The progress in dynamical studies by x-ray coherent scattering: example of critical fluctuations

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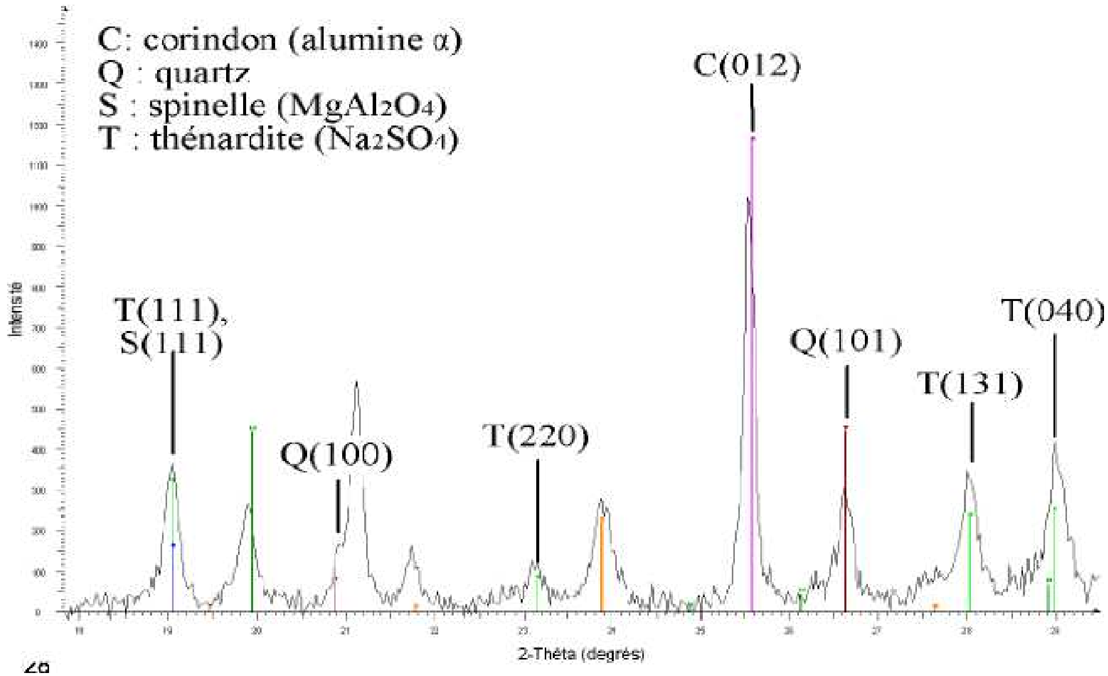
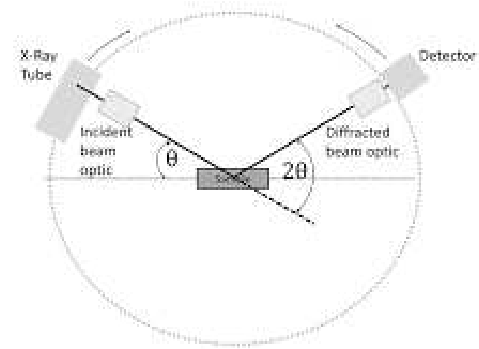
An x-ray experiment

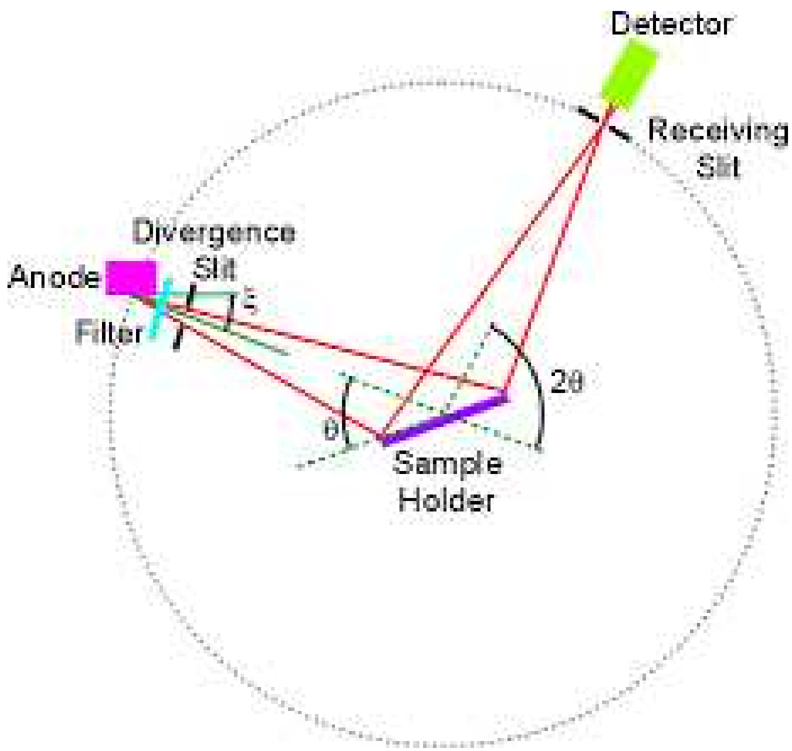




A typical powder diffraction setup

Here a mixture of various crystalline phases is studied. From the position of the Bragg peaks, one can identify theses and estimate the sample composition. These peaks correpond to interferences in the sample: a limited domain in the sample produces these interferences. It is called “the coherence volume”. The size of this coherence volume is connected to the x-ray source (and detection!).

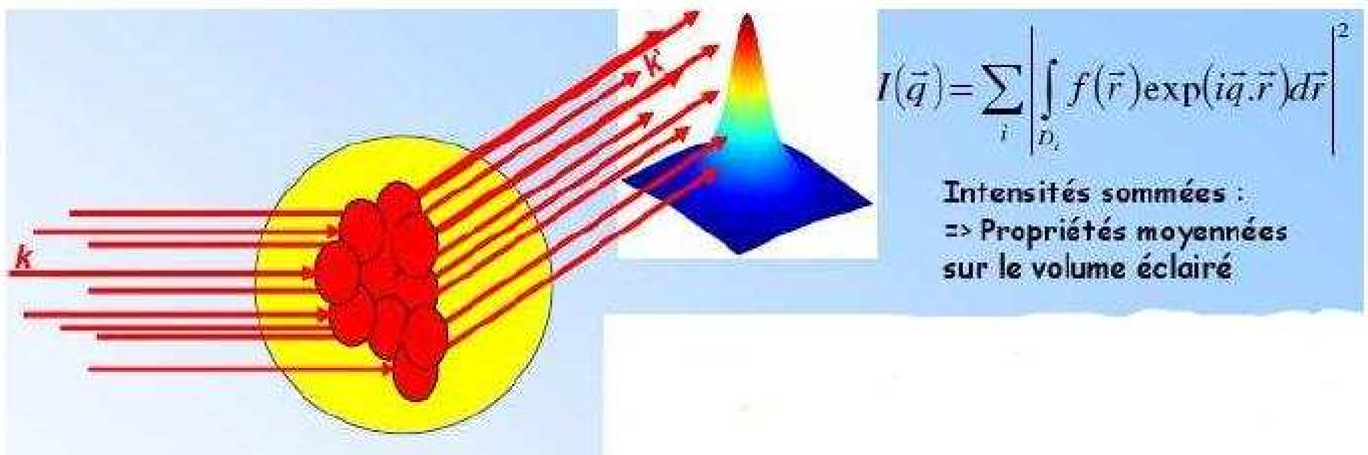
Domain of coherence?



Classical powder diffraction experiment:
In the beam transverse direction:
$\Lambda_t \simeq \lambda * L/\phi$, λ is wavelength, $\phi \simeq 0.3mm$ is the source size and $L \simeq 0.3m$ is the source-to sample distance: $\Lambda_t \simeq 0.15\mu$m.
In the longitudinal direction:
$\Lambda_l \simeq \lambda \times (\lambda/\Delta\lambda)$, $\Delta\lambda/\lambda$ is the beam monochromaticity. In a lab experiment: $\Delta\lambda/\lambda \simeq 1. \times 10^{-3}$ and $\Lambda_l \simeq 0.15\mu$m (have to take account of θ, the relevant length is $\simeq 2 \times \Lambda_l/sin^2(\theta)$).

An extremely small volume. This volume is directly connected to the experimental resolution.

Adding intensities

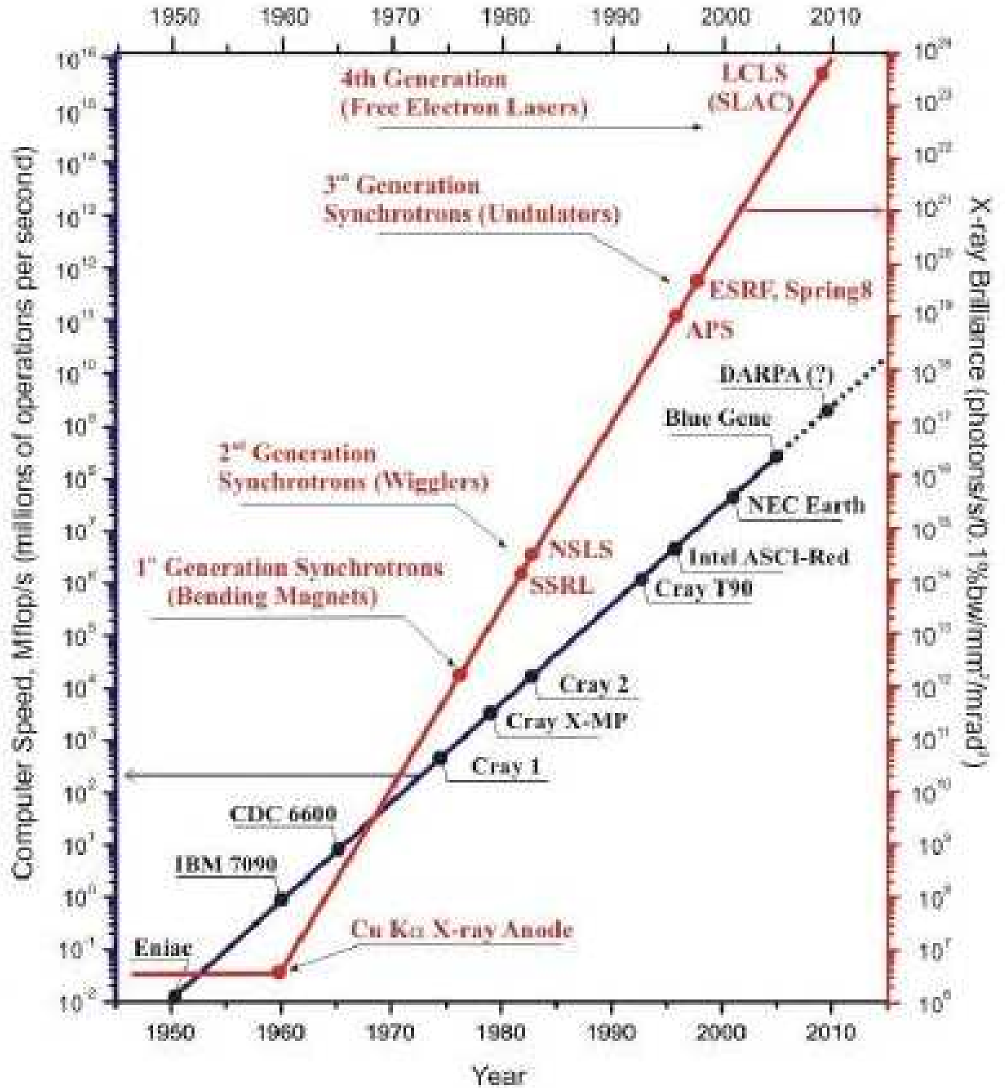


In a lab experiment, the intensities of a large number of coherent domains are added, and we measure an average.

The beam intensity in one domain of coherence can be estimated from the source brilliance, B, given in units of x-rays per second per source area (mm^2) per solid angle (mrad^2) for a bandwith of $\Delta\lambda/\lambda = 1. \times 10^{-3}$:

$$I_{coh} \simeq B \times \lambda^2 \times \Delta\lambda/\lambda$$

Historical x-ray Brillance



Brillance grew faster than CPU speeds!

For a CuKα x-ray tube, we have:

$1.*10^7 \times (1.4*10^{-4})^2 \simeq 0.2$ ph/s.

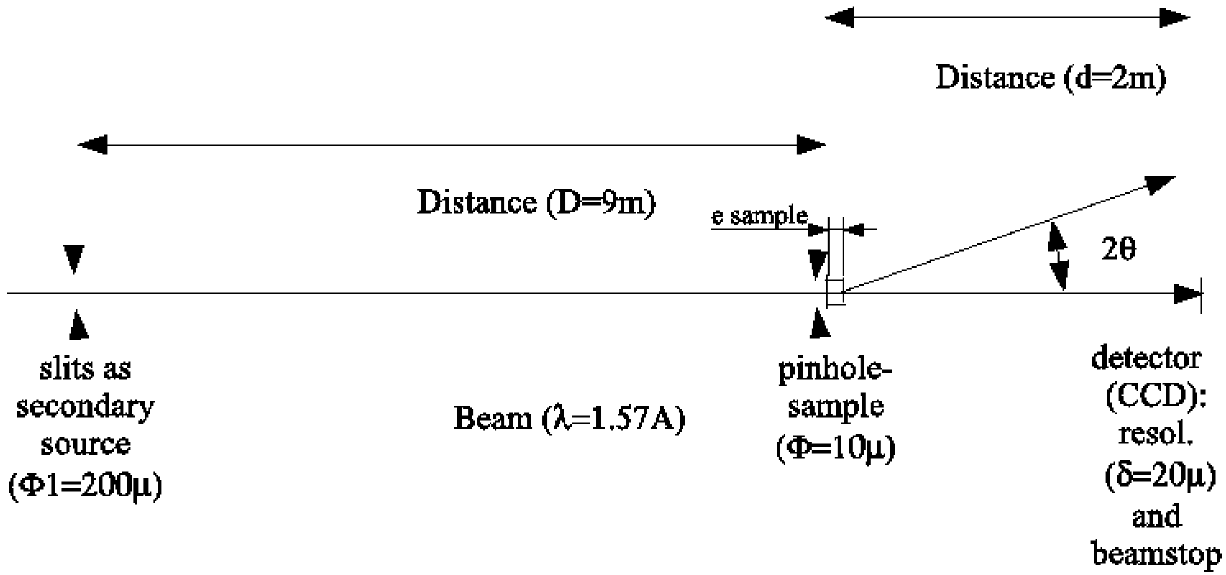
For a rotating anode, it is 1 ph/s

For an esrf undulator, it is $\simeq 1.*10^{10}$ ph/s.

Within 2 years, a factor 50 improvement!

With x-ray lasers, still three orders of magnitude!

Coherent measurements



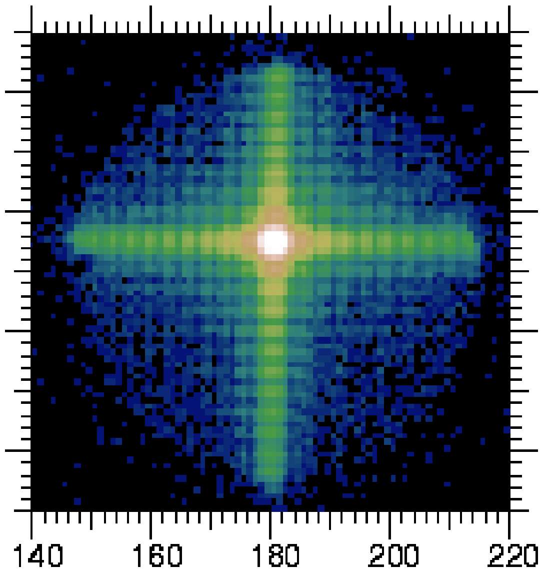
Experiments are similar to "Young's hole" experiments which were used in optics before the occurence of optical lasers. The esrf source is essentially incoherent. One selects an x-ray beam:

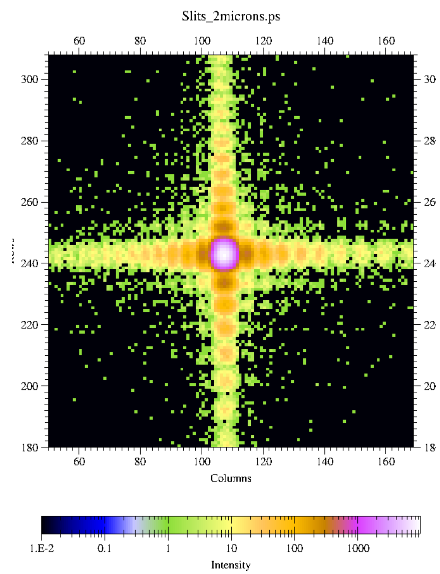
$$\phi \times \epsilon \approx \lambda$$

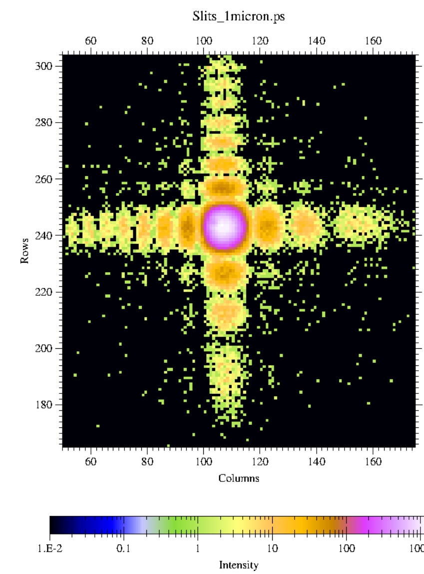
Roughly, it may be $\phi \simeq 10\mu$m and $\epsilon \simeq 10\mu$rad, and usually $\Delta\lambda/\lambda \simeq 1. * 10^{-4}$ ($\lambda^2/\Delta\lambda \simeq 1.\mu$m).

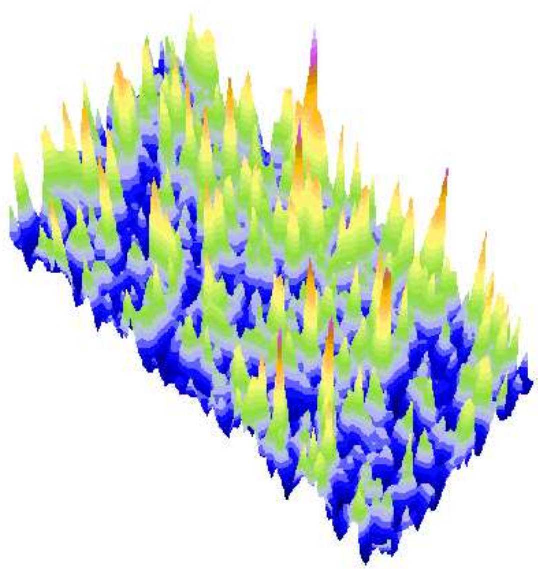
The same relations must hold for detection: high resolution is mandatory. Area detectors are used (here CCD with 20μm resolution).

Beautiful diffractions





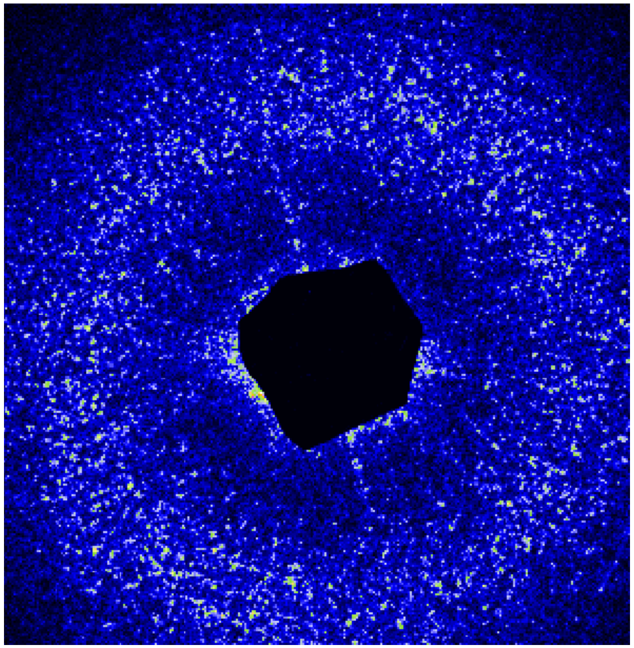


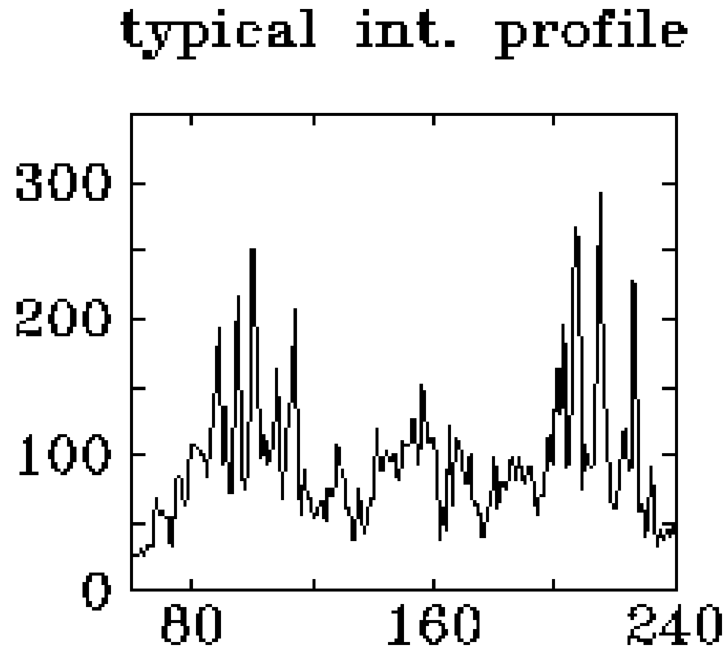


(top) scattering of slits with a coherent beam.

(left) speckles observed in a large region of a high resolution CCD detector .

How many coherence volumes?





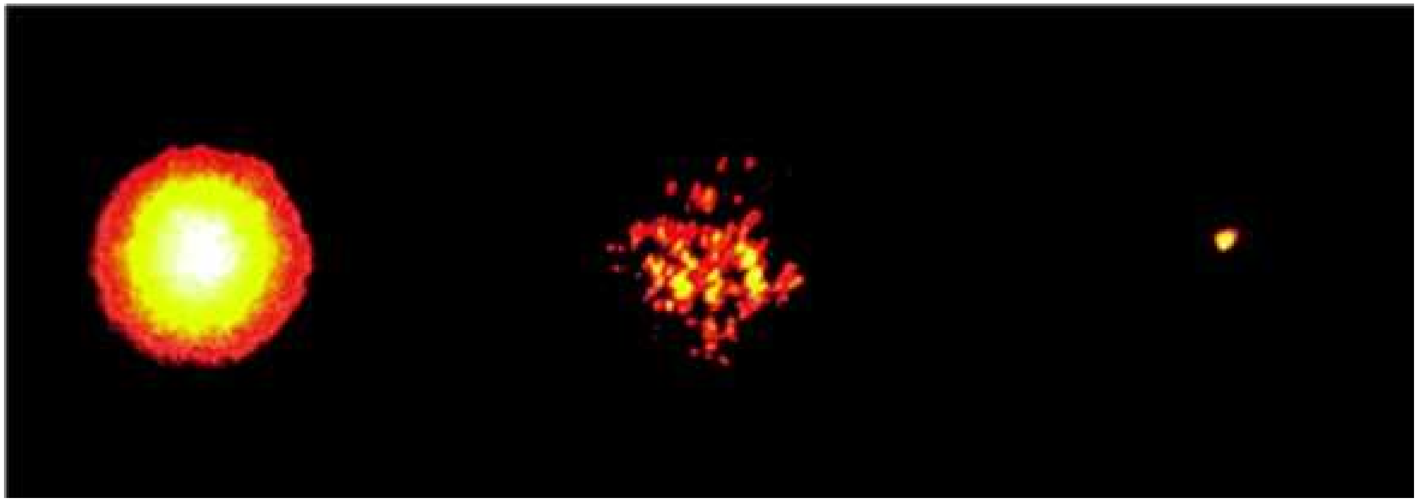
The speckle contrast can be discussed for a macroscopic isotropic sample:

$$\beta(q) = (< I(q)^2 >_q - < I(q) >_q^2 - < I(q) >)/(< I(q) >_q^2)$$

This contrast is roughly the ratio N between the coherence volume and the irradated volume ($\beta < 1.$, here $\beta \simeq 0.2 \simeq 1/N$ and $N \simeq 5$)

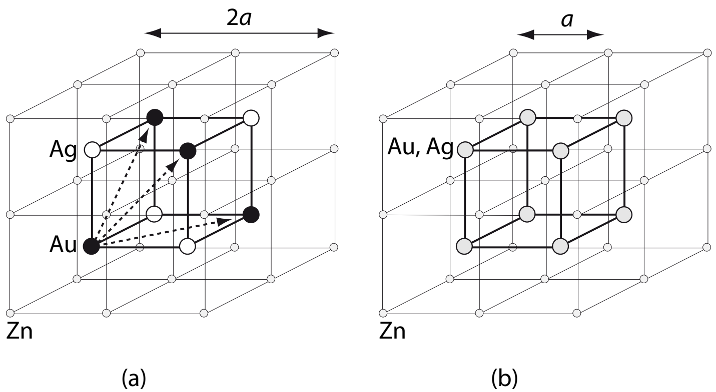
fluctuations: stars and speckles

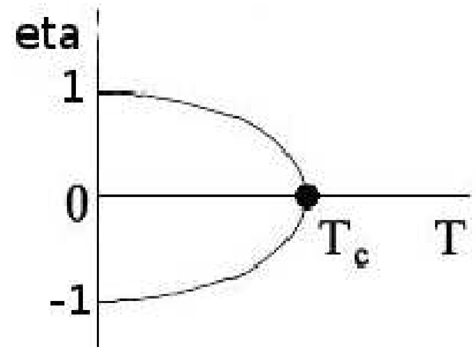
Here we consider the image of a star in a high resolution telescope.

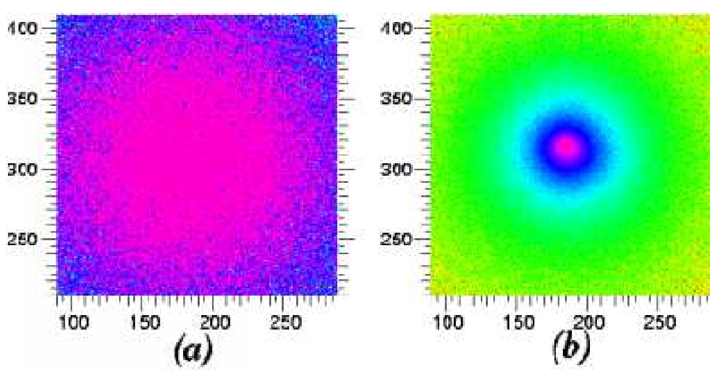


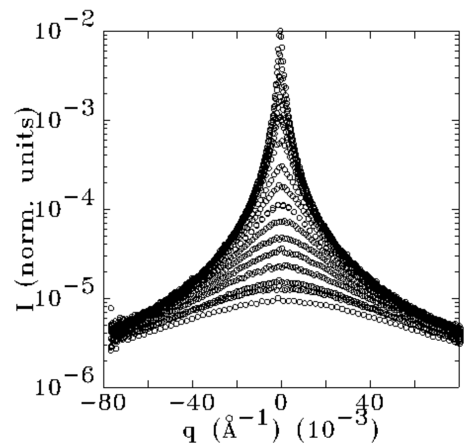
Three images of a star. The light of the star in fully incoherent, but at the distance and resolution (a 4 m telescope) of the figure, it can be considered as a purely coherent point source. By crossing the atmosphere, the image of the star is blurred by atmospheric fluctuations (left). For short time exposure, we obtain speckles, a "photograph" of the fluctuations (middle). Right: the image of the star with an adaptative optics. This image has the same extension as the speckles.

Ordering transition in $AuAgZn_2$









An ordering transition, inducing a (1/2 1/2 1/2) superstructure peak ($\eta = c_{Ag} - c_{Au}$, intensity $\propto \eta^2$) and strong isotropic fluctuations, observed in an area detector, diverging in the vicinity of $T_c \simeq 336°$C.

A typical "Ising" system.

Static scaling laws

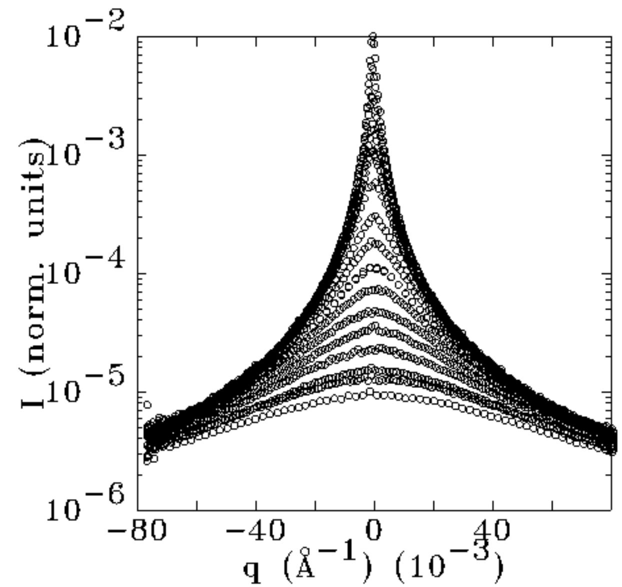
The measured intensity $I(q,T)$ correponds to fluctuations for $T > T_c$, and scales like:

$$I(q,T) = k((T - T_c)/T_c)^{-\gamma} \times G(q\xi)$$

with:

$$\xi \propto ((T - T_c)/T_c)^{-\nu}$$

This system is said to belong to the “Ising universality class”, with $\gamma \simeq 1.241$ and $\nu \simeq 0.631$

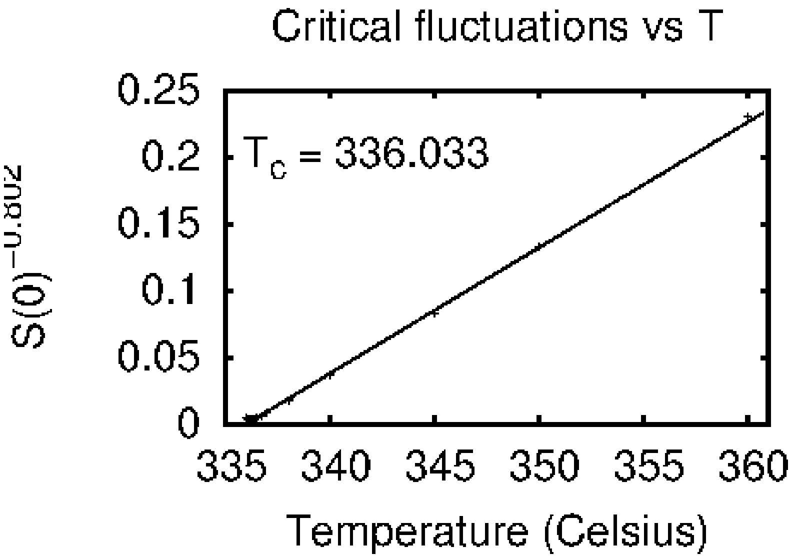


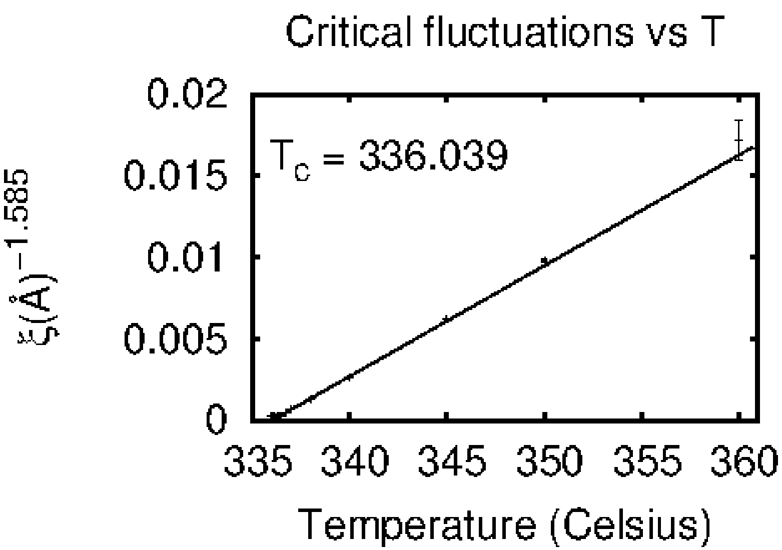
The Ising Hamiltionian of nearest neighbour interactions: $H = \Sigma s_i s_j$ is equivalent to the field theoretical model of Landau-Wilson:

$$H(\phi) = (\nabla\phi)^2/2 + r_0(T)\phi^2/2 + u_0\phi^4/4! \text{ for } r_0(T) \simeq 0$$

Both models give the same values of ν and γ

critical behaviour

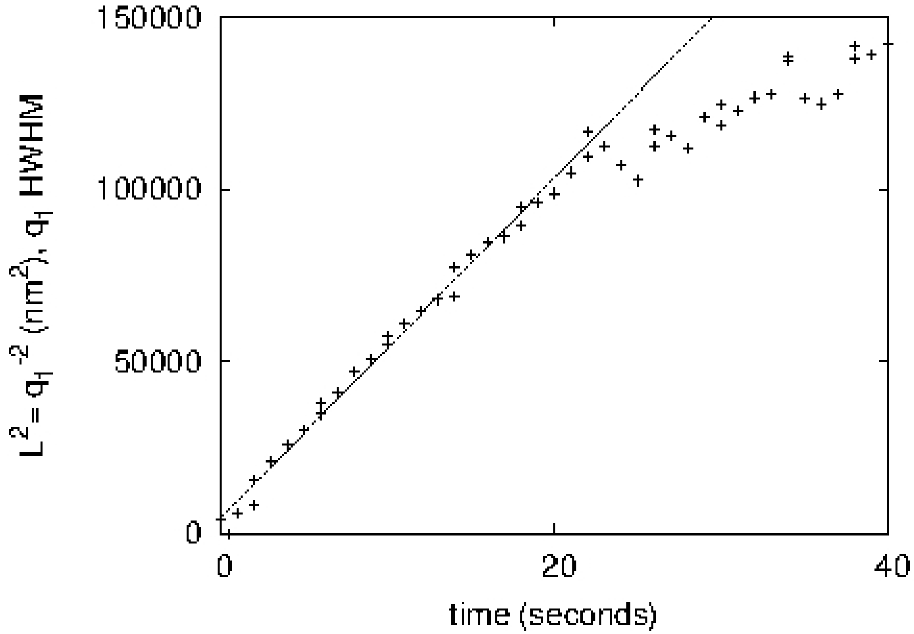




These two linear fits show that the Ising static exponents correspond to this system and provide a precise estimate of the second order transition:

$$T_c \simeq 336.035(10)$$

Coherence and dynamics



The dynamics of this system is equivalent to the model of the second order phase transition with "non conserved order parameter", model A of Hohenberg et al/1/.

For $T < T_c$, the classical behaviour of size increase of ordered domain size: L: $L^2 = Dt$ was verified.

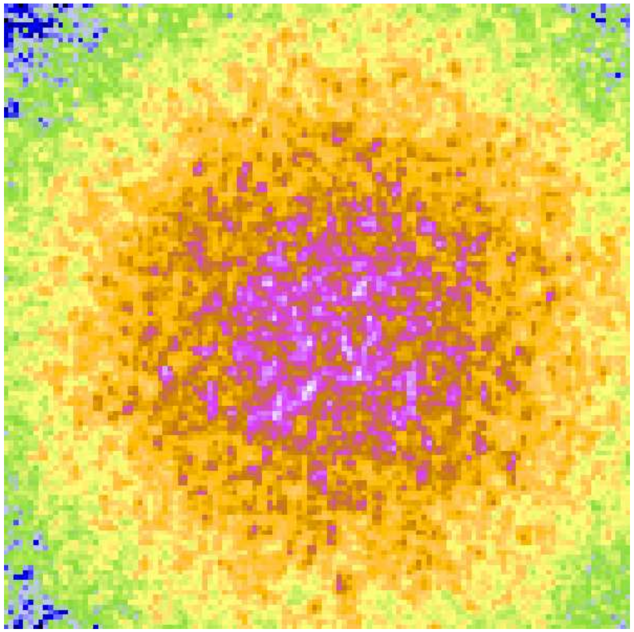
Writing:

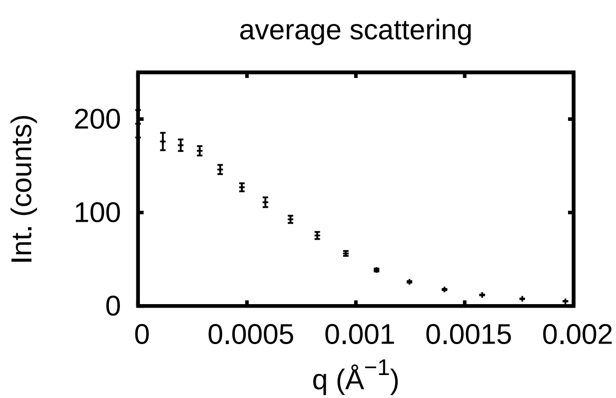
$$(L/a)^2 = (t/\tau_0)$$

we have for a=3.17Å, $\tau_0 \simeq 20\mu$s (rough! depends on definition of L)

/1/*Hohenberg and Halperin, Rev. Mod. Phys. 48-3 (1977)*

Speckles





For a quenched sample, speckles corresponding to the Fourier transform of the static domain configuration are observed. Here, they are observed in the enlarged Bragg peak. The HWHM of the (average) peak is: $\Delta q \simeq 0.0007$Å^{-1}, i. e. $L \simeq 1400$Å

For T close to T$_c$, the equilibrium fluctuations of length ξ have a fluctuation time $\tau \propto \xi^z$. This fluctuation time diverges in the vicinity of T_c. The observation of this “critical slowing down" by means of speckle dynamics is the aim of the experiment.

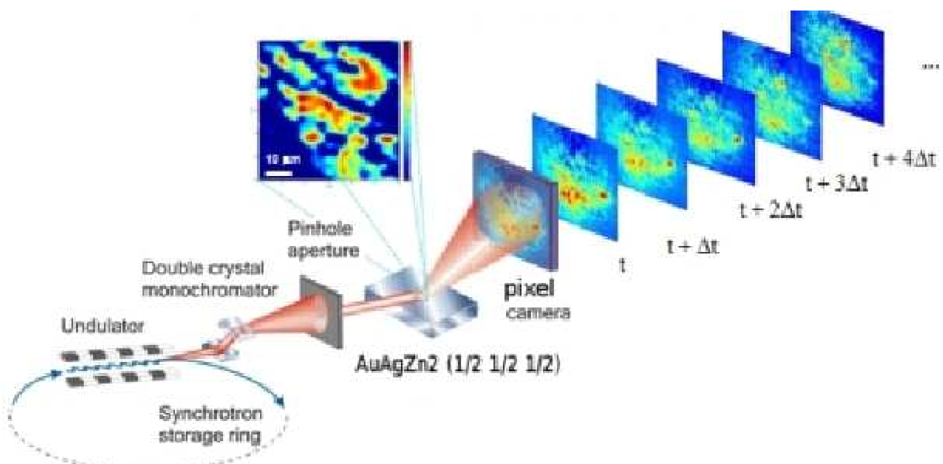
Speckle dynamics (XPCS)

The time correlations are obtained from:

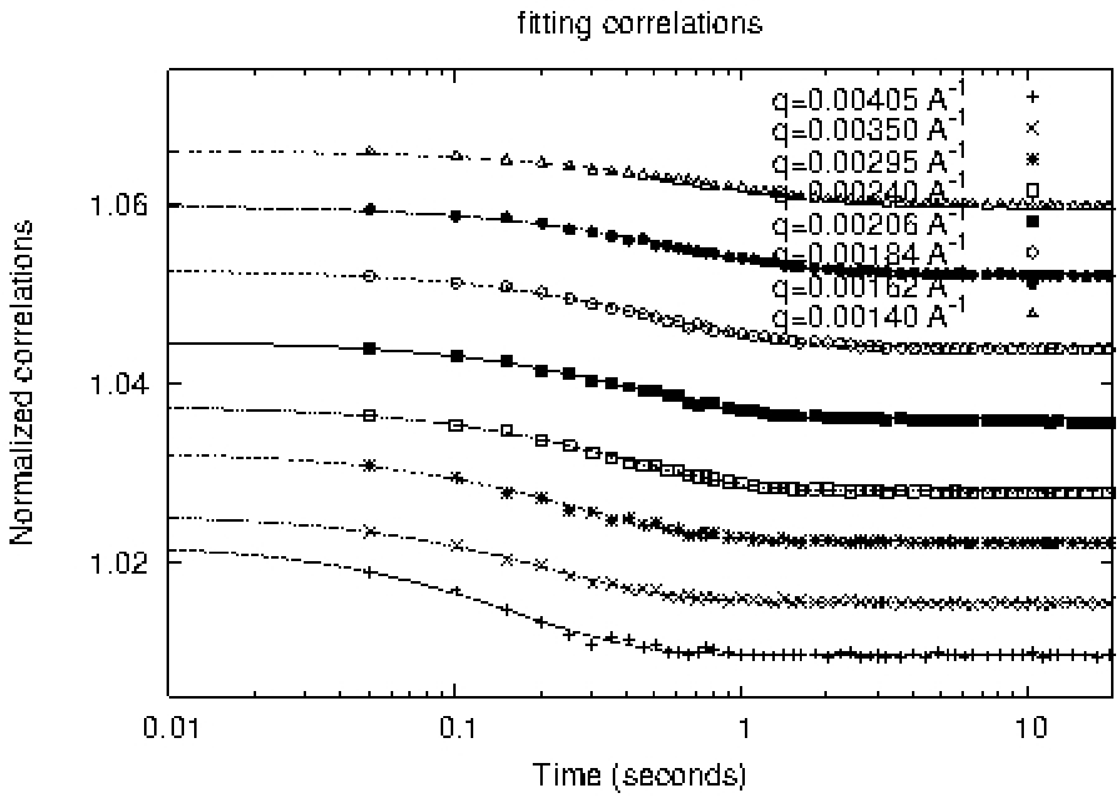
$$< I(\vec{q}, t+t')I(\vec{q}, t') >_{t'}$$

For each pixel of wavevector $\vec{q}$. Not enough statistics! One carries out also an average over a suitable $\vec{q}$ domain D_q:

$$\gamma(t,q) =< I(q,t+t)I(q,t) >_q \,/ < I(q,t) >_q^2$$



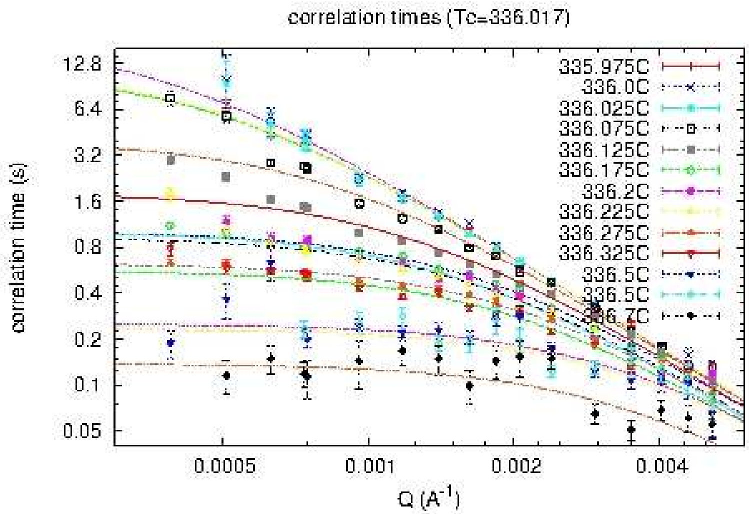
Example of correlations

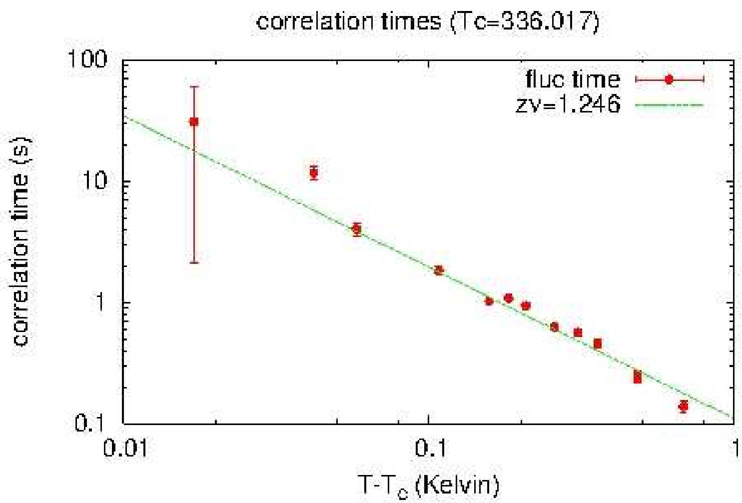


Measurements at $T_c + 0.09K$. Fit with the equation:

$$\gamma(t, q, T) = 1 + b0 + c0 \times exp(-t/\tau(q, T))$$

The dynamic scaling laws





A plot (top) of the various results $\tau(q,T)$. Writing:

$$\tau(q,T) = \tau_0(T)/(1 + (\xi' q^2))$$

a temperature variation of τ is obtained $\tau_0(T)$ vs $T - T_c$ is plotted (bottom) in a log scale. The scaling law:

$$\tau(T) \propto (T - T_c)^{-z\nu}$$

gives $z\nu = 1.246$, i. e. $z \simeq 1.97(5)$
In agreement with theoretical models ($z \simeq 2.02$), with a poor precision.

Improvements?

Beatrice Ruta will explain improvements!

The needs of coherence (β) are different here and in Direct imaging (Federico and Julio).