Coherent X-ray scattering of charge order dynamics and phase separation in titanates

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Publication date
2017

Document Version
Other version

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Chapter 3

Coherent X-ray Scattering: Techniques, Principles and Instrumentation

In this chapter, we discuss various aspects of coherent x-ray scattering. We will briefly introduce x-ray correlation spectroscopy, and describe the ways one can generate the necessary coherent x-ray beams. Then we give a fairly detailed description of the statistics of the coherent x-ray scattering data, the speckle patterns, and use this to explain two methods by which dynamical information can be obtained by analyzing time series of speckle patterns. These speckle techniques also demand ultra-stable instrumentation, and how this has been realized during this research project is also recounted in detail.

3.1 X-ray correlation spectroscopy

When a coherent light beam scatters off a disordered medium, the resulting diffraction pattern observed on an area detector is found to consist of a complicated spatial arrangement of more or less equally sized blobs of intensity, called a speckle pattern. This speckle pattern is a reciprocal space representation of the disordered spatial structure of the scattering volume. However, the phase information of the scattered light is lost, making a direct inversion of speckle data to real space structure impossible.

One way to make use of the speckle pattern is to apply a so-called phase retrieval iteration algorithm [64–69], through which the lost phase information of the scattered wave can be recovered, in order to reconstruct the real space image of the spatial structure, in principle, down to the diffraction limit. This approach is especially interesting in the x-ray range, because theoretically it allows one to obtain microscopic images with Ångström spatial resolution.
In this thesis, apart from using regular, high-resolution scattering of synchrotron X-rays to study the charge order and phase separation in doped perovskite titanates, we also examine possible dynamics of the charge order in these systems using coherent X-ray scattering, via the analysis of speckles. In the ideal case, our approach would involve analysis of the correlations between a series of speckle patterns as a function of time delay between each image. When the spatial arrangement of the disordered medium changes, the speckle pattern also changes, and by studying these variations one can determine the dynamical properties of the system, such as correlation times.

Originally, this technique, called Photon Correlation Spectroscopy or Dynamical Light Scattering (DLS), was developed after the appearance of the first practical coherent light source, the laser. It has been very successful for the study of the dynamics of soft condensed matter systems such as colloids and gels, at timescales ranging from nanoseconds to hours and with correlation lengths of the order of the optical wavelengths [70–72].

When the first truly intense x-ray beams from third generation synchrotrons became available in the mid 1990’s, DLS has been ported to the x-ray range, where it became known as X-ray (photon) correlation spectroscopy (XPCS or XCS) [73–78]. Again, the advantage of X-rays lie in their Ångström wavelengths, which greatly improves the principle limits for the spatial resolution. Furthermore, hard X-rays have a greater penetration depth, which allows one to probe the bulk properties. XCS is now a proven technique for soft matter dynamical studies.

The open challenge is to see whether this technique can be used to study the dynamics of interesting electronic phases in hard condensed matter systems. Especially in the soft X-ray range, a number of strong resonance lines are found at which core levels interact with valence states. At these resonances, X-rays are sensitive to charge, orbital and spin signatures, opening up the possibility to probe the equilibrium dynamics of the corresponding ordered phases individually.

Pioneering hard condensed matter XCS work has been performed on the atomic diffusion in solid solutions [79], CDW/SDW domain dynamics [80] and spin dynamics [18, 81], and the Amsterdam group has been active in this field from the outset [18]. It should be pointed out that the technique is still very much under development. Since even storage ring based coherent X-ray sources are weak and scattering cross sections are relatively small, most experiments use CCD or other pixel detectors in order to intercept in parallel as many scattering angles as possible. Unfortunately, these detectors have quite long readout times. Because of this, at the moment XCS on hard condensed matter systems can only probe slow dynamics spanning time-scales from seconds to hours. This however is set to change with the advent of the X-ray Free Electron Lasers described later in this chapter.
3.2 Coherence

As is clear from the previous section, coherent X-rays are a key prerequisite of XCS. A real synchrotron beam is a superposition of a set of ideal plane waves with finite spectral bandwidth and divergence. As a result, the amplitudes at two different points in time or space are correlated only over finite times or lengths. The coherence of such a beam can be captured by two important length scales, the longitudinal (temporal) coherence length and the transverse (spatial) coherence length. Together they define the region over which the beam is coherent, and this region should be larger than the scattering volume of interest.

3.2.1 Longitudinal coherence length

The longitudinal coherence is a measure of how monochromatic a light source is. The longitudinal coherence length is defined as the shortest length over which two waves with a wavelength difference $\Delta \lambda$ run out of phase, and therefore measures the self-interference of the light beam, as can be seen from Fig. 3.1. The longitudinal coherence length is denoted by $\xi_l$, which is given by

$$\xi_l = \frac{1}{2} \frac{\lambda^2}{\Delta \lambda}.$$  \hspace{1cm} (3.1)
Within the longitudinal coherence length, the light source can be treated as coherent radiation. In practice, the longitudinal coherence is controlled by the bandwidth $\Delta \lambda / \lambda$ of a monochromator at a particular X-ray beamline. The longitudinal coherence length can be converted into a coherence time by dividing it by the speed of light.

### 3.2.2 Transverse coherence length

The light produced by a storage ring electron beam of finite spatial size has finite divergence, reducing the coherence in the transverse plane (which can be considered as a screen perpendicular to the propagation direction of light). The amplitudes of two spatial points in the transverse plane are correlated when the distance between the points is within the transverse coherence length $\xi_t$. This can be quantified by considering a light source with diameter $D$. The waves emitted from the middle and the edge of the light source will interfere constructively with each other at a distance $R$ in the far field when their separation on the screen is less than [83]

$$\xi_t = \frac{\lambda}{2D}.$$  \hspace{1cm} (3.2)

Any incoherent source, such as a synchrotron undulator beam or for that matter a candle flame, can be made transversely coherent by passing the beam through two aligned apertures, a technique known as spatial filtering [84–86]. The filtering condition is given by,

$$D_1D_2 \simeq \lambda L,$$  \hspace{1cm} (3.3)

where $D_1$ and $D_2$ are the diameter of the two apertures, $\lambda$ is the wavelength of the light, and $L$ is the distance between the two apertures. (See Fig. 3.2.)

We had to employ spatial filtering in the soft X-ray experiments ($\lambda \sim 1$ nm). In these experiments $D_2$ was typically chosen to be 5 or 10 $\mu m$, which is one to two orders of magnitude larger than the correlation lengths that we are looking for. In order to avoid broadening of the resulting beam by diffraction, this aperture ($D_2$) therefore has to be placed as close as possible to the sample. $L$ is typically chosen to be 0.5 m, determined by the available space and $\lambda$ is determined by the experimental needs. The size of the upstream aperture $D_1$ can be tuned separately in the horizontal and vertical direction to accommodate the fact that the vertical divergences of the soft X-ray beamlines used in this work are generally 10 times larger than the horizontal divergence (Bessy-II). As a consequence of the spatial filtering required to generate a sufficiently coherent X-ray beam, approximately
3.3 X-ray free-electron laser and self-amplified spontaneous emission

Fig. 3.2 Spatial filtering for transverse coherence. Left panel: schematic of the filtering mechanism. With two apertures, the transverse coherent portion of the light can be filtered out by selecting one of the plane waves of the incoming light fronts. The diameters of the two apertures depend on the wavelength of the incoming light. Right panel: an overexposed far field image of the filtered beam after the second aperture showing the interference fringes, known as the Airy pattern, showing that the beam is first order transversely coherent. Courtesy of J. Peters and J. B. Goedkoop.

3 orders of magnitude of the light flux is discarded. After monochromatization and spatial filtering the resultant coherent flux is typically of the order of $10^9$ photons per second.

Fortunately, the source size of synchrotrons continues to improve, and the Hamburg storage ring Petra-III currently holds the record for the smallest source size/divergence. At hard X-ray beamline P10 at Petra-III ($\lambda \sim 1$ Å), is specially designed for coherent scattering, and the available coherent flux is $10^{11}$ photons per second in a spot diameter of $\sim 10$ µm, obtained by focusing the beam. Under these conditions, no spatial filtering is necessary. Data from both Bessy-II and Petra-III will be presented in chapter 4 and chapter 5.

3.3 X-ray free-electron laser and self-amplified spontaneous emission

Given the complications of spatial filtering and the strong reduction of the available intensity, clearly having an intrinsically coherent X-ray source, also in the soft X-ray region of the spectrum, has long been a big dream of the community. However, the generalization of the laser concept to the X-ray regime has been an difficult process, since X-rays cannot be constrained to a optical cavity using mirrors. In the 1970s, the first free-electron laser (FEL)
was built by John Madey and his colleagues in Stanford University [87–90]. The advantage of a FEL is that it does not require the optical cavity, opening up a pathway to X-ray lasers. In this thesis, we describe our pioneering experiments at the Linac Coherent Light Source (LCLS) on X-ray correlation spectroscopy (XCS) from hard condensed matter systems. LCLS is the world’s first operating X-ray FEL, and therefore we give a short introduction to how the FEL radiation comes about here, and give a brief introduction to this unique facility later in this chapter.

A relativistic charged particle radiates X-rays when it travels in a curved path forced by a magnetic field. In a conventional undulator used at synchrotron facilities, an array of permanent magnets with alternating polarity causes the charged particles to oscillate transversely. Typically the number of periods of the undulator is around 50 and the length of the so-called insertion device is \( \sim 1 \) meter. The light emitted at the successive bends interferes constructively at wavelengths fitting to the undulator period, resulting in a high-brilliance X-ray beam. While the beam is intense, it is only partially coherent, due to the electron beam divergence and the finite number of undulator periods, so that in general spatial and spectral filtering is necessary with concomitant losses in flux, as described earlier.
3.4 Speckle methodology

Nevertheless, the same concept lies at the basis of the X-ray laser. In order to obtain an X-ray laser, the self-amplified spontaneous emission (SASE) principle was first exploited in 1984 by Bonifacio, Pellegrini and Narducci [92]. When the undulator is long enough, i.e. several tens of meters, the X-ray intensity becomes so great that the electromagnetic field starts to interact with the original electron bunch, modulating the electron bunch into ‘micro-bunches’ with interval equal to the wavelength of the light. This strongly nonlinear self-amplifying process leads to a true coherent motion of the electrons with the electromagnetic field. This process can boost the coherent X-ray flux by several orders of magnitude compared to 3rd generation synchrotron sources [93–96]. In Fig. 3.3, an illustration is given for a high gain XFEL. As we will see later, however, because of the spontaneous nature of the emission process, it is not predictable which micro-bunch will be amplified, and also the emitted X-ray pulse intensity can vary strongly from bunch to bunch. Nevertheless, the excitement in the field due to the game-changing characteristics of XFEL radiation is very clear, and it was both an enormous opportunity and challenge to use such an unique source of some of the experiments described in this thesis.

3.4 Speckle methodology

In this section we will briefly recap some results on speckle statistics from the book of Goodman [97]. This will provide a good basis to understand the speckle phenomenology, and to discuss the coherent properties of the LCLS beam that can be extracted from our XFEL data in what follows. For more detailed discussions, readers are kindly requested to refer to this book.

3.4.1 Phasor sum

A speckle pattern results from the interference of the light. A speckle is formed by a number of randomly phased electromagnetic waves, or *phasors*, which can be expressed by a sinusoidal function of time and space,

$$\mathcal{A}(x,y,t) = A(x,y,t)\cos[\omega t - \theta(x,y,t)],$$  \hspace{1cm} (3.4)

where $A$ represents the space and time dependent amplitude, $\omega$ is the ‘carrier’ frequency and $\theta$ represents the phase of the signal $\mathcal{A}$. Here the polarization of the signal is not included and $\omega$ is typically much larger than the bandwidth of the amplitude or phase [97]. By using...
Euler’s formula, this signal is simply projected on to the complex plane,

\[ \mathcal{A}(x, y; t) = \frac{A(x, y; t)}{2} (e^{-i(\omega_0 t - \theta(x, y; t))} + e^{i(\omega_0 t - \theta(x, y; t))}) , \]  

(3.5)

For the sake of notation convenience, we suppress the positive frequency and double the negative one. Then the signal (phasor) is now represented by,

\[ \mathcal{A}(x, y; t) = A(x, y; t)e^{-i\omega_0 t} , \]  

(3.6)

where \( A(x, y; t) = A(x, y; t)e^{i\theta(x, y; t)} \), is the carrier frequency suppressed amplitude.

Moreover, a speckle can be seen as a phasor sum formed by the superposition of \( N \) phasors at a single point in space-time. It can be expressed by,

\[ A(x, y; t) = \frac{1}{\sqrt{N}} \sum_{n=1}^{N} a_n e^{i\phi_n(x, y; t)} , \]  

(3.7)

where \( \frac{1}{\sqrt{N}} \) is the normalization factor, \( a_n \) is the amplitude of a single phasor component, and \( \phi_n \) is its phase.

### 3.4.2 Probability density function of the speckle amplitude

Phasor sums cover the complex plane randomly, but it turns out that one can derive some important statistical properties. Under the assumptions that the amplitudes and phases of all phasors are statistically independent and that the phases are uniformly distributed ([97]-p.8), it can be shown that the probability density function of the amplitude of a phasor sum is given by a Rayleigh density function:

\[ P_A(A) = \int_{-\pi}^{\pi} p_{A, \theta}(A, \theta) d\theta = \frac{A}{\sigma^2} \exp\left(-\frac{A^2}{2\sigma^2}\right) , \]  

(3.8)

### 3.4.3 From amplitude to intensity

CCD detectors only measure the intensity, \( I = |A|^2 \) of the phasor sum. Following this procedure, one finds that for a speckle series with an amplitude probability density function as in Eq. 3.8, the intensity probability density function is given by a negative exponential distribution,
\[ P_I(I) = \frac{1}{\bar{I}} \exp\left(-\frac{I}{\bar{I}}\right), \quad (3.9) \]

where \( \bar{I} \) is the average intensity. The standard deviation of this distribution is \( \sigma_I \). Speckle intensity statistics meeting this distribution are said to be resulting from fully developed speckle.

Often, parts of the above assumptions are not met, also for the important case where some of the speckles are not fluctuating. The scattering from this fixed part adds an additional fixed phasor component, which strongly modifies the above statistics in manners described in Goodman [97]. As the fixed phasor component increases, the intensity probability density function approaches, ultimately, a Gaussian distribution.

### 3.4.4 Speckle contrast

Strictly speaking, for a speckle pattern, the term contrast describes the strength of the intensity fluctuations in the pattern, compared to the average intensity [97], which is defined as,

\[ C = \sqrt{\langle I^2 \rangle_{xy} - \langle I \rangle_{xy}^2} = \frac{\sigma}{\bar{I}}, \quad (3.10) \]

where the bracket stands for averaging over pixels, \( \sigma \) is the standard deviation of the intensity, and \( \bar{I} \) is the average intensity of the pattern. This definition will be used in the coming sections.

### 3.4.5 Low intensity statistics

For the X-ray free electron laser experiments described in this thesis, the scattered intensities are often weak and the resulting image is no longer a recognizable pattern with dim and bright speckles, but instead consists of only discrete photon events. The statistical properties of such low intensity speckle images are discussed here.

With certain assumptions, the possibility of recording \( K \) photon events in an area within a time interval is described by a Poisson probability distribution,

\[ P(K) = \frac{\bar{K}^K}{K!} e^{-\bar{K}}, \quad (3.11) \]
where $\bar{K}$ is the average photon number detected from this area within a time interval. $\bar{K}$ is linearly related to the integrated intensity $I$ via the relation $\bar{K} = \alpha I$, where $\alpha$ is a conversion coefficient from recorded intensity to photons events.

Due to the stochastic nature of the SASE process at LCLS, the incident intensity itself is also a speckle-like time series. In this case the intensity can be approximated as a gamma density distribution function $P(I)$, which is given by,

$$P(I) = \left( \frac{M}{I} \right)^M \frac{e^{-M/I}}{\Gamma(M)}, \quad (3.12)$$

where the intensity $I$ is zero or positive, and the $M$ is the number of coherent modes.

This same is true for $\bar{K}$. Thus the probability of observing $K$ photon events, given the distribution of incident intensity, can be expressed using the conditional probability, $P(K|I)$ as

$$P(K) = \int_0^\infty P(K|I)P(I)dI. \quad (3.13)$$

By inserting the Gamma distribution of $P(I)$ in Eq. 3.12 into this formula, we obtain a general expression for the photon count statistics for low intensity speckle:

$$P(K) = \frac{\Gamma(K+M)}{\Gamma(K+1)\Gamma(M)} \left( \frac{\bar{K}}{K+M} \right)^K \left( \frac{M}{K+M} \right)^M. \quad (3.14)$$

Here $\Gamma$ is the Gamma function, and $M$ denotes the number of modes in the speckle pattern, which is the reciprocal of the contrast [97–99]. The total expression is known as the negative binomial distribution. When $\bar{K}$ is fixed, this expression converges to the Poisson distribution for large $M$.

### 3.4.6 Time-correlation function

A central concept in the analysis of XCS data is the time-time correlation function. Consider a signal $I(t)$ as a function of time at a given scattering vector $\mathbf{q}$. At any given time $t_0$, a value $I(\mathbf{q},t_0)$ is defined, and after a time delay by $\tau$, the signal becomes $I(\mathbf{q},t_0+\tau)$. The product $I(\mathbf{q},t_0)I(\mathbf{q},t_0+\tau)$ measures the correlation of the two values. For an infinite time series, the time average of the infinite number of pairs separated by $\tau$ is $G(\mathbf{q},\tau) = \langle I(\mathbf{q},t_0)I(\mathbf{q},t_0+\tau) \rangle_{t_0}$, where the brackets indicates averaging over all possible starting times. The function $G(\mathbf{q},\tau)$ is called the time-correlation function of the signal $I(\mathbf{q},t)$. For each scattering vector $\mathbf{q}$ in reciprocal space, such a correlation function can be measured allowing one to obtain the correlation length dependent dynamics of the disordered system [70].
3.4 Speckle methodology

It is convenient to define the normalized time-correlation function,

\[ g_2(q, \tau) = \frac{\langle I(q, t)I(q, t + \tau) \rangle_t}{\langle I(q, t) \rangle_t^2}, \]  

where the triangular brackets again indicate the averaging over the subscript variable. As shown in Fig. 3.4, for a dynamical system that produces speckle patterns with fully developed speckle, the normalized time-correlation function is 2 for \( \tau = 0 \) and decays to 1 at long times, when all correlations are lost. Such a correlation function can be written as

\[ g_2(q, \tau) = 1 + \beta |f(q, \tau)|^2, \]  

where \( \beta \) is the coherence factor of the light source, which ideally is 1 for fully coherent beam, and \( f(\tau) \) is the intermediate scattering function, which represents the dynamics of the form factor, whereby a scattering vector dependence has been added. Thus, by measuring the \( q \) dependent time correlation function one can obtain the decay of the intermediate scattering function at different length scales.

In the analysis of the speckle movies obtained during the research reported here, we made extensive use of the dedicated package XPCSGUI developed by M. Sprung and Z. Jiang at Advanced Photon Source in Argonne National Laboratory, which allows one to calculate
the correlation function per speckle. The resulting functions can be averaged over ranges of scattering vectors in order to enhance the sampling statistics.

### 3.4.7 Contrast reduction for dynamical speckle

The time-correlation function is the standard way to analyze speckle time series in XCS. However, there are two important cases for which it is not an appropriate method of data analysis. Firstly, if the detected intensity is too low due to insufficient incoming flux, strong absorption or low scattering cross sections, or secondly, if the source has fluctuating intensity. The latter can happen sometimes at synchrotron beamlines and is always intrinsic to the unseeded SASE beam from LCLS.

Under these circumstances, for which $g_2$ method is unsuitable, it is more productive to analyze the data in terms of the evolution of the speckle contrast. To picture how the contrast can give access to the dynamics, consider how a photo of an object that moves during the exposure time is ‘blurred’ and the details of the object cannot be identified clearly. This can be quantified in terms of the contrast reduction in comparison with the static case. The same holds for speckle patterns as they are the reciprocal representation of the real space image.

Consider the sum of two speckle patterns $S(\tau) = I(t_0) + I(t_0 + \tau)$, where $I(t_0)$ and $I(t_0 + \tau)$ denote the speckle intensity at time $t_0$ and $t_0 + \tau$. By the definition given in Eq. 3.10, the contrast of the summed image is given by

$$C_2(\tau) = \sqrt{\frac{\langle S(\tau)^2 \rangle_{xy} - \langle S(\tau) \rangle_{xy}^2}{\langle S(\tau) \rangle_{xy}^2}}. \quad (3.17)$$

For a fully developed speckle pattern one can use the Siegert relation to write the contrast of the form [100, 101],

$$C_2(\tau) = \sqrt{\beta^2(1 + f(\tau)^2)} + \alpha, \quad (3.18)$$

where $\beta$ denotes the coherence factor of the light source, $f(\tau)$ the intermediate scattering function, and $\alpha$ the shot noise, which is important at a pulsed light source, such as X-ray free electron laser. It is not hard to see that in the absence of shot noise, the contrast of the sum of two uncorrelated speckle patterns is reduced to $\frac{1}{\sqrt{2}}$ with respect to the contrast of the individual patterns. In Fig. 3.5, we plot the evolution of the contrast of the sum of two images
3.4 Speckle methodology

Fig. 3.5 Evolution of the contrast of a dynamical system with exponentially decaying correlations resulting from Eq. 3.18 with shot noise set to zero. The correlation time is denoted by $\tau_c$.

separated by a given delay time $\tau$ for a sample with exponentially decaying fluctuations of the intermediate scattering function $|f(\tau)|$.

As mentioned, this contrast method is useful when the sequential $g_2$ analysis cannot be applied. The two procedures differ with regards to the order of averaging: in the contrast method, the contrast of a region of the diffraction pattern within a certain scattering vector range $q$ is calculated before calculating the time correlations, while in the time correlation method first all time correlations per pixel are determined, after which an averaging over a $q$ range is performed in order to enhance statistics.

3.4.8 Pixel effect on contrast

When using a pixelated 2D detector care has to be taken to match the pixel size to the speckle size according to the spatial Nyquist sampling criterion [102, 103]. However, it has been shown that the contrast will always be reduced by the pixelation of the image. The effective contrast $C_{eff}$ is found to be

$$C_{eff} = C \frac{1}{1 + \left(\frac{P}{S}\right)^2},$$

(3.19)

where $P$ is the pixel size, and $S$ is the speckle size [104]. The $\frac{P}{S}$ ratio can be increased by moving the detector further from the sample, but this naturally goes at the cost of signal to noise ratio. Alternatively, the pixel density can be increased. An experimental rule of thumb states that if pixel size is equal to speckle size, one obtains the best signal to noise
ratio. However, as Eq. 3.19 shows, even in the P=S case, the contrast obtained is 50% of the theoretical value.

### 3.5 Instrumentation

The experiments performed on hard condensed matter systems require an uncompromising set-up that meets the following requirements:

- a brilliant 3rd generation synchrotron light source in order to have sufficient coherent intensity,
- both spectral and spatial filtering to obtain the monochromatic and coherent portion of the light
- a vacuum diffractometer, both because of the necessity of reducing air scattering and absorption and because of the necessity of cryogenic cooling of the sample,
- an X-ray CCD detector with high quantum efficiency and appropriate readout time, as the latter determines the sampling rate. For most current detectors the read out time is over one second. New generation devices, such as the Timepix detector discussed later on in chapter 4 and Appendix B of this thesis, allow a faster data acquisition,
- stability over the duration of the experiment, which is typically several hours, this time scale being set by the limitation of the detector’s read-out speed.

The soft X-ray experiments described here used a beamline that is designed and laid out for spectroscopic experiments. This means that we had to take care of the spatial filtering ourselves. To this end, during this project, a special in vacuum ultra-stable sample and pinhole stage was designed in Amsterdam to fit the Köln/Bessy in-vacuum soft X-ray diffractometer (AG Schüßler-Langeheine). In this section, we will first describe these two parts: the Köln-Bessy chamber and the Amsterdam insert, which was designed to optimize the stability of the beam position with respect to the sample during temperature or diffraction angle changes.

#### 3.5.1 Vacuum diffraction chamber for soft X-ray scattering

The chamber (see Fig. 3.6) [105] has two levels of each 8 CF150 ports equally distributed in the horizontal plane. On the rotating top flange of the chamber, 6 CF100 ports are placed for mounting in vacuum CCD cameras and photodiodes. The chamber has two differentially pumped concentric rotation circles for $\theta - 2\theta$ motions in the standard Bragg diffraction
Fig. 3.6 A sketch of the vacuum diffraction chamber. The colors indicate the different construction parts. Adopted from PhD thesis of M. Buchholz [105].
Fig. 3.7 Drawing of the ultra-stable coherent scattering insert. (a) Overview of the diffractometer. (b) The sample cavity; it includes a silver braid, sample stage and in-plane (XY) piezoelectric driven motors. The red housing is used to couple the stage with the $\theta$ rotation and the vertical (Z) motion at the top of the main chamber. (c) The mobile pinhole stage, with (YZ) piezoelectric driven motors. It is fixed to the bottom support tube to eliminate the relative motion between the incoming light (red beam) and the sample. (d) The bottom support tube with ball bearings allowing $\theta$ and Z motions.

gometry. A Z-axis motorized vacuum manipulator (not shown in Fig. 3.6) can be mounted on top of the $\theta$ motion, inside which a cryostat can be installed at the center of rotation of the chamber.

### 3.5.2 Coherent setup

The ultra-stable insert which consists of a sample stage and a pinhole stage, both mounted on a stable platform is shown in Fig. 3.7. The cooling power is transported from the cryostat via a flexible silver braid. On the two ends of the silver braid, copper connectors are used to make rigid connection to the cryostat and the sample stage.

The sample stage itself consists of a vacuum transferable sample holder and receptor designed by AG Schüßler-Langeheine at BESSY and a thermal isolator that is mounted on
3.5 Instrumentation

Fig. 3.8 (a) The sample stage: vertical section. (b) Exploded view of the elephant foot, showing the two concentric titanium cylinders cut by spark erosion. In each of these, eight narrow beams are cut, which support the sample rigidly while minimizing thermal conduction. The two cylinders are welded together on the free back end. The sample holder part of the construction is electrically insulated to allow the drain current measurements and hosts slots for the cryogenic heater and temperature sensor.

The thermal isolator (elephant foot) is a cylindrical titanium device. It was designed to minimize the thermal motion of the sample during temperature changes. The elephant foot in this coherent setup is an upgraded version of its predecessor developed for ARPES manipulators by Patthey and Golden. The version consists of two coaxial titanium cylinders of 1.5 mm thickness in which diagonal cuts define 8 beams with a width of 1 mm, thus minimizing the cross section for thermal conduction whilst maintaining rigidity. (See Fig. 3.8.) The elephant foot has a free back end allowing the two cylinders to compensate each others’ thermal expansion or contraction, ensuring that the sample surface stays aligned with the theta rotation axis over all the temperature range.

The temperature of the flow cryostat is typically set to be some 10 K below the set point of the sample. A cryogenic heater and a temperature sensor on the sample stage stabilize the sample temperature at the desired value to within 20 mK.

As shown in Fig. 3.7 (b), the sample stage sits on two piezoelectric driven motors with encoders having 100 nm resolution that position the sample in the XY horizontal plane. The whole assembly is kept on the rotation axis of the chamber by a set of vertical ball bearings.
mounted on a stainless steel support tube that is rigidly attached to the concentric bottom flange of the diffraction chamber.

The coherent pinhole stage is also mounted on the support tube in order to reduce drifts of the pinhole with respect to the sample (see Fig. 3.7). Thanks to another pair of piezoelectric driven motors, the coherence defining pinhole can be positioned accurately in the center of the incoming light beam defined by an upstream aperture. The downstream pinhole has a diameter of $15 \mu m$ that is laser drilled in a $100 \mu m$ thick tungsten foil. In order to bring it close to sample, the pinhole is mounted on a tube that is concentric to the incoming beam and the tube is clamped to the piezoelectric motors via an aluminum connection piece (see Fig. 3.7 (c)).

During the experiments it became clear that the encoders of the in-vacuum piezoelectric motors produce considerable amounts of infrared radiation that is visible to the X-ray CCD camera. In order to prevent this infrared contamination, the encoders had to be switched off during measurements.

The horizontal and vertical divergence of the incoming beam can be controlled by slits that are located at 50-70 cm upstream of the pinhole. The horizontal slit is formed by the controlled gaps between 2 vertically mounted razor blades on 2 horizontal translational motors. The vertical divergence of the beam is defined by a set of vertically arranged rectangular slits machined in a thin metal plate. Slits of differing vertical widths can be placed in the beam by means of a motorized vertical translation stage acting on the whole slit assembly.

3.5.3 Vacuum diffractometer at the hard X-ray beamline P10 at Petra-III

The hard X-ray beamline P10 (see Fig. 3.9) is dedicated for coherent x-ray scattering experiments. Here the previously described setup shown in Fig. 3.7 was slightly modified to accommodate the P10 end station. A custom, compact diffraction chamber is mounted on an aluminum frame above the 4-circle Huber diffractometer. In this case the Huber is not used to minimize the modifications required for the existing setup.

The chamber hosts an simplified version of the sample stage shown in Fig. 3.7 (b). Since the beamline is optimized to deliver coherent light, the pinhole aperture option of the sample assembly is thus removed. The chamber is connected to the beamline via bellows. Using turbo pumping, we could maintain a vacuum to about $10^{-5}$ mbar even given the large Kapton window which represented the vacuum seal for the slit-shaped opening through which the diffracted x-rays exit the sample chamber (see Fig. 3.10 (b)). After exiting through the
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Fig. 3.9 An overview of the P10 beamline layout. The main parts are indicated with color coding.

Fig. 3.10 (a) The vacuum diffractometer at P10. Its highlighted sections are indicated by color overlays. (b) the zoomed in chamber section. The beam entry port is on the back of the chamber and the diffracted light exits through the horizontal slit, covered by a Kapton window.
3.5.4 LCLS and the X-ray correlation spectroscopy station

In 2009, the first X-ray laser pulse was sent out from the Linac Coherent Light Source (LCLS) [95, 107], operated by SLAC National Accelerator Laboratory. This is the first FEL-based hard X-ray source that can be accessed by general users. The LCLS has 2 experimental halls, which host 6 end stations, each of which is dedicated to a particular research field. Among them is a station for X-ray correlation spectroscopy (XCS) [75, 108, 109].

LCLS operates at a frequency of 120 Hz, where the 14 GeV electron beam pulses generated by the LINAC travel through a 132 meter long undulator producing a broad bandwidth SASE radiation, dubbed the pink beam. This beam passes the Front End Enclosure (FEE), which hosts a series of beam diagnostics.

A four-jaw X-ray slit system defines the beam before the gas-filled attenuators. The attenuated pink XFEL pulse energy is then measured indirectly using the UV fluorescence with a pressurized gas detector [110, 111]. A number of motorized scintillators can be moved in to directly image the beam profile with a camera.
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Fig. 3.12 LCLS diffractometer. (a) The construction of the chamber on the 4-circle Huber stage at XCS beamline with color coding. (b) The sample holder is the same as described in Fig. 3.7.

Passing through the FEE, the pink-beam X-rays are band-filtered by a monochromator set, before coming into the XCS experimental station. The monochromator set consists of two Si (111) crystals that offset the beam in the horizontal plane, followed by a channel cut Si(111) monochromator, with a spectral resolution of 1.2 eV at 8.8 keV. Further slits and a set of Be-lenses are used to define and focus the beam down to 12x18 µm². An intensity-position monitor is located downstream of the monochromator [112], which measures the monochromatic intensity with a transmission rate of 99%. This monochromatic XFEL beam contains a coherent flux of \( \sim 10^{11} \) photons per \( \sim 30 \text{ fs pulse} \) after the XCS monochromator [98], which is the same number of coherent photons as the P10 beamline at one of the very best 3\(^{rd}\) generation synchrotron sources, Petra-III emits in one second.

The XCS diffractometer setup is similar to that used at P10 at PETRA-III. However, this time we mounted a modified version of the vacuum chamber (see Fig. 3.12) directly on the 4-circle Huber stage for the sample motions. The chamber is detached from both the beamline and the flight tube (see Fig. 3.11), and thus has both entrance and exit windows covered by Kapton foils. In the sample space, a vacuum of \( 10^{-5} \text{ mbar} \) is maintained, similar to the situation at P10. After a 7 meter long evacuated flight tube, the diffracted light can be captured by the Timepix (see Appendix B) or the Pixis 1300 detector.
The XCS station at LCLS is still under commissioning, and in fact for the experiment described in Chapter 4, we were only the second user group at this station, where we exploited the time structure of LCLS by recording speckle movies at 120Hz, as reported later in this thesis.

As the next step in XCS development at LCLS, a so-called split-and-delay line is being tested, in which a single light pulse can be separated into two pulses with controllable delay time in the range of femto- to nanoseconds. This will make it possible by means of the contrast reduction method to correlate two speckle images separated at these ultrafast time scales, opening up a completely new time window to speckle techniques [75, 113, 109] in the foreseeable future.