6

Conclusions

We have shown that one can produce a beam of coherent X-rays by *spatially filtering* an incoherent beam of *monochromatic* X-rays to dimensions smaller than its transverse coherence lengths using small pinholes with diameters of a few μm. A second generation synchrotron radiation source like beamline X-25 at NSLS provides a sufficient coherent flux to perform *coherent* X-rays scattering experiments.

When a coherent beam is scattered from a disordered sample, a graininess in the structure factor is observed, called speckle. The speckle pattern is sensitive to the exact arrangement of the atoms in the illuminated volume. If the state of disorder of the sample changes with time, the speckle pattern will fluctuate in time. An analysis of the time correlations of the scattered light at a given wavevector allows a direct measurement of the dynamics of the system, in and out of thermodynamic equilibrium.

This thesis is the first study of the ordering kinetics of first order phase transitions in a classical binary alloy with a *non-conserved order* parameter, using *coherent hard X-rays*. One of the important contributions of this thesis has been to develop techniques to measure the speckle patterns produced by coherent X-ray scattered from a binary alloy with a two-dimensional CCD detector, as well as techniques to analyze these patterns.

In Chapter 3, the development of techniques based on statistical estimators like the mean, variance and spatial correlation function has been essential in our understanding of the noise, quantum efficiency, and spatial resolution of the CCD. The use of spatial correlation functions to measure the resolution of the CCD is an original contribution [37]. The spatial autocorrelation function of the signal generated by a spatially uniform incident beam is broadened by the spread of the detected electrons to several nearest neighbor pixels. By assuming an identical response for each detected photon, the resolution function can be extracted from the measured correlation function. The spatial extent of the resolution function reduces the spatial or temporal variance of the signal when compared to the variance due to Poisson noise. This effect must be accounted for in the calculation of the quantum efficiency. These techniques can be applied to any typical one and two-dimensional position-sensitive detectors, and can also characterize non-linear detectors.

Section A.2 also contains central and original derivations, which have been used throughout the thesis to calculate error bars of various functions of the estimated average, $<V>$, and variance, S_V^2, of the signal V. We have shown that by combining standard error analysis with the variances and covariance of $<V>$ and S_V^2, one can approximate the standard deviation of several functions of the estimated mean and variance $f(<V>, S_V^2)$. We derived, for example, the error on the contrast $\sqrt{S_V^2/<V>^2}$ and on the ratio $S_V^2/<V>$. This ratio of variance over mean is an important quantity which can be used to calibrate a detector, or to determine whether or not the observed time fluctuations of the signal are above the fluctuations expected from counting statistics.

The development of the detector characterization techniques was based on spatial correlation functions introduced to measure the speckle size of Cu_3Au speckle patterns. Using spatial correlation functions, it was shown that this detector has a sufficient spatial resolution to measure speckle patterns of Cu_3Au (100). The speckle pattern properties were studied by changing the horizontal transverse coherence length, and the illuminated sample area. The speckle size was measured by the width at small displacements of the spatial crosscorrelation function of two subsequent scans of the CCD. The speckle width is reduced when the illuminated sample area is increased, until the speckle size is comparable to the resolution of the CCD. Then the widths become resolution limited. Increasing the horizontal coherence length reduced the speckle size horizontally. Because the CCD resolution is anisotropic and broader than one pixel, and because the beam is partially coherent, the effect of different optical conditions requires subtle analysis. A theoretical model based on the autocorrelation function of the Fraunhofer diffraction pattern of the pinhole convolved with the detector resolution is in good agreement with the observations. Some difference is seen between the model and the data for the smallest pinhole $D = 3.5$ μm. Perhaps this indicates that the illuminated sample area is larger than the pinhole area, since the sample is placed at the limit of the near-field diffraction of the pinhole. Note that these effects should be easier to resolve if the angular resolution of the detector is improved, whether by choosing a detector with finer spatial resolution, or placing the detector further away.

The contrast of the speckle pattern was also measured by subtracting the expected Poisson noise contribution from the spatial fluctuations. The contrast is reduced as expected when the input beam diameter increases, and when the upstream source size is increased. The measured contrast is smaller than expected. We discussed some of the possible causes for this difference earlier.

These measurements of the contrast and speckle size are major contributions of this thesis. It is the first *detailed quantitative* analysis of X-ray speckle. The measurements on static speckle from Cu_3Au agree well with the well known properties of speckle. Because the CCD detector can measure thousands of independent speckles simultaneously, the contrast and autocorrelation functions can be measured quite accurately. The calculation algorithm are also simple to implement. This work has some applications in microscopy, like atomic force and electron microscopy. For example, it is straightforward to measure the average domain size in an image with the autocorrelation function.

We measured the time fluctuations of the structure factor for the (100) superlattice reflection of Cu_3Au after a quench from the disordered phase to the ordered phase. Soon after the quench, speckles appear at fixed positions in reciprocal space. Their intensities grow like the average scattering from antiphase domains. The overall sharpening of the structure factor and the increase in peak intensity can be measured by least-squares fits of the whole speckle pattern. The widths and peak intensity of the Gaussian fits follow power laws, as expected from the coarsening of domains.

The approach we took to study the time fluctuations is to look at the time fluctuations with respect to a time-dependent growing average, provided by two-dimensional fits. By studying the statistics of the relative fluctuations with respect to the fit, one can test whether these fluctuations are above counting statistics or not. The interested reader will find some of our experience gained in analyzing the time fluctuations in equilibrium and out of equilibrium compiled in section A.4. For the X-25 data, the time fluctuations on a time scale of one half hour are within the expected fluctuations due to Poisson noise of the detected photons and incident intensity fluctuations, thus the speckles are constant over this time scale within fluctuations due to counting statistics. Intrinsic fluctuations with larger time scales due to the coarsening dynamics cannot easily be separated from fluctuations caused by beamline instabilities at present.

The amplitude of the time fluctuations of the data is small, and consistent with theory. As opposed to an IFS experiment performed in thermal equilibrium (see Fig. 2.13), the relative amplitude of these fluctuations is much smaller than 100 % and the fluctuation times are long. This makes quantitative analysis particularly difficult because of the limited count rate and the beamline instabilities. If the amplitude of these fluctuations had been larger, the time fluctuations would have been more easily measured. We believe that the small size of the fluctuations is an important property of the speckle patterns generated by the non-equilibrium growth which produced the domain structure.

This observation is qualitatively consistent with a numerical simulation of model A. After the quench, speckles appear at fixed $\vec{q}$, and their dynamics are slow. This observation is consistent with the simple picture that the speckle pattern is dominated by the scattering from large domains which slowly grow at the expense of small domains. This dynamics makes the small domains disappear, leaving the macroscopic domain arrangement over the sample size changing slowly with time (see Fig. 5.19). Fourier modes representing the domain structure changes only slowly because the macroscopic domain structure correlates with itself for long times.

The last conclusion of this thesis is that individual speckles remain *fixed* in reciprocal space after the quench. This observation violates simple minded scaling in the experimental data and the numerical simulation. Although the speckle pattern does not scale, the overall scattering sharpens as expected for scaling. This persistence of speckle seems to be a general property of the structure factor of non-equilibrium growth measured with coherent light, for both conserved and non-conserved order parameters. These experiments started out as a way to test some of the ideas of Grant and Roland [36]. There is good reason to believe that these types of measurements will lead to a deeper understanding of this type of phenomena, but much work still need to be done. This XIFS work has also generated a lot of interest in the visible IFS community, and many experiments are underway to study non-equilibrium phenomena.

To measure the time fluctuations in more detail, longer experiments with higher incident coherent flux are planned in the future. A factor of 100-1000 in intensity is accessible by using the new undulator facilities such as the ESRF or the APS. This increase in flux would allow to reduce the error bars in Fig. 5.17 by a factor of more than ten, and enables smaller changes of the structure factor to be measured. As demonstrated recently [19], these beamlines are also more stable in terms of beam intensity and position. Ideally, one should also look for systems with faster intrinsic dynamics than Cu_3Au, and samples which scatter isotropically[1].

Improvements in detecting scheme are also achievable. For example, very low noise two dimensional gas detectors with nearly unit quantum efficency (a factor 4 larger than the detector used in this experiment) are under development at the ESRF, and should be operational within the next year. These detectors allow measurements

[1] For example, although an Fe_3Al sample scatters less that a Cu_3Au one due to smaller scattering contrast, it scatters isotropically in $2\Theta_{||}$ and $2\Theta_{\perp}$, making it possible to ensemble average correlation functions with the same $|\vec{q}|$. This should improve the statistics of the correlation function and allow for the measurement of the wavevector dependence of the correlation function [89].

of the time arrival of each photon, and have very low dark noise. Similarly, CCD performance can be improved by using low noise detectors, with a CCD chip having a larger depletion layer. A larger depletion layer is found in EEV chips, for example [90], and this increases the quantum efficiency for hard X-rays. At 7 keV, the EEV chip has a quantum efficiency of 50 %, which is a factor two better than the chip used here. Another approach with CCDs is to couple the array optically with an X-ray sensitive phosphor which greatly enhances the quantum efficency but reduces the CCD resolution. By placing such a detector a few meters away, the resolution should be sufficient to resolve speckle patterns. A completely different approach with CCDs should be used when the count rates are low. For low count rates, it is possible to detect single photons events. This removes most of the electronic noise in the data, and in principle allow subpixel spatial resolution!

These measurements are some of the first results using this newly developed scattering technique. As a feasibility experiment, our measurements on Cu_3Au were a success! We demonstrated that the experiment can be performed, and pointed out some of the major problems encountered on second generation synchrotron sources. These results have important applications in the design of the next generation of beamlines optimized to produce coherent X-rays. These measurements are also exciting because they give new insights on the dynamics of phase transitions not available before. Beside our IFS results, the simple example of coherent X-ray scattering from an isolated edge dislocation was given in Chapter 2. This proposed experiment is already feasible with existing sources.

Coherent X-ray scattering will be a powerful technique at the newly commissioned third generation synchrotron sources, and many group are developing strong scientific programs in this field. Because the coherence lengths of synchrotron radiation are on the order of the microstructure of many materials, coherent X-rays will be a tool of choice to investigate disorder in these materials.

Appendices

A.1 Some useful probability distributions

A Poisson distribution is discrete. The probability of measuring n events is

$$P(n) = \frac{\mu^n e^{-\mu}}{n!}. \tag{A.1}$$

The mean and the variance are μ. Higher central moments are $\overline{(x-\mu)^3} = \mu$, and $\overline{(x-\mu)^4} = 3\mu^2 + \mu$.

A Gaussian distribution is continuous, with probability density

$$\rho(x) = \frac{1}{\sqrt{2\pi}\sigma} \exp[\frac{(x-\mu)^2}{2\sigma^2}]. \tag{A.2}$$

The mean is μ and the variance is σ^2. Higher moments are $\overline{(x-\mu)^3} = 0$ and $\overline{(x-\mu)^4} = 3\sigma^4$.

A chi-square random variable is given by

$$\chi^2 = \sum_{i=1}^{M} (y_i - \overline{y_i})^2. \tag{A.3}$$

It is the sum of the squares of M Gaussian random variable y_i, with average $\overline{y_i}$ and standard deviation σ. The probability density of this random variable is

$$\rho(\chi^2) = (\sqrt{2}\sigma)^{-M} (\chi^2)^{M/2-1} \exp(-\frac{\chi^2}{2\sigma^2})/\Gamma(M/2). \tag{A.4}$$

The average is $M\sigma^2$ and the variance $2M\sigma^4$, with a contrast $\sqrt{2/M}$.

The chi-square distribution is the probability distribution followed by speckle statistics for M=2. For speckle, the scattered intensity $I = \mathrm{Re}\{E\}^2 + \mathrm{Im}\{E\}^2$, i.e. the sum of the squares of the real and imaginary part of the Gaussian scattered electric field E. Here the electric field is linearly polarized. For $M = 2$, Eq. A.4 is an exponential with average $\overline{I} = 2\sigma^2$ and standard deviation $\sigma_I = 2\sigma^2$. The fluctuations of the scattered intensity are as large as the mean!

When a detector averages several speckles because its area integrates over many speckles, Eq. A.4 is also the distribution which *approximately* describes the scattered intensity I. The probability density of the scattered intensity is

$$p(I) = (\frac{N_s}{\overline{I}})^{N_s} I^{N_s-1} \exp(-\frac{N_s I}{\overline{I}})/\Gamma(N_s), \tag{A.5}$$

where N_s is the number of independent speckle in the detector area, or the ratio of the detector area over the speckle area introduced in Eq. 5.3, and $\overline{I}$ is the average intensity. This is known as the gamma probability density [39]. For $N_s = 1$, it reduces to the exponential distribution we derived earlier in Eq. 2.34. Its average is $\overline{I}$ and its variance is $\frac{\overline{I}^2}{N_s}$, so that the contrast $\frac{\sigma}{\overline{I}} = \frac{1}{\sqrt{N_s}}$.

A.2 Error analysis

One often wants to evaluate the uncertainty in the estimated average $<x>$ and variance S^2 of a random variable X, with theoretical mean μ and variance σ^2. Assuming that the probability density $\rho(x)$ of the random variable X is stationary, the true mean and variance of $\rho(x)$ are defined by

$$\overline{x} = \int_{-\infty}^{\infty} x\rho(x)dx = \mu, \text{ and} \tag{A.6}$$

$$\overline{(x-\mu)^2} = \int_{-\infty}^{\infty} (x-\mu)^2\rho(x)dx = \sigma^2, \tag{A.7}$$

with $\int_{-\infty}^{\infty}\rho(x)dx = 1$. The discussion below is limited to distributions with finite μ and σ^2. The statistical estimators of the mean $<x>$ and variance S^2 are defined by

$$<x> = \frac{1}{N}\sum_{i=1}^{N} x_i, \text{ and} \tag{A.8}$$

$$S^2 = \frac{1}{N-1}\sum_{i=1}^{N}(x_i - <x>)^2, \tag{A.9}$$

where x_i is a measured outcome of the random variable X, chosen from the same probability distribution $\rho(x)$. The estimators $<x>$ and S^2 are unbiased estimators of the mean and variance and on average are equal to μ and σ^2. The uncertainties in these statistical estimators depend on the number of random events observed, N. For small N, the fluctuations in the measured or estimated value are comparable to the expected value. Therefore, its dependence on N must be well characterized.

The uncertainty on the mean is well known and goes as $\sqrt{\sigma^2/N}$ [64]. For a Gaussian distribution, the uncertainty on the variance is $\sqrt{2/(N-1)}\sigma^2$ [64]. In section A.2.1, we derive a general relation for the uncertainty of the estimated variance, and apply it to the Poisson distribution. In error propagation analysis, one may want to evaluate the uncertainty in a function of the mean and variance $f(<x>, S^2)$ as required in section 3.0.3. To evaluate the error in f, one uses the uncertainty in $<x>$ and S^2, and needs to know the covariance between $<x>$ and S^2. A general expression for this covariance is derived in section A.2.2, and a derivation for Eq. (3.7) is shown in section A.2.3 and A.2.4.

A.2.1 Uncertainty in the measured mean and variance

It is important to note that $<x>$ and S^2 are themselves random variables since they are functions of several random variables. A useful estimator of the uncertainty of these new random variables is their standard deviations $\sigma_{<x>}$ and σ_{S^2} defined in Ref. [64] by

$$\sigma^2_{<x>} = \overline{<x>^2} - \overline{<x>}^2, \text{ and} \tag{A.10}$$

$$\sigma^2_{S^2} = \overline{S^4} - \overline{S^2}^2, \tag{A.11}$$

where the horizontal line refers to an average over the joint probability distribution of the random variable $<x>$, a function of N random variables x_i, i.e. $\overline{<x>} \equiv \int <x>(x_1, ..., x_N)\rho(x_1, ..., x_N)dx_1...dx_N$. $\sigma_{<x>}$ and σ_{S^2} are the standard deviations of the probability densities of $<x>$ and S^2. The N dependence in Eq. (A.10-A.11) is not explicitly written in order to keep the notation simple. From Eq. (A.8), recalling that $\overline{x_i} = \mu$, it is easy to show that $\overline{<x>} = \mu$. In order to simplify the algebra below, we introduce a new variable $y_i = x_i - \mu$, so that $\overline{y_i} = 0$ and $\overline{y_i^2} = \sigma^2$. One can now rewrite Eq. (A.10) , using Eq. (A.8) and square it to get

$$\sigma^2_{<x>} = \frac{1}{N^2}\sum_{i,j=1}^{N} \overline{y_i y_j}. \tag{A.12}$$

Assuming that each y_i is independent, i.e.

$$\overline{y_i y_j} = \delta_{ij}\sigma^2, \tag{A.13}$$

where δ_{ij} is a Kronecker delta, and replacing Eq. (A.13) in Eq. (A.12), one gets

$$\sigma^2_{<x>} = \frac{\sigma^2}{N}, \tag{A.14}$$

which is a well known result valid for any N, and any probability distribution with finite mean and variance, given that no correlation exists between the x_i's. This result states that the probability density of $<x>$, $\rho(<x>)$, is narrower than $\rho(x)$ by a factor $\sqrt{\frac{1}{N}}$.

In Eq. (A.11), the $\overline{S^2}$ term can be evaluated by substituting x_i by y_i and replacing Eq. (A.8) into Eq. (A.9). Then, one finds

$$S^2 = \frac{1}{N-1}\left(\sum_{i=1}^{N} y_i^2 - \frac{1}{N}\sum_{i,j=1}^{N} y_i y_j\right). \tag{A.15}$$

Taking averages on both sides of Eq. (A.15), and evaluating the sums with Eq. (A.13), one finds $\overline{S^2} = \sigma^2$, as stated before. The $N-1$ denominator comes from the fact that there is one less degree of freedom used to evaluate the mean. Taking the square of Eq. (A.15), and taking averages on the right and left hand side of the equation, one finds

$$\overline{S^4} = \frac{1}{(N-1)^2}\left(\sum_{i,j=1}^{N} \overline{y_i^2 y_j^2} - \frac{2}{N}\sum_{i,j,k=1}^{N} \overline{y_i^2 y_j y_k} + \frac{1}{N^2}\sum_{i,j,k,l=1}^{N} \overline{y_i y_j y_k y_l}\right). \tag{A.16}$$

Evaluating the averages recalling that $\overline{y_i} = 0$ and $\overline{y_i y_j} = \delta_{ij}\sigma^2$, one finds

$$\overline{y_i^2 y_j^2} = \begin{cases} \overline{y^4}, & \text{for } i = j, \quad N \text{ terms}, \\ \sigma^4, & \text{for } i \neq j, \quad N(N-1) \text{ terms}, \end{cases} \tag{A.17}$$

$$\overline{y_i^2 y_j y_k} = \begin{cases} \overline{y^4}, & \text{for } i = j = k, \quad N \text{ terms}, \\ \sigma^4, & \text{for } i \neq j = k, \quad N(N-1) \text{ terms}, \\ 0, & \text{otherwise}, \end{cases} \tag{A.18}$$

$$\overline{y_i y_j y_k y_l} = \begin{cases} \overline{y^4}, & \text{for } i = j = k = l, \quad N \text{ terms}, \\ \sigma^4, & \text{for } i = j \neq k = l, \quad 3N(N-1) \text{ terms}, \\ 0, & \text{otherwise}. \end{cases} \tag{A.19}$$

Here, the nth order central moment is defined by $\overline{y^n} \equiv \int y^n \rho(y) dy$. Using Eq. (A.17-A.19) to evaluate the sums in Eq. (A.16), and replacing Eq. (A.16) into Eq. (A.11), one finds

$$\sigma_{S^2}^2 = \frac{\overline{y^4}(N-1) + \sigma^4(3-N)}{N(N-1)}. \tag{A.20}$$

This result depends on the fourth order central moment of the distribution, $\overline{y^4}$, and the standard deviation, σ. For a Gaussian distribution, (see section A.1) $\overline{y^4} = 3\sigma^4$ and Eq. (A.20) gives as expected $\sigma_{S^2}^2 = \frac{2}{N-1}\sigma^4$. For a Poisson distribution on the other hand, $\overline{y^4} = 3\mu^2 + \mu$ which yields

$$\sigma_{S^2}^2 = \frac{2}{N-1}\mu^2 + \frac{\mu}{N}. \tag{A.21}$$

Note that for $\mu < 1$, the second term is an important correction and a Gaussian approximation would underestimate the error on the variance. For large means, Eq. (A.21) becomes identical to the error for a Gaussian with variance equal to the mean, as expected since the Poisson distribution crosses over to such a Gaussian for large means. Finally, note that since in general $\rho(S^2)$ is not Gaussian, one cannot easily quantify the confidence limits of S^2, but can nevertheless calculate its variance.

A.2.2 Correlation of the sample mean and variance

One might think that there is no correlation between the estimator of the mean $<x>$ and the estimator of the variance S^2. The independence of the estimated mean and variance is discussed in several statistical books for Gaussian distributions [73, 64]. The measure of correlation between the measured mean and variance, the covariance, is defined as

$$\begin{aligned} \text{cov}(<x>, S^2) &= \overline{(<x> - \mu)(S^2 - \sigma^2)} = \overline{<y> S^2} \\ &= \frac{1}{N(N-1)} \left(\sum_{i,j=1}^{N} \overline{y_i y_j^2} - \frac{1}{N} \sum_{i,j,k=1}^{N} \overline{y_i y_j y_k} \right), \end{aligned} \tag{A.22}$$

since $\overline{<y>\sigma^2} = 0$. Evaluating the averages as in Eq. (A.17-A.19), one finds

$$\overline{y_i y_j y_k} = \begin{cases} \overline{y^3}, & \text{for } i = j = k, \quad N \text{ terms}, \\ 0, & \text{otherwise}. \end{cases} \tag{A.23}$$

The same results holds for $\overline{y_i y_j^2}$. Replacing those in Eq. (A.22) and simplifying, one gets

$$\mathrm{cov}(<x>, S^2) = \frac{\overline{y^3}}{N}, \tag{A.24}$$

where $\overline{y^3}$ is the 3rd order central moment of the distribution. Thus, the covariance depends on N, and on how symmetric with respect to the mean is the probability distributed. The estimated mean and variance are independent only as N goes to infinity, or if $\overline{y^3} = 0$. For a Gaussian distribution, the covariance is zero because odd powers of the central moments are zero. For a Poisson distribution, $\overline{y^3} = \mu$ (see Appendix A.1) and

$$\mathrm{cov}(<x>, S^2) = \frac{\mu}{N}. \tag{A.25}$$

A.2.3 Evaluation of the error on a function of S^2 and $<x>$

From standard error propagation analysis [73], one can readily calculate the uncertainty of a function $f(<x>, S^2) = \frac{S^2}{<x>}$. It is given by

$$\frac{\sigma_f^2}{f^2} \approx \frac{\sigma_{S^2}^2}{\overline{S^2}^2} + \frac{\sigma_{<x>}^2}{\overline{<x>}^2} - \frac{2\mathrm{cov}(S^2, <x>)}{\overline{<x>}\,\overline{S^2}}, \tag{A.26}$$

where the terms on the right hand side have been calculated above. The approximate sign comes from the fact that σ_f is derived from a first order Taylor expansion of f around its average. Replacing the uncertainties and covariance derived previously for a Poisson distributed random variable X in Eq. (A.14) ,(A.21), (A.25), one finds

$$\frac{\sigma_f^2}{f^2} \approx \frac{2}{N-1}. \tag{A.27}$$

The result is independent of the mean μ. By averaging N independent measurements, one can measure f accurately. On the other hand, if the signal is Gaussian distributed, the covariance term in Eq. (A.26) would be zero, and one gets $\frac{\sigma_f^2}{f^2} \approx \frac{2}{N-1} + \frac{\sigma^2}{\mu^2 N}$. Depending on the mean and variance of the distribution, the second term may be important.

A.2.4 Including electronic noise for an X-ray detector

For an X-ray detector, Eq. (A.27) is not sufficient to estimate the errors on k, defined in Eq. (3.5), because for weak signals, the electronic noise will contribute significantly to σ_k. V_e, the dark electronic noise, increases the relative error on the time averaged mean $<V>_t$ and variance $S_{t,V}^2$ when the contribution from the detected photons approaches zero. To find σ_k, the standard deviation of k, we can simply replace in Eq. (A.26) $<x>$ by $<V>_t$, and S^2 by $S_{t,V}^2 - \sigma_{Ve}^2$, where σ_{Ve}^2 is the theoretical variance of the electronic noise probability distribution. Then the first term of Eq. (A.26) can be rewritten as

$$\frac{\sigma^2_{S^2_{t,V} - \sigma^2_{Ve}}}{(\overline{S^2_{t,V}} - \sigma^2_{V_e})^2} = \frac{\sigma^2_{k^2 S^2_{n_d}} + \sigma^2_{S^2_{Ve}}}{(\overline{S^2_{t,V}} - \sigma^2_{V_e})^2}$$

$$= \frac{2}{N-1} + \frac{1}{Nn_d} + \frac{2\sigma_{Ve}^4}{(N-1)(\overline{S_{t,V}^2} - \sigma_{V_e}^2)^2}. \qquad \text{(A.28)}$$

To obtain this relationship, we used the equation $\sigma^2_{S^2_{t,V}-\sigma^2_{Ve}} = \sigma^2_{S^2_{t,V}}$ and Eq. (3.3), and assuming that V_e follows Gaussian statistics. The first two terms are the detected photon contributions which are included in Eq. (A.27), and the last term is due to an error on the evaluation of the dark variance. Next, we can evaluate the second term of Eq. (A.26) using Eq. (3.3),

$$\begin{aligned}\frac{\sigma^2_{<V>_t}}{\overline{<V>_t}^2} &= \frac{k^2\sigma^2_{n_d} + \sigma^2_{V_e}}{N\overline{<V>_t}^2} + \frac{\sigma^2_{V_e}}{N_d\overline{<V>_t}^2} \\ &= \frac{1}{Nn_d} + \frac{\sigma^2_{V_e}}{\overline{<V>_t}^2}\left(\frac{1}{N} + \frac{1}{N_d}\right), \qquad \text{(A.29)}\end{aligned}$$

where n_d and $\overline{<V>_t}$ are the hypothesized means of the number of detected photons and of the detector signal, while $\sigma^2_{n_d}$ and $\sigma^2_{V_e}$ are the theoretical variances of n_d and of the dark electronic noise. The first term in this equation comes from the statistics of the detected photons and is already included in Eq. (A.27). The second term explains the increased uncertainty in k for small signals due to the electronic noise. The third term is due to the statistical error in the measurement of the dark pattern subtracted from V, where N_d scans are averaged without X-rays. To minimize the error, one should choose $N_d = N$. Finally, the last term in Eq. (A.26) is unchanged from the Poisson case because the dark signal and the detected photon signal are not correlated. Collecting terms, we find

$$\frac{\sigma_k^2}{k^2} \approx \frac{2}{N-1}\left(1 + \frac{\sigma_{Ve}^4}{(\overline{S_{t,V}^2} - \sigma_{V_e}^2)^2}\right) + \frac{\sigma^2_{V_e}}{\overline{<V>_t}^2}\left(\frac{1}{N} + \frac{1}{N_d}\right). \qquad \text{(A.30)}$$

It is therefore easier to measure k when $<V>_t^2$ and $S^2_{t,V}$ are much larger than $\sigma^2_{V_e}$. To evaluate σ_k for a given experiment, one would replace $\sigma^2_{V_e}$, $\overline{S^2_{t,V}}$ and $\overline{<V>_t}$ by the experimentally measured values S^2_{t,V_e}, $S^2_{t,V}$ and $<V>_t$.

Note that the error analysis approach developed above can be extended to other problems, such as the evaluation of the error on the measured contrast in optics if the signal is not Gaussian distributed. It is also useful to estimate the errors in our coherent X-ray experiments.

For example, the error on the measured contrast for the time-averaged scattered intensity in section 2.6.2 is derived with the same technique. In section 2.6.2, the contrast is defined by

$$Ct = \frac{\sqrt{S^2}}{<x>}. \qquad \text{(A.31)}$$

Neglecting the term for the correlation between the estimated mean and variance, it is easy to show that the error on the contrast

$$\frac{\sigma_{Ct}}{\overline{Ct}} \approx \sqrt{\frac{\sigma^2_{S^2}}{4\overline{S^2}^2} + \frac{\sigma^2_{<x>}}{\overline{<x>}^2}} \approx \sqrt{\frac{1}{2(N-1)} + \frac{Ct^2}{N}}. \qquad \text{(A.32)}$$

Here, the error on the estimated variance σ_{S^2} was approximated by the error for Gaussian statistics.

A.3 Derivation of the autocorrelation function for a Gaussian

If one assumes that the structure factor is a smoothly varying function of $\vec{q}$, one can understand quantitatively the $\vec{q}$ dependence of the crosscorrelation function (see Eq. 5.5) which is caused by the presence of antiphase domains.

Let us study the autocorrelation function by choosing $dt = 0$ in Eq. 5.5. For the two dimensional Gaussian function defined in Eq. 5.11, one can show that its spatial autocorrelation function calculated by a two dimensional integral over a rectangle in reciprocal space with edges $L_\perp \times L_{||}$ is

$$<v^*(\vec{q},t)v^*(\vec{q}+\vec{dq},t)>_{\vec{q}} = \frac{\pi\sigma_\perp\sigma_{||}I_{max}^2}{4L_\perp L_{||}}\exp(\frac{-dq_\perp^2}{4\sigma_\perp^2})\exp(\frac{-dq_{||}^2}{4\sigma_{||}^2})\left[\mathrm{erf}(\frac{L_{||}+dq_{||}}{2\sigma_{||}})+\mathrm{erf}(\frac{L_{||}-dq_{||}}{2\sigma_{||}})\right] \times \left[\mathrm{erf}(\frac{L_\perp+dq_\perp}{2\sigma_\perp})+\mathrm{erf}(\frac{L_\perp-dq_\perp}{2\sigma_\perp})\right], \quad (A.33)$$

where I_{max} is the peak intensity, and $\sigma_\perp$ is the Gaussian width along $2\theta_\perp$. The error functions are present because of the finite size of $L_\perp$ and $L_{||}$. For a small displacement with $|dq_\perp| << L_\perp$ and $|dq_{||}| << L_{||}$, the autocorrelation in Eq. A.33 is also approximately a two dimensional Gaussian with width along $2\theta_\perp$ equal to $\sqrt{2}\sigma_\perp$.

In Fig. 5.5–5.7, the autocorrelation is normalized by the square of the spatial average. For a two dimensional Gaussian, the spatial average of the linearized signal is

$$<v^*>_{\vec{q}} = \frac{2\pi\sigma_\perp\sigma_{||}I_{max}}{L_\perp L_{||}}\mathrm{erf}(\frac{L_\perp}{2\sqrt{2}\sigma_\perp})\mathrm{erf}(\frac{L_{||}}{2\sqrt{2}\sigma_{||}}). \quad (A.34)$$

Thus the normalized autocorrelation function for small displacement $|dq_\perp| << L_\perp$ and $|dq_{||}| << L_{||}$ is approximately

$$\frac{<v^*(\vec{q},t)v^*(\vec{q}+\vec{dq},t)>_{\vec{q}}}{<v^*>_{\vec{q}}^2} \propto \frac{L_\perp L_{||}}{16\pi\sigma_\perp\sigma_{||}}\exp(\frac{-dq_\perp^2}{4\sigma_\perp^2})\exp(\frac{-dq_{||}^2}{4\sigma_{||}^2}). \quad (A.35)$$

Note that the maximum of the normalized autocorrelation is inversely proportional to the product of the widths $\sigma_\perp\sigma_{||}$.

A.4 Tools for time fluctuations analysis

Here are some of the tools that might be useful for simple analysis of time fluctuations for XIFS in systems in thermal equilibrium and out of equilibrium. In equilibrium, to make sure that the time fluctuations are larger than the fluctuations due to counting statistics and electronic noise, one can measure the fluctuations of the structure factor with respect to its long time average, and compare these fluctuations to those caused by Poisson noise. We define the ratio of these two quantities as

$$R(\vec{q},t) = \frac{S^2_{t,S}(\vec{q})}{<S(\vec{q},t)>_t}, \tag{A.36}$$

where $S^2_{t,S}$ is the variance and $<S(\vec{q},t)>_t$ the time average of the structure factor $S(\vec{q},t)$ measured from a sequence of N measurements. In deriving Eq. A.36, the detector is assumed to have unit quantum efficiency and a spatial resolution much smaller that the speckle size. With such a detector, the expected variance from Poisson noise is simply equal to the time average $<S(\vec{q},t)>_t$. Note that we have already derived this equation earlier since this is simply the equation for k in Eq. 3.5 for a detector without electronic noise! If the fluctuations are only caused by Poisson noise, $R = 1$, and its error bars are given by Eq. A.27. A ratio greater than unity implies that the fluctuations are significantly larger than fluctuations due to counting statistics.

In an intensity fluctuation spectroscopy experiment, one wants to know what is the standard deviation of the structure factor corrected for trivial fluctuations due to counting statistics[1]. This standard deviation of $S(\vec{q},t)$, $\sigma_{t,S}$ is simply

$$\sigma_{t,S}(\vec{q}) = \sqrt{(R(\vec{q})-1)<S(\vec{q},t)>_t}. \tag{A.37}$$

Therefore, the relative fluctuation of the structure factor is simply the ratio of Eq. A.37 over the time average $<S(\vec{q},t)>_t$ equal to

$$\frac{\sigma_{t,S}(\vec{q})}{<S(\vec{q},t)>_t} = \sqrt{\frac{R(\vec{q})-1}{<S(\vec{q},t)>_t}}. \tag{A.38}$$

If the ratio $R(\vec{q}) = 1.1$ and the time average of the structure factor is 10 counts, then the relative rms fluctuation of the signal with respect to its time average is $\sqrt{0.1/10} = 10$ %.

Out of equilibrium, since the time average of $S(\vec{q},t)$ changes in time, Eq. A.36 must be modified to take this into account. We have found it useful to measure the fluctuations of the speckle pattern with respect to two dimensional least-squares fit of the whole speckle pattern. A natural variable to study fluctuations is

$$\eta(\vec{q},t) = \frac{S(\vec{q},t)}{S_f(\vec{q},t)} - 1. \tag{A.39}$$

Here η is the relative difference between the structure factor $S(\vec{q},t)$, and the fit $S_f(\vec{q},t)$. Assuming for simplicity that the detector is ideal, the expected standard deviation of η is

$$\sigma_\eta = \frac{\sqrt{S(\vec{q},t}}{S_f(\vec{q},t)}. \tag{A.40}$$

[1] An excellent introduction on the subject can be found in Chapter 9 of Goodman [39].

To calculate whether the fluctuations of $\eta(\vec{q},t)$ are above its expected error bars, we define the ratio

$$R_\eta(\vec{q}) = \frac{S^2_{t,\eta}(\vec{q})}{<\sigma^2_\eta(\vec{q},t)>_t}. \tag{A.41}$$

Here the variance of η, $S^2_{t,\eta}$, and the time average of the calculated variance σ^2_η, $<\sigma^2_\eta(\vec{q},t)>_t$, can be calculated from N subsequent measurements. The ratio $R_\eta(\vec{q},t)$ is a two dimensional array in our measurements.

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