The X-ray Correlation Spectroscopy instrument at the Linac Coherent Light Source

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Abstract. The X-ray Correlation Spectroscopy instrument (XCS) at the Linac Coherent Light Source (LCLS) is a dedicated instrument using coherent x-ray scattering techniques to investigate dynamics in condensed matter systems. XCS can probe both slow and ultrafast dynamics on lengthscales of interest. It employs an extensive suite of X-ray instrumentation to tailor the LCLS X-ray beam properties to experimental requirements. Results demonstrating the full transverse coherence of the LCLS beam are presented.

1. Introduction
The unprecedented brilliance and narrow pulse duration of the Linac Coherence Light Source (LCLS) provides a unique opportunity to observe dynamical changes of large groups of atoms in condensed matter systems over a wide range of time scales using Coherent X-ray Scattering (CXS) in general and X-ray Photon Correlation Spectroscopy (XPCS) in particular. The X-ray Correlation Spectroscopy (XCS) instrument at the LCLS investigates equilibrium and non-equilibrium dynamics in disordered or modulated materials [1, 2].

Coherent X-rays are particularly well suited for investigating disordered system dynamics down to nanometer and atomic length scales by using X-ray Photon Correlation Spectroscopy [1]. When coherent light is scattered by a disordered system, the scattering pattern presents a peculiar grainy appearance known as speckles. The speckles originate from the exact position of all scatterers within the coherently illuminated volume. XPCS thus characterizes the temporal fluctuations of such speckle patterns yielding information about the dynamic behavior of the system [1].

XPCS, as currently performed on 3rd generation synchrotron light sources is complementary to Photon Correlation Spectroscopy (PCS) with visible coherent light, which probes slow dynamics (< 10^6 Hz) but can only access the long wavelength regime (Q < 4 · 10^{-3} Å^{-1}). Neutron-based techniques (inelastic, quasi-elastic neutron scattering, neutron spin-echo) and Inelastic X-ray Scattering (IXS) can access the same Q range as XPCS, but these techniques probe the dynamic properties of matter at high frequencies (typically from 10^8 Hz to about 10^{17} Hz), as illustrated in Figure 1. The brown XPCS areas (in transmission/diffraction and Grazing Incidence (GI) geometry) in Figure 1 are defined from existing XPCS measurements performed at existing 3rd generation synchrotron light sources (the light purple XPCS area is the range
2. XPCS at the XCS instrument at LCLS

The XCS instrument can operate in two different modes which allow the investigation of different time scales.

2.1. Sequential mode

The shortest time scale of this mode of operation is limited by the LCLS repetition rate (i.e. up to 120Hz) and is illustrated by the Sequential Mode area in the frequency-wavevector graph in Figure 1. It employs the very large time-averaged coherent X-ray flux from the LCLS (averaged over the 120 Hz repetition rate) to investigate dynamics by means of 2D-XPCS data collection. These experiments consist of collecting time-resolved sequences of speckle patterns with an area detector. The temporal evolution of speckle patterns is quantified by calculating intensity autocorrelation functions, from which one can access characteristic relaxation times of the dynamics of the system [2]. Typical characteristic times ranging from a few inter-pulse periods up to many minutes can thus be measured on length scales of interest for a given system.

2.2. Split and Delay mode

The pulsed nature of the LCLS opens up opportunities to probe dynamics on much shorter timescales than those achievable at storage rings. Utilizing a split and delay scheme allows probing ultrafast dynamics in the $10^{-12}$ to $10^{-8}$s range, as indicated by the ”Ultra Fast Mode” area in Figure 1. This range was previously accessible solely by light, neutron and x-ray inelastic scattering techniques. The split and delay technique takes advantage of the peak brilliance of the LCLS beam. The concept is to split each LCLS X-ray pulse into two equal-intensity pulses separated in time, but propagating along the same path [3]. The scattering from the two pulses is collected during the same exposure of an area detector. If the sample is static (i.e. no dynamics on the time scale of the time delay between the two pulses), the contrast in the summed speckle patterns will be identical to that of a single pulse. If on the other hand, the sample evolves on this time scale the summed speckle pattern will show a reduction of contrast [4]. A characteristic relaxation time can then be extracted from the time-delay dependence of the measured contrast. This novel scheme provides access to ultrafast dynamics in condensed matter systems such as liquids and glasses.
3. XCS instrument configuration
The XCS instrument is a hard X-ray instrument that was designed to utilize the LCLS FEL beam for coherent X-ray scattering techniques such as XPCS. A schematic representation of the various optical components of the XCS instrument is provided in Figure 2.

![Figure 2. Schematic view of the optical components and diagnostics of the XCS Instrument.](image)
The distances from each component to the sample are indicated in meters. XCS can operate various monochromators: a Si(111) or (220) Large Offset Double Crystal Monochromator (LODCM) and an artificial Si(511) Channel Cut Monochromator (CCM). The beam can be focused to small sizes (typically 5 × 5µm²) with Compound Refractive Lenses (CRL) available for installation at two different locations. XCS can also operate in pink beam mode by translating most of its components into the main LCLS beamline as indicated in red.

Along the XCS beamline and its various components, a series of slits and diagnostics, such as intensity monitors [6] and YAG viewing cameras, are available for the characterization and tuning of optical components. The XCS instrument operates primarily with monochromatic radiation, as required for controlling the longitudinal coherence of the beam. A Large Offset Double Crystal Monochromator (LODCM) is located 44m upstream from the sample. It has either Si(111) or (220) single crystals which deliver the X-ray beam over energies ranging from 5 to 24keV. It provides a 60cm horizontal offset from the main LCLS hard X-ray line (i.e. as indicated by the black beam path in Figure 2). If further monochromatization is required, a double crystal artificial Channel Cut Monochromator (CCM) [5] utilizing Si(511) can be used for energies ranging from 7.5 to 9.5keV. A fixed energy (i.e. 7.908 or 8.4keV) split and delay unit [3], located 12.5m upstream of the sample, can generate double X-ray pulses allowing access to ultrafast dynamics as described in Sec. 2.2. Standard X-ray optics such as Compound Refractive Lenses (CRL), attenuators and harmonic rejection mirrors are used for tailoring the X-ray beam to desired experimental conditions. A 4-circle diffractometer operating in horizontal scattering geometry is used in conjunction with a Large Angle Detector Mover. It covers horizontal scattering angles up to 55 degrees in addition to providing sample detector distances up to 7.5m. In addition it permits to operate in various scattering geometries: Small Angle X-ray Scattering, diffraction but also reflectivity and Grazing Incidence. The XCS instrument can also operate in pink beam, if required, where the first crystal of the LODCM is removed from the beam path and most of the optical components located downstream are translated into the main LCLS line (as shown by the red scheme in Figure 2).
4. Commissioning results on transverse coherence

In order to confirm the full transverse coherence of the LCLS beam (as is expected for Free Electron Laser sources), we generated 9.5keV coherent scattering patterns from a slit located at 196m (cf. Figure 2). The data were recorded as a function of the slit opening (0 to 500µm) on a high resolution YAG viewing system located 167.9m downstream at 28.1m (cf. Figure 2). Diffraction patterns showing Fraunhofer and Fresnel diffraction are presented in Figure 3.(a), (b) and (c) for openings (gaps) of 100, 150 and 400µm respectively. A movie showing the continuous evolution of the diffraction pattern as a function of the slit size is shown as Supplementary Material. The Fresnel diffraction of the 400µm slit (as defined for slit openings larger than 150 – 200µm when transitioning from the far to the near field regimes), clearly confirms that the beam is fully transversely coherent over the opening of the slit, which also turns out to be the Full Width at Half Maximum of the beam size at this location.

![Figure 3](image)

**Figure 3.** Coherent diffraction patterns of a square slit (with an opening defined by the gap in the upper-right corner). The patterns were measured at 9.5keV in pink beam with a slit-to-YAG screen distance of 167.9m. A movie showing the evolution of the diffraction pattern as a function of the continuous opening of the slit gap is presented in Supplementary Material.

References

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