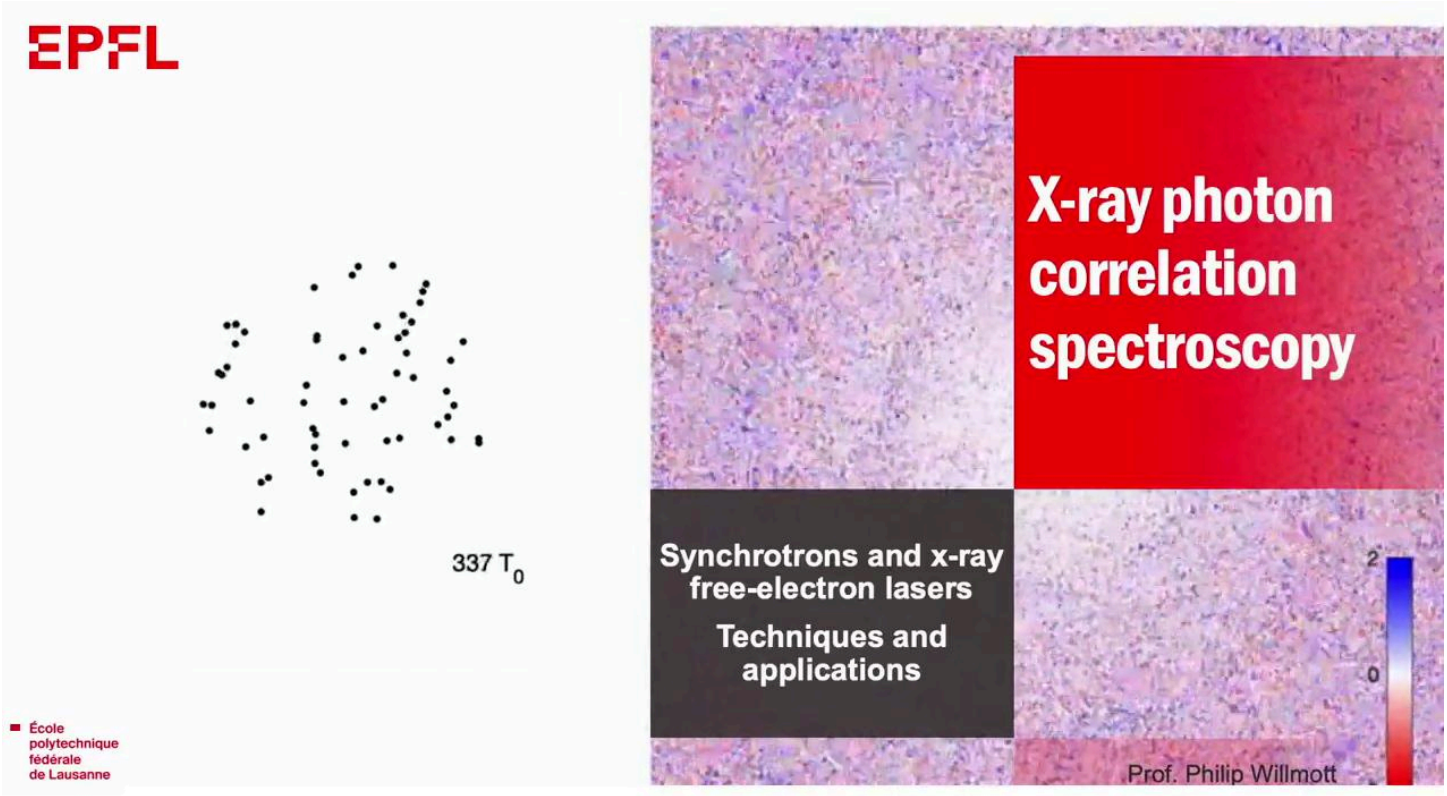
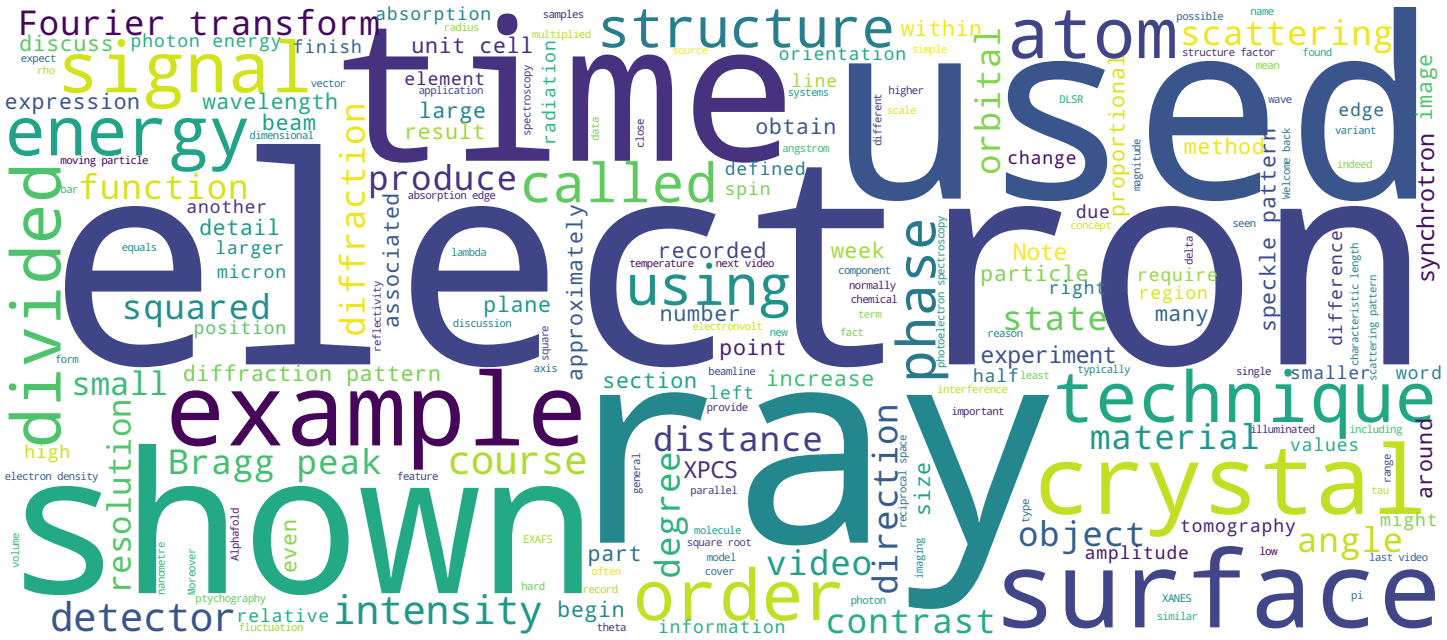




Search MOOC

Video

EPFL

Contents and objectives of this video

- Purpose and applications of XPCS
- Description of XPCS
- XPCS at DLSRs

Hi again. In this very last video of this course, we discuss the techniques of X-ray photon correlation spectroscopy, or XPCS. We'll look at which types of studies might profit from XPCS, then look at XPCS in some more detail. We'll finish with a brief summary of how XPCS is likely to develop since the operation of DLSRs began in the last couple of years.

Notes

Summary

0m 05s

XPCS – Introductory comments

- Used in materials science, soft-matter science, and condensed-matter physics
- Study dynamic behavior of systems on the nanoscale down to μs or shorter
  - Insights into temporal fluctuations and correlations of structures within a material
- Suited for complex systems
  - Soft matter
  - Colloids
  - Glasses
  - Phase transitions

XPCS can be used in many fields of the natural sciences, such as in colloid research, protein dynamics, vitreous glass systems, and the biosciences, to name just a few examples. It can be applied to any system consisting of nanoscale features that are liable to move with characteristic durations down to the microsecond scale, or even potentially shorter still since the advent of diffraction-limited storage rings. In this manner, insights into the internal dynamics and structural correlations of materials can be gained.

Notes

Summary

0m 33s

XPCS – Introductory comments

- XPCS: analysis of progress of coherent x-ray scattering patterns (“speckle movies”) to gain insights into the movement, interactions, and fluctuations of nanoscale structures.
  - Information about spatial arrangement of scattering features within sample
  - How they evolve over time
  - Analyze temporal intensity fluctuations

Essentially, an XPCS experiment is nothing more than a speckle movie, in which the progress of the scattering pattern produced by coherent illumination of the sample is recorded. This provides information about the spatial arrangement and temporal dynamics of scattering features within that sample.

Notes

Summary

1m 08s

XPCS – Introductory comments

- Why include XPCS in the imaging section?
  - Sits somewhat uncomfortably in this section as one does not normally produce any real-space images - real-space dynamics are simply inferred from those recorded in reciprocal space
  - However, it uses the phenomenon of speckle to extract information about dynamical processes
  - Doesn't fit anywhere else either!

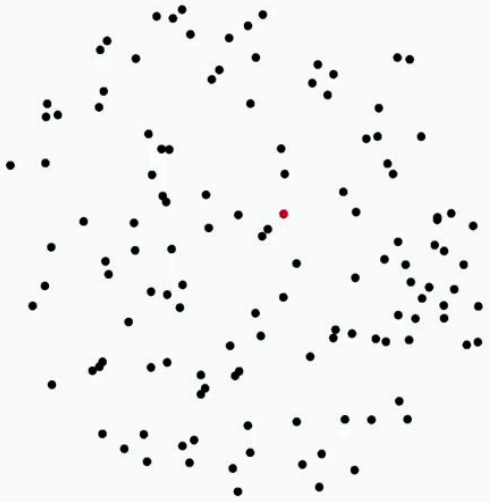
Before we continue, I feel it behoves me to explain why XPCS has been included in a section which is nominally on imaging. One can, of course, argue the speckle movie, once Fourier transformed, produces a real movie, which itself is nothing more than a succession of still images. But normally, one doesn't attempt to Fourier transform each individual frame. That's the point of XPCS, to extract real space dynamics directly from the reciprocal space. Nonetheless, because it exploits speckle, which is used in all the techniques described in this week's videos with the exception of scanning tensor tomography, I decided to include it here. It sure as hell fits even less well anywhere else.

Notes

Summary

1m 28s

Simple case – one itinerant particle in an ensemble



- Ensemble of randomly distributed particles
  - All stationary except one
  - Itinerant particle moves with random "Brownian" motion
  - What happens to the speckle pattern of such a system?

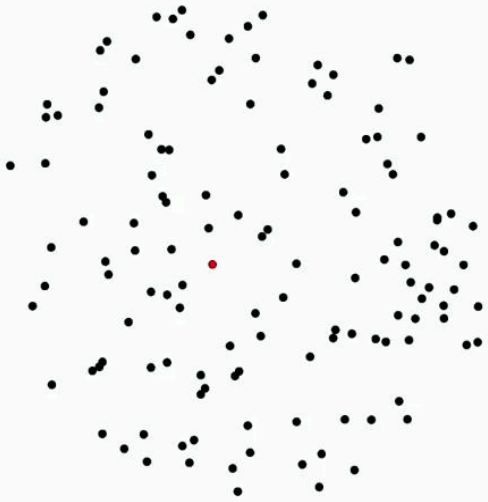
Let's now begin by considering a very simple case of a single particle moving randomly among an ensemble of 127 other identical particles that are frozen in position. What can we expect to happen to the speckle pattern of such a system?

Notes

Summary

2m 19s

Simple case – one itinerant particle in an ensemble



- Very weak fluctuations
- Mostly visible in dark nodal circles
- Why?
  - Small differences in FT most prominent where FT is weakest
  - Similar to CTR signal in SXRD…

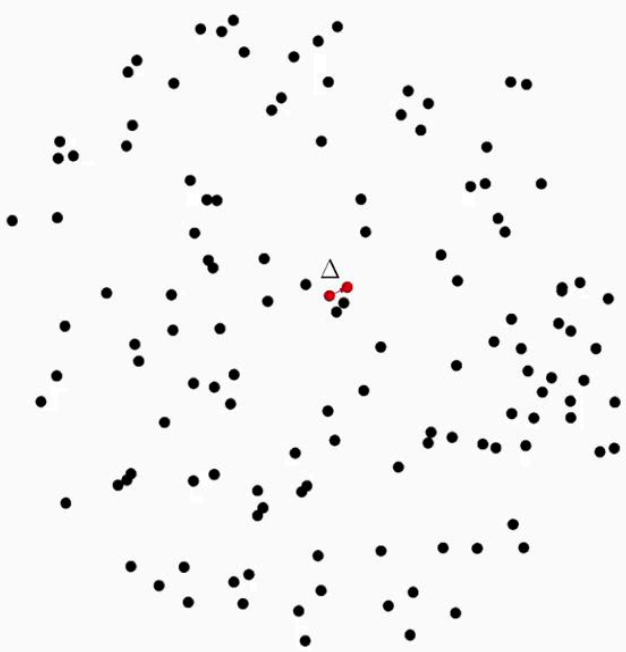
If you look extremely carefully, you might see very weak fluctuations, which are most evident in regions of the dark rings of the envelope Airy function. It's not surprising that the fluctuations are so difficult to detect. As the single-moving particle, only contributes a small amount to the total structure factor, composed of contributions from the other 127 particles. And don't forget, this is then squared to produce the intensity. The reason why the fluctuations are clearest in the dark nodal regions it's here, that the individual contributions are small, and hence from the moving particle, it can have a relatively significant contribution.

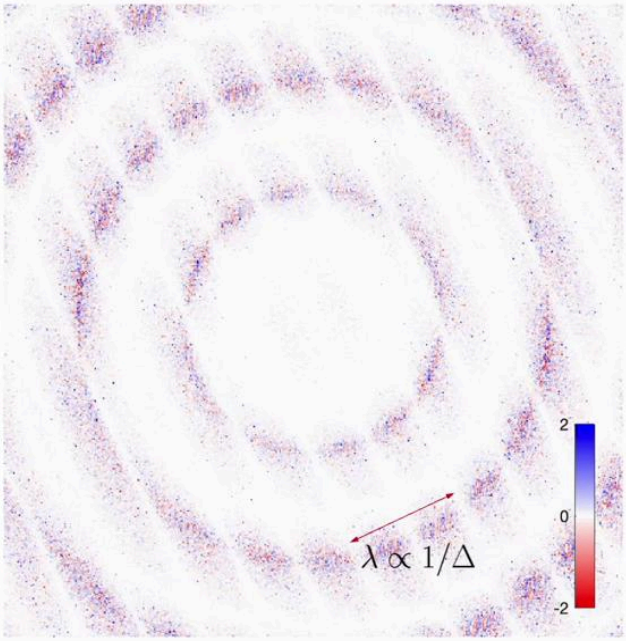
Notes

Summary

2m 38s

Simple case – one itinerant particle in an ensemble





Let's now increase the contrast by subtracting one speckle image from the successive image and dividing by their average, shown here mathematically. Now the fluctuations are much more apparent. What is also clearly seen is that the difference speckle pattern is modulated by a peculiar wave function which changes its wavelength and direction for each new frame. What's going on here? Let's consider any two successive frames, such as these two. The particle has moved a distance delta, shown by the red arrow. The corresponding different speckle pattern shows a wave that points in the same direction. Analysis of other pairs of frames demonstrates that, moreover, the wavelength of this wavelike modulation is inversely proportional to the distance that is moved between successive shots, delta.

Notes

Summary

3m 27s

Simple case – one itinerant particle in an ensemble

- Why the modulation?
- In general

$$\mathcal{F}[f(x+a)] = \mathcal{F}[f(x)]\ \exp(-ika)$$

- Hence

$$\Rightarrow \mathcal{F}(\text{circle}) \rightarrow \mathcal{F}(\text{circle})\cos(kd)$$

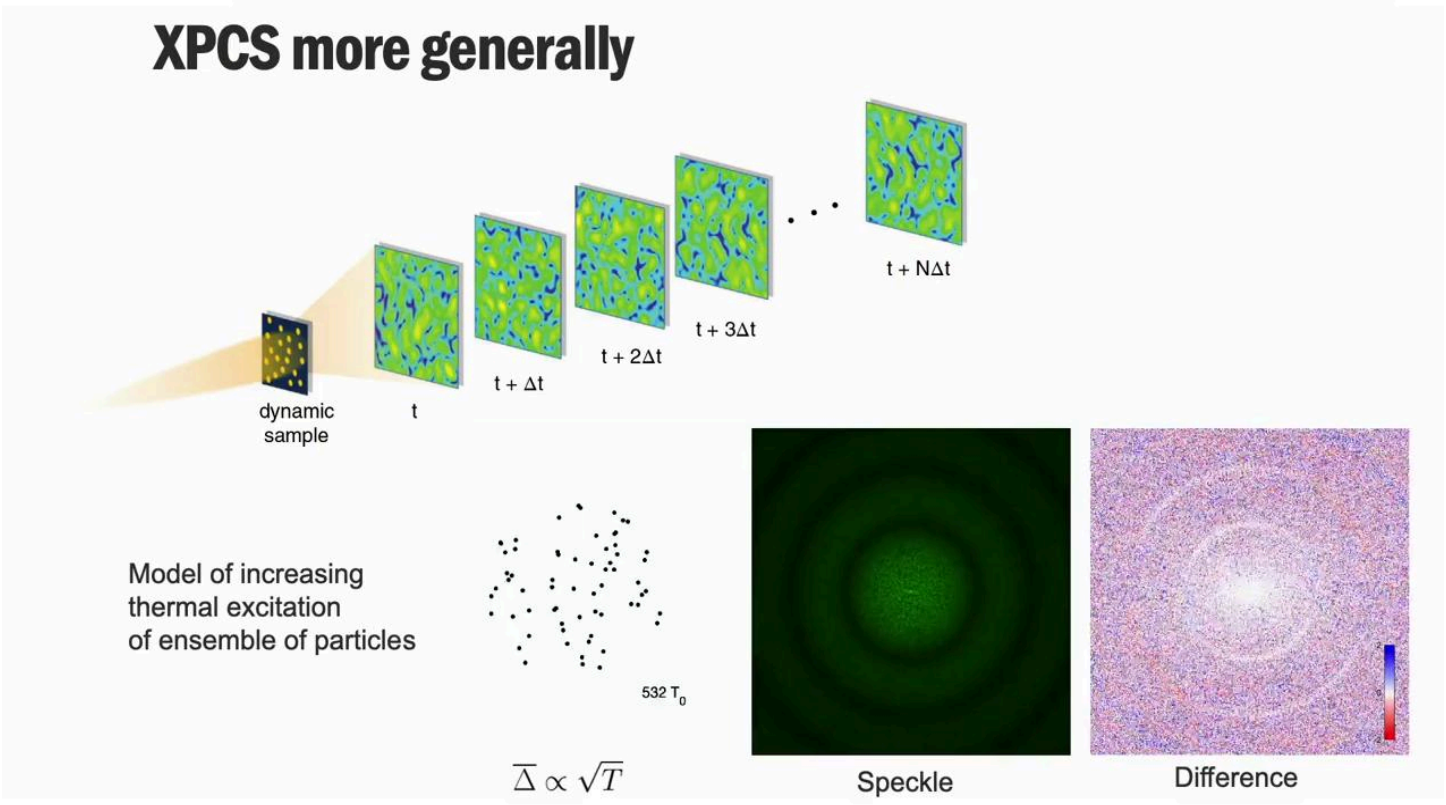
  - Signal modulation defines difference in temporal images in XPCS

Why do these different speckle patterns show this modulation? The reason has to do with the fact that the Fourier transform of a function here, the moving particle, when shifted by a distance a, produces a Fourier transform of that particle multiplied by the exponent of minus ika, this is just a rule for Fourier transforms. Hence, the FT of the circle is modulated by ikd, or for the symmetric function, simply cosine kd. The take-home message here is that the single modulation defines the difference in the system as a function of time. With many moving particles, many sinusoidal waves with different directions and wavelengths modulate the signal, thus changing the speckle pattern much more strongly.

Notes

Summary

4m 32s



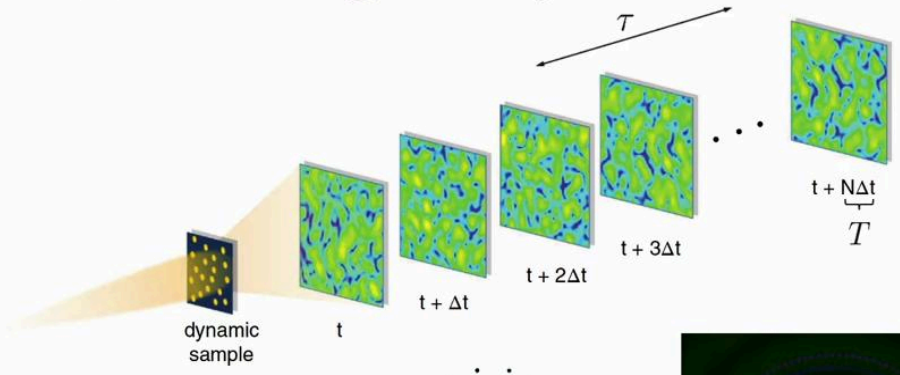
If we consider now a model where all the particles move thermally, whereby the average movement is proportional to the square root of the temperature, and starting with a very low temperature, we will see that at first the speckle pattern changes only marginally, and the difference pattern has mainly zero intensity, shown here as being white. With increasing temperature, however, the difference pattern becomes dominated by large fluctuations.

Notes

Summary

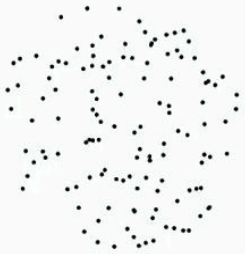
5m 24s

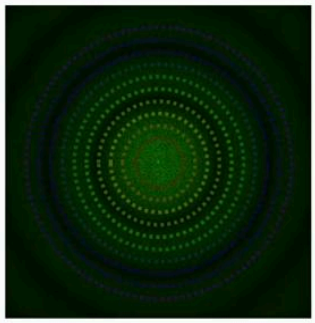
XPCS more generally

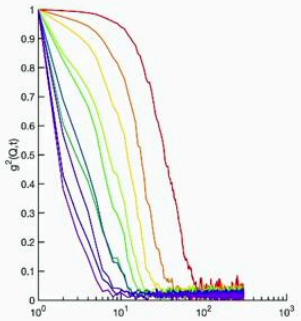


$$g^{(2)}(\mathbf{Q},\tau) = \frac{\langle I_t\, I_{t+\tau}\rangle_T}{\langle I_t\rangle_T^2}$$

Normalized intensity correlation function







See also e.g., A.R. Sandy *et al.*, https://www.annualreviews.org/doi/pdf/10.1146/annurev-matsci-070317-124334 and G. Grübel *et al.*, https://link.springer.com/referenceworkentry/10.1007/978-1-4020-4465-6_18

We can quantify this still further by plotting the normalised intensity correlation function as a function of time, or here, frames. This is defined as being the average of the intensity I at time T, multiplied by the intensity for a later time, T plus tau, over the entire recording time, large T, and divided by the square of the intensity at time T squared, also over the entire recorded time, large T. What this expression is doing is to look at how much the pattern has changed. If, for example, none of the particles at a time T plus tau are in the same position as they were at time T, then multiplying these two particle distributions together will produce a zero result. In contrast, if the increase in time tau is made to be so small that the particles have had no time to move any significant distance, the multiple of IT and IT plus tau, will be unity if divided by IT squared. This is now shown for different Q values. We can expect for low Q, which probes large characteristic lengths, that it will take some time for the correlation function to decay. In contrast, for large Q, sampling small distances, it will drop rapidly as the particles drift away from their original positions. Sure enough, this is the case. In this manner, different length scales are probed in XPCS.

Notes

Summary

5m 52s

XPCS more generally

- Correlation analysis: temporal correlations of speckle fluctuations
  - Analysis of how I(Q) correlations change over different time intervals
    - Relaxation timescales
    - Diffusion coefficients
    - Other dynamical properties of the material.

- XPCS sample requirements
  - Well-suited for studying samples with relatively slow dynamics, with correlation timescales of the order of ms to seconds – DLSRs should significantly extend temporal resolution, see next slide
  - Sample should be relatively uniform
  - Sample should have scattering features that are smaller than the x-ray coherence length

Such correlation analysis of temporal changes in the speckle can be used to probe different characteristic lengths and timescales, resulting in valuable information regarding relaxation rates, diffusion coefficients, and many other dynamical properties of materials. XPCS is thus well suited for systems which have dynamics which can be tracked on the minimum timescale, which until recently has been in the microsecond region, and that only for point detectors following a very small region corresponding to a typical speckle size. It's expected that XPCS at DLSRs will significantly extend the time resolution as discussed briefly in the following slide. Secondly, the sample should be relatively uniform, or at least that part of it being illuminated by coherent X-rays, which also requires that the scattering features should be significantly smaller than the transverse coherent lengths available at the beamline.

Notes

Summary

7m 31s

XPCS at DLSRs

- XPCS limitation until recently: modest temporal resolution.
- XPCS measures the correlation between two photons and not interference of a photon with itself (as in 'normal' diffraction)
  - ⇒ Signal-to-noise ratio scales with intensity rather than the scattering amplitude

$$S/N = F_{\mathrm{coh}}(T\,\Delta t\;n_x n_y)^{1/2}$$

    - S/N of correlation function between two speckle patterns separated by Δt in an experiment over a total time of T. F_{coh} is the coherent flux
- X times increase in F_{coh} thus allows an X^2 reduction in minimum Δt
- DLSRs increase F_{coh} by ca. 50. Δt might decrease by over 1000 to near ns regime!!
- Many systems with characteristic time-scales are now becoming accessible
  - Magnetic spin reorientation (GHz)
  - Ferroelectric-domain switching (100 MHz)
  - Structural phase transitions (100 kHz to GHz)

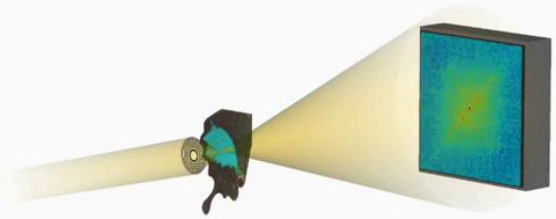
The resolution of XPCS, especially if area detectors are used, has been very modest until recently, rarely ducking under the millisecond regime. Importantly, XPCS measures the correlation between two photons, and not interference of a photon with itself, as is the case in normal diffraction. The signal to noise ratio thus scales with the intensity rather than the amplitude, and is proportional to the coherent flux and the square root of delta T, the time separation between successive measurements. This means that an increase by, let's say, a factor of 50 in the coherent flux at a DLSR will allow delta T to be reduced by over three orders of magnitude, potentially allowing nanosecond scale dynamics to be studied. Processes that might then be accessible to XPCS might include magnetic spin reorientation, ferroelectric-domain switching, or structural phase transitions, to name just three phenomena that have until now been entirely excluded from XPCS studies.

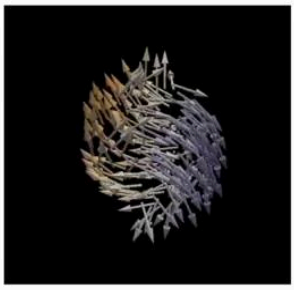
Notes

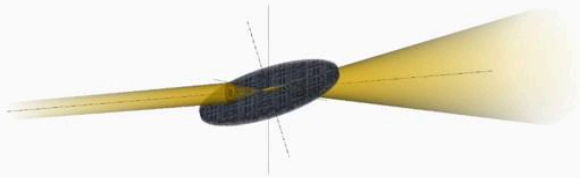
Summary

8m 34s

Summary of this section





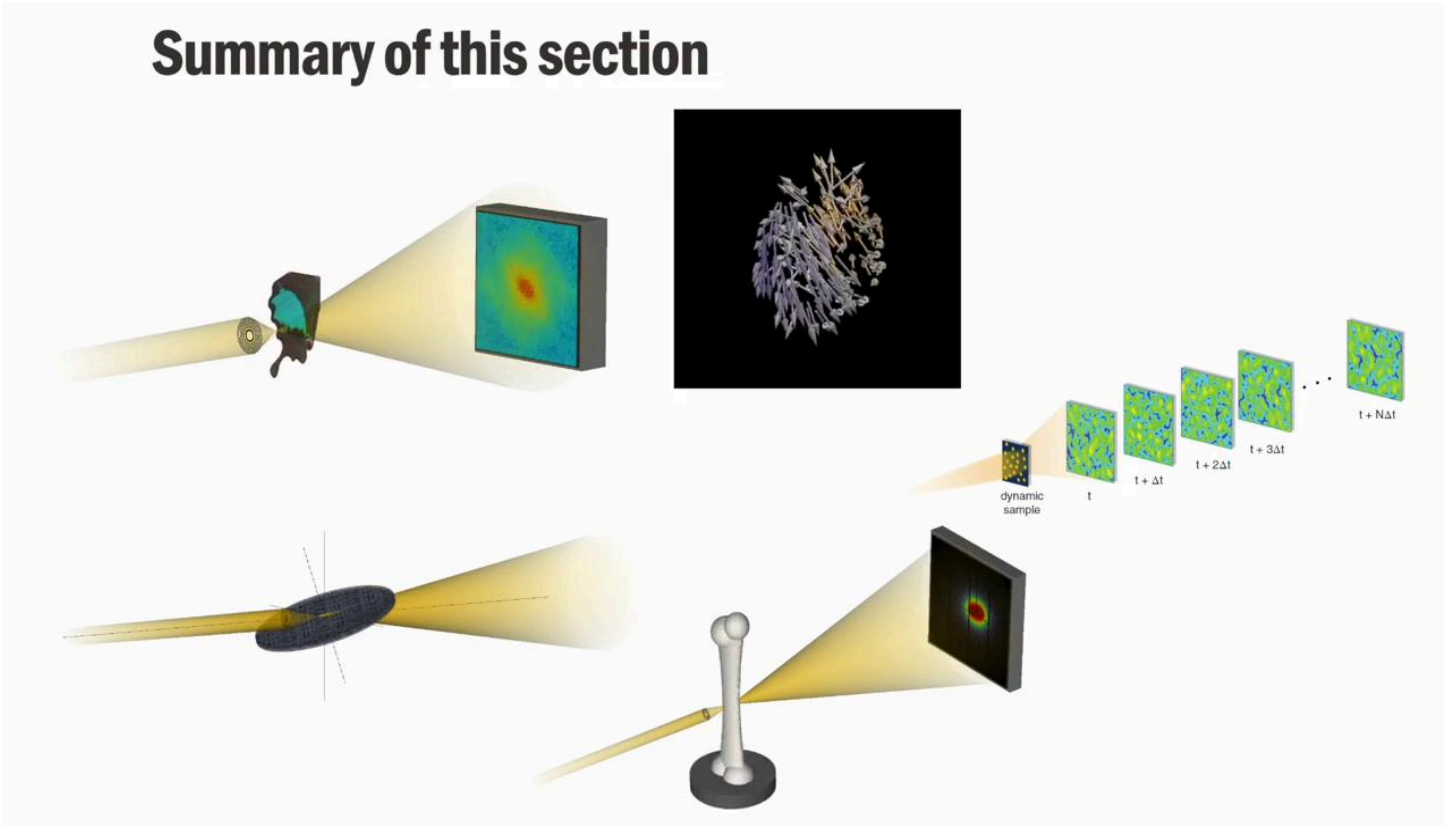


In summary, this last section has concentrated on ptychography and laminography, higher-order imaging techniques such as vector tomography and scanning SAXS tensor tomography.

Notes

Summary

9m 47s

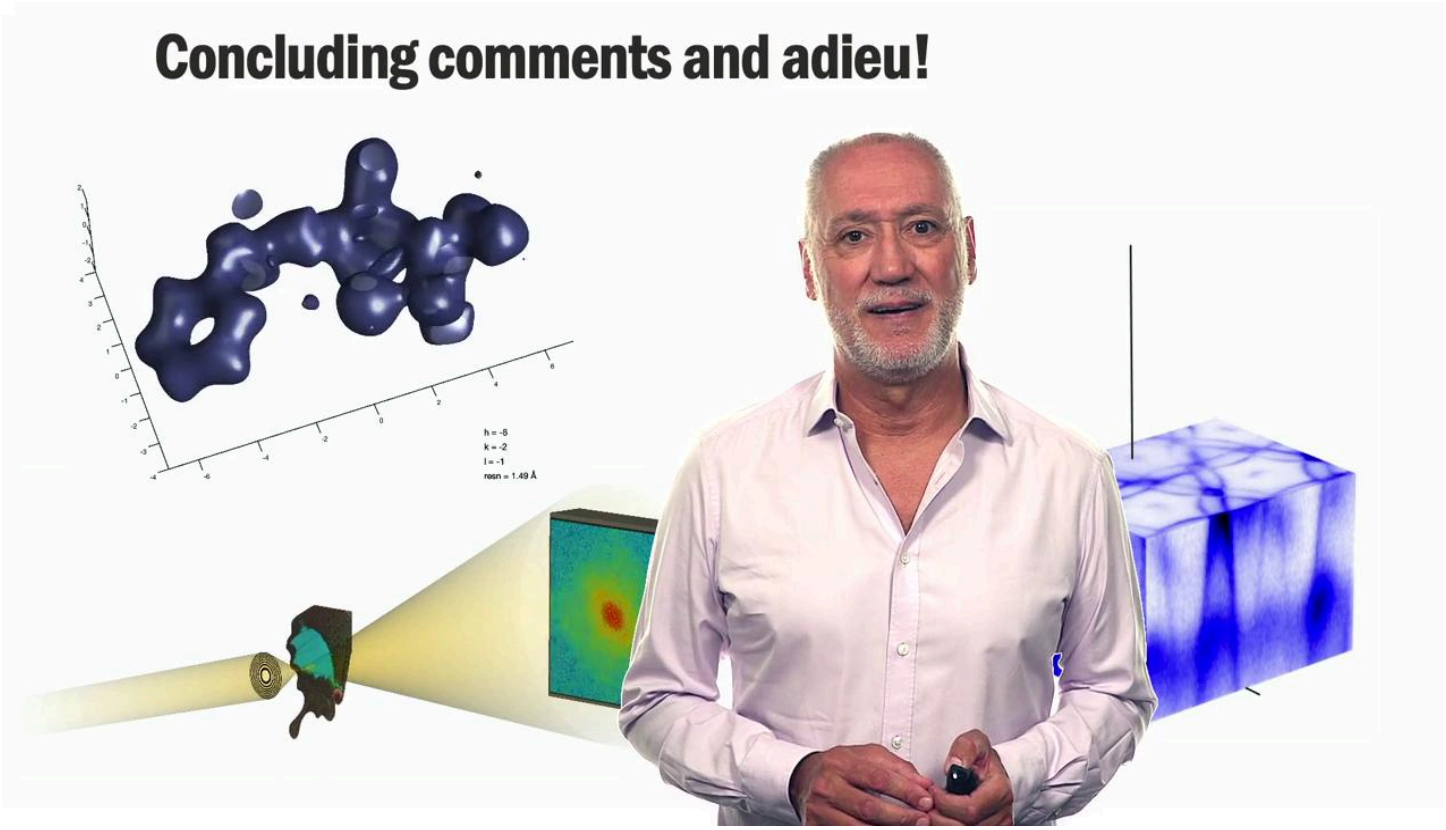


We finished in this last video with an introductory description of X-ray photon correlation spectroscopy.

Notes

Summary

10m 00s



So, we're done. Thank you very much for those of you who have seen this course, and hopefully also this sister course through. Needless to say, many, in fact, most of the methods and techniques covered in these last six weeks have, perforce of the limited time available, been described only very briefly and to a constrained depth and breadth. For many of these, I'm sure that you can find whole six-week MOOC courses or even longer, that concentrate on details of any one of these alone. There are, moreover, specialised summer schools and courses to be found all over the world. There is, of course, an overwhelming choice of literature in the form of journal reviews and textbooks, including my own, I should mention. Nowadays, more and more excellent online material in the form of interactive applets and so forth is available. That said, I hope that the broad strokes that have formed the character of this course will prove to be for at least some of you a solid foundation for exploring further the extremely exciting and burgeoning world of synchrotron and XFEL science. Thank you again, and goodbye.

Notes

Summary

10m 08s