Studies of Nanoscale Dynamics in Condensed Matter using X-ray Photon Correlation Spectroscopy at LCLS

Category A Letter of Intent
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(Contact information for all team members is given in Appendix 1.)

Scope of Work:

Summary: We propose a research program on nanoscale dynamics in condensed matter, carried out primarily using X-ray Photon Correlation Spectroscopy (XPCS) at an experimental endstation optimized specifically for XPCS experiments. This endstation will also support other types of scattering experiments that use the transverse coherence and short pulses of the LCLS source to characterize samples with minimal disturbance (heating/radiation damage) from the beam.

Science Case:

Complex nanoscale dynamics is a ubiquitous phenomenon at the frontier of research in condensed matter. Viscoelastic flow of liquids, polymer reptation, protein folding, crystalline phase transitions and domain switching, and countless other *collective processes* show both fast and slow equilibrium dynamics, often sensitive functions of temperature, applied fields, and fabrication parameters. Using time- or energy-resolved light-scattering techniques, we can currently probe the full range of these time scales, from femtoseconds through kiloseconds. However, since scattering probes structure at length scales of the wavelength or longer, measurements with visible light are not sensitive to phenomena below 0.1 micron. But in almost all materials, intermolecular interactions are short-range and correlations in quantities such as polymer or ferroelectric alignment, motions of polymer or protein subunits, or liquid state molecular orientation extend over nanometers rather than microns. Inelastic neutron and x-ray scattering techniques allow study of these short length scales, but are insensitive to time scales longer than nanoseconds or to small samples.

The unprecedented brilliance and narrow pulse width of the LCLS provides a unique opportunity to observe nanoscale dynamics in condensed matter systems over a wide range of time scales using X-ray Photon Correlation Spectroscopy (XPCS) [1]. Figure 1 shows the relationship between XPCS and other techniques for probing equilibrium dynamics [2]. Various techniques can be used to measure the dynamic structure factor $S\ (Q,\omega)$ or the corresponding

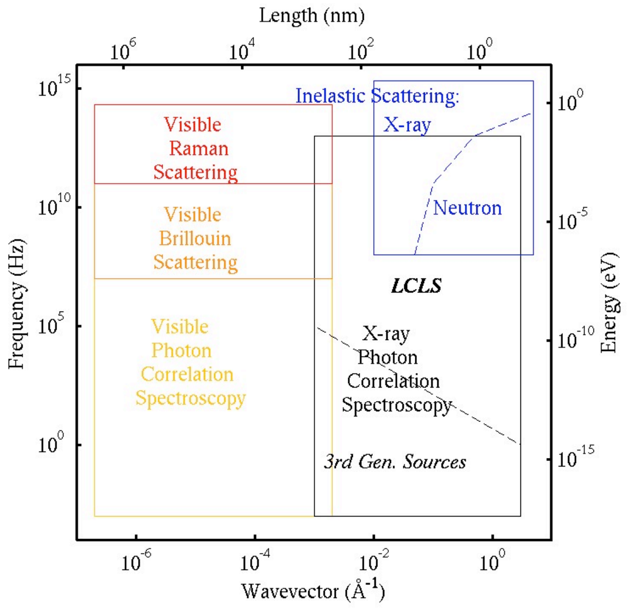

FIG. 1: Plot of the frequency-wavevector space relevant to dynamics in condensed matter and the regions covered by various probes of equilibrium dynamics (after [2]).

response function $S(Q,t)$ in different regions of frequency ω (or time t) and wavevector Q space. The figure shows that XPCS provides a time-domain, small-length-scale spectroscopy highly complementary to other techniques, uniquely covering ω lower than 10^8 Hz and Q larger than about 10^{-3} A^{-1}, as well as the gap between visible and neutron techniques at high ω in the nanoscopic length scale regime.

The time and length scales covered by XPCS experiments at LCLS are of great interest for the study of dynamics in many forefront research areas in condensed matter physics. The scientific case for experiments using XPCS techniques at 4th generation x-ray sources such as LCLS has been developed in several workshop reports and design report documents [1,3,4]. Examples of problems that could be addressed are:
***Simple Liquids* –** Transition from the hydrodynamic to the kinetic regime.
***Complex Liquids* –** Effect of the local structure on the collective dynamics.
Polymers – Entanglement and reptative dynamics.
Biological Macromolecules – fluctuations between conformations (e.g. folded and unfolded).
***Glass Formers* –** Vibrational and relaxational modes in the mesoscopic region.
***Dynamic Critical Phenomena* –** Order fluctuations in alloys, liquid crystals, etc.
***Charge Density Waves* –** Direct observation of sliding dynamics.
***Quasicrystals* –** Nature of phason and phonon dynamics.
***Surfaces* –** Dynamics of adatoms, islands, and steps during growth and etching.
Defects in Crystals – Diffusion, dislocation glide, domain dynamics.
***Ferroelectrics* –** Order-disorder vs. displacive nature; anisotropy and size effects.
***Magnetic Films* –** Observation of magnetic relaxation times.
***Lubrication* –** Correlations between ordering and dynamics.
Detailed discussions of specific cases have been given in other documents (see e.g. discussion of polymers, liquids, and glasses in [3] and dynamic critical phenomena, surfaces, polymers, magnetic films, and lubrication in [4]) and for brevity will not be repeated here.

The use of XPCS has been limited to date by the availability of sufficient coherent x-ray flux. Since the flux of coherent x-rays is proportional to the source brilliance, XPCS studies benefit directly from higher brilliance sources. The modest coherent x-ray flux available at 3rd-generation synchrotron sources makes XPCS measurements currently feasible at ω up to the kHz range, depending upon Q and the scattering efficiency of the material. The impact of the high coherent x-ray flux from LCLS will be to allow XPCS measurements at higher ω and on a greater variety of materials, overlapping with energy-domain measurements using neutron and x-ray inelastic scattering. Time-domain XPCS measurements will provide truly complementary information for understanding relaxational processes (e.g. often associated with the 'central peak' in inelastic scattering).

To exploit the unique features of LCLS, we propose to develop a beamline with the capability for two types of XPCS techniques:

- Using the very high time-averaged coherent x-ray flux from the LCLS to carry out XPCS measurements over time scales from 10^{-3} to 10^{3} seconds by recording a sequence of speckle patterns ('Sequential' technique);
- Using the extremely high-peak coherent x-ray flux from the LCLS to carry out XPCS measurements over time scales from 10^{-12} to 10^{-6} seconds using a 'Split-Pulse' technique.

Sequential Technique: Important aspects of the microscopic dynamics of systems such as long-chain polymers occur on relatively slow time scales, e.g. longer than 10^{-2} s. This means that it will be possible to employ the very high time-averaged coherent x-ray flux from the LCLS, averaged over the 8 ms repetition rate, to investigate the dynamics using XPCS data collection and analysis techniques that are similar to those used now. Such an experiment consists of collecting a sequence of speckle patterns on an area detector. From an analysis of these sequences, correlation times from a few repetition times up to many minutes can be measured. The advantage of LCLS will be in higher signal rates than currently available. For example, a preliminary study of dynamics in a binary blend of poly(ethylene oxide) and poly(methyl methacrylate) found that XPCS measurements of this system at the Advanced Photon Source were not feasible because of the small scattering cross sections. The high average brilliance of LCLS will be required for XPCS studies of many systems of interest.

Split Pulse Technique: The short pulse duration at LCLS will allow extension of XPCS studies to much faster time scales than currently possible. For example, to understand the structural nature of the dynamics in glass-forming systems at the nanoscale, it will be important to carry out studies spanning a very large range of time scales (10^{-12} to 10^{3} s) in order to observe the evolution of the dynamics from liquid to glassy behavior as the temperature is lowered. In order to probe time scales between 10^{-12} and 10^{-6} s, we propose to develop a Split-Pulse technique, taking advantage of the instantaneous brilliance of the LCLS. The concept of the technique is to split each x-ray pulse into two equal-intensity pulses separated in time, but propagating along the same path. The scattering from the two pulses will then be collected during the same exposure of an area detector. If the sample is static on the time scale of the two pulses, then the contrast in the summed speckle pattern will be the same as that from a single pulse. If the sample evolves on this time scale, then the summed speckle pattern will have lower contrast. Thus by analyzing a set of such patterns, each for a different time delay, the correlation times of the system can be measured, on time scales down to the pulse duration. A pulse splitter with a path length difference variable from 3×10^{-4} to 3 m would give delay times from about 10^{-12} to 10^{-8} seconds. Longer time delays between pulses may be possible using two electron bunches in the free electron laser, assuming reasonable reproducibility between bunches. With the planned pulse structure of the LCLS, there may be a gap between about 10^{-6} and 10^{-3} s in the time scales accessible by the Sequential and Split-Pulse techniques.

Heating of the sample by the x-ray beam: The pulsed structure of the LCLS presents a significant issue for the design of a beamline optimized for XPCS measurements, because of the potential for heating of the sample by the beam. For dynamics studies, one would like to avoid heating the sample by more than a few degrees during the measurement. This is an important issue not just for XPCS but also for any application of a fourth-generation source as a non- or weakly interacting probe. Based on a simple analysis presented in [3], it appears that XPCS studies in a weakly interacting regime will be feasible, but will require careful choice of x-ray energy and flux depending upon sample absorption and scattering characteristics. In particular, the ability to use the 3rd harmonic of the source at energies up to 24 keV will be advantageous. One area of work we propose to carry out is to better understand the threshold for beam heating of the sample, and its implications on beamline and detector design (e.g. x-ray energy, focusing, and expected signal levels).

Figure 2 shows a comparison of three quantities: N_{MIN} , the minimum required number of photons per pulse to give sufficient signal per speckle; N_{MAX} , the maximum tolerable photons per pulse to avoid sample disturbance; and N_{AVAIL}, the photons per pulse available from the LCLS.

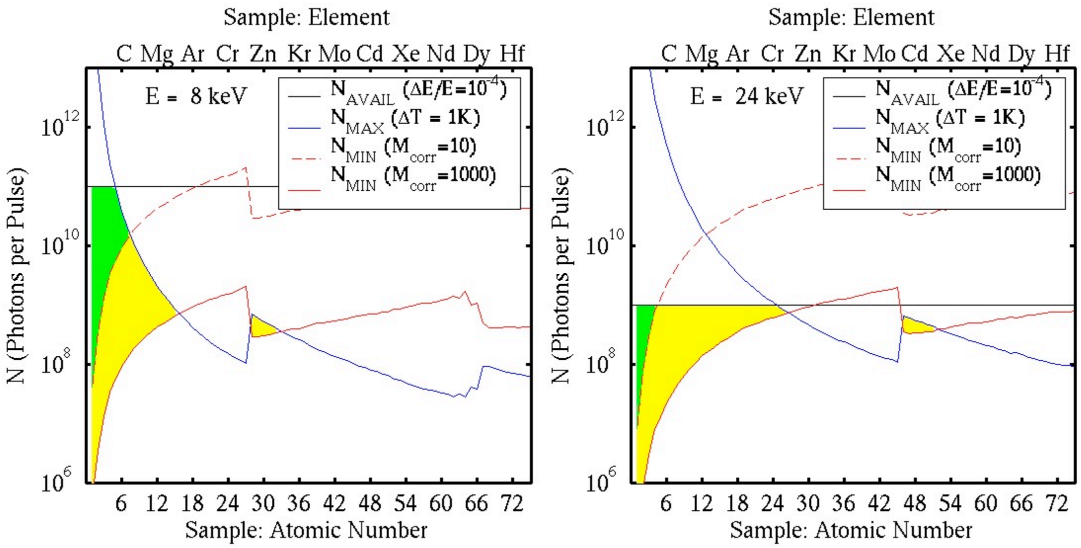

FIG. 2: Comparison of calculated number of photons per pulse available from the LCLS N_{AVAIL}, that tolerable due to beam heating of the sample N_{MAX}, and that required for XPCS N_{MIN} from two types of samples, corresponding to a typical liquid or glass (M_{corr} = 10) or to nanoscale clusters in a polymer blend (M_{corr} = 1000), as a function of sample composition, at 8 and 24 keV photon energies. Experiments are feasible in the shaded regions.

These are plotted as a function of sample composition for two energies, using $\Delta T_{MAX} = 1K$, a beam area of 10^{-4} cm^2, and two types of samples. The calculation predicts that XPCS will be feasible at the LCLS when the value of N_{MIN} is lower than both N_{MAX} and N_{AVAIL} (shaded areas). The result depends strongly on sample composition and photon energy. Because of the order-of-magnitude uncertainty in several quantities, these calculations do not provide exact limits, but do indicate that feasibility will depend upon photon energy, sample composition, and scattering power.

The threshold calculated here for adiabatic sample heating of 1K is a more stringent damage criterion than typical radiation damage thresholds found at third-generation x-ray sources for “soft” materials. For example, an x-ray damage threshold of 2000 kGy has been found for polymers in XPCS studies by Dierker, which corresponds to about $5\text{x}10^{13}$ photons per 10^{-4} cm^2 at 8 keV. The thresholds given in Fig. 2 are significantly lower than this value. Thus the unprecedented x-ray flux per pulse from the LCLS introduces a new facet to radiation damage issues.

Conceptual Design Summary and Expected Performance:

The proposed experiments require preservation of the transverse coherence of the beam, and thus minimal optics that could degrade collimation. Ideally no mirrors would be used to steer the beam to the experimental station (i.e. the endstation should be on the undulator centerline). Experiments will typically require monochromatic radiation (e.g. using a silicon (220) reflection, or diamond (111) if perfect crystals are available) to provide adequate temporal coherence. Minimization of sample heating will require energy tunability as well as the option of operation at the highest possible energies (e.g., using the third harmonic at 24 keV). The balance between maximizing signal and avoiding sample heating will require some flexibility in focusing options, from unfocused to mildly focused (e.g. 1:1 imaging of source). The Split-Pulse technique will involve a pulse-splitter optical element with variable delay, or in other cases the ability to provide two pulses with variable delay from the source. An x-ray sensitive streak camera with appropriate time resolution will be needed to set up and monitor the split pulses. Several sets of beam definition slits to provide clean coherent beams of adjustable size will be required. The sample chamber will have three rotation/tilt axes and high-resolution translations, with sample temperature control. Sufficient room in the experimental area is required for a long distance (5-10 m) between the sample and detector. Vibration and temperature control of the experimental station will be required for stability.

The major component of instrumentation will be an x-ray sensitive area detector system optimized for XPCS measurements. Some of the requirements for this detector are likely to be different from those developed for other experiments. Although development of detailed detector specifications is one of the initial activities to be carried out as part of this proposal, a possible scenario can be summarized here. The practical upper limit on the sample-to-detector distance fixes the maximum usable coherence length (and thus incident beam size and required area detector resolution). For example, to resolve the speckle pattern at 10 m the beam size must be less than 39 or 23 microns at 8 or 24 keV, respectively. This relatively high spatial resolution requirement (e.g. 25 microns) in turn leads to a large number of pixels (e.g. 100 million or more) required to cover sufficient solid angle. The average signal level per pixel will typically be small (e.g. 10^{-2} counts per pulse). For the Split-Pulse technique, in order to determine the contrast with sufficient accuracy, it will be necessary to have very low error rates in distinguishing pixels with zero, one, two, and more counts.

The proposed beamline will provide the best XPCS performance in the world in both the millisecond-to-kilosecond and sub-picosecond-to-microsecond regimes. The time-averaged brilliance of LCLS will be 2 to 4 orders of magnitude higher than at current x-ray sources, allowing XPCS studies of phenomena on 10 ms and longer time scales that are not currently feasible. The peak brilliance of LCLS will allow sufficient signal from single pulses to enable XPCS studies in split pulse mode down to the sub-picosecond time scale, six orders of magnitude beyond the current limit.

The optimization of the beamline to produce conditions in which the sample is not affected by the x-ray beam will be important for other types of x-ray studies (non-XPCS) that take advantage of the unique characteristics of LCLS. These include laser pump / x-ray probe experiments using the short pulse nature for time-resolved studies, as well as experiments using coherent x-ray beams, e.g. to reconstruct images of nanostructures without destroying them. The large area, high resolution detector will also be optimal for many coherent diffraction experments. The streak camera will be ideal for studying the dynamics of ultrafast phase transitions by time resolved conventional diffraction techniques, such as the metal-insulator transition in VO_2 [5] or structural changes caused by electrical stimulation in ferroelectrics and piezoelectrics.

Roles and Responsibilities:

The team members on this proposal are playing a leadership role in developing XPCS techniques and have significant experience in beamline design. We have been involved in the development of coherent x-ray techniques since the initial x-ray speckle measurements at NSLS [6] and throughout the exploration of XPCS capabilities and development of facilities at 3rd-generation x-ray sources. Team members are co-authors on most of the major publications using XPCS (see e.g. [7-22]). We bring the resources of several national laboratories with hard x-ray synchrotron facilities, the X-ray FEL project at DESY, as well as university-based coherent x-ray research and education programs.

All team members will participate with LCLS staff in developing an XPCS user facility for the scientific community. We will help set goals for capabilities and provide specifications and consultation for beamline instrumentation that will be designed by LCLS, such as monochromators, focusing optics, and slits. The team will participate in facility commissioning and early experiments.

In addition to this overall role, team members will lead subgroups to develop specific hardware and techniques in coordination with LCLS staff, as described in Subtasks A-G below. Several components of the required instrumentation will involve significant development and testing of unique systems. These include the area detector, streak camera, pulse splitter, and sample chamber. In order to determine certain hardware specifications and develop the required new aspects of XPCS technique in anticipation of the first experiments at LCLS, team members will also carry out studies of pulsed x-ray beam heating of samples, the effects of focusing optics on XPCS measurements, and the principles of the proposed Split-Pulse XPCS technique and associated data analysis.

Subtask A. Area detector: The XPCS program at LCLS will need an optimized area detector array with special characteristics (e.g. 100 million pixels, 25 micron resolution, 8 or 24 keV, 120Hz, low signal, extremely low noise). We will coordinate an effort with LCLS staff to determine the required specifications for this detector system, consult detector experts to determine the best solution(s), and carry out appropriate aspects of system acquisition (e.g. build or buy an initial prototype, test the prototype, determine specifications for the final system, commission the final system). Team members bring specific expertise in detector development [23] and access to the expertise of the Brookhaven detector group. Responsibility: Dierker and Mochrie.

Subtask B. X-ray streak camera: Kieffer and his group have developed an ultra-fast x-ray streak camera [24] that has a measured time resolution of better than 350 fs, making it the fastest x-ray streak camera in existence. The spatial resolution is less than 40 microns. Currently under development is a new streak camera that is expected to have a time resolution better than 100 fs. We plan to carry out tests of this design and then coordinate with LCLS staff to build x-ray streak camera(s) optimized for LCLS. A streak camera will be extremely useful as a diagnostic tool to study the beam stability to insure that we will have the high Q resolution needed for XPCS experiments, to study and monitor timing issues with the beam splitter/delay instrumentation, and to measure pulse widths to characterize effects of high instantaneous x-ray fluxes on samples. It will also be very useful as the primary detector in time-resolved measurements. Responsibility: Sutton and Kieffer.

Subtask C. X-ray beam splitter / delay: A critical component of the Split-Pulse mode for XPCS measurements will be a pulse split-and-delay instrument capable of preserving the transverse coherence between the two sub-pulses. We will participate with LCLS staff in building a prototype of this instrument and testing it at APS and SPPS. Responsibility: Fuoss and Stephenson.

Subtask D. Sample chamber: A sample chamber will be required for XPCS and other types of diffraction experiments to be carried out at the endstation. We will design, construct, and test the sample chamber in coordination with LCLS staff. Responsibility: Brennan and Fuoss.

Subtask E. Understanding effects of x-rays on sample: Experimental determinations of the nature of sample heating and other interactions from fast x-ray pulses will be important for designing XPCS and other LCLS instrumentation. We will carry out measurements (e.g. with focused beam at SPPS) and analyze the results to establish sample heating limits and optimum instrumentation specifications for both Split-Pulse and Sequential XPCS experiments. Responsibility: Stephenson and Dufresne.

Subtask F. Understanding effects of focusing on XPCS measurements: We expect that different amounts of focusing will be required to obtain optimal signal levels from XPCS measurements at LCLS, to match incident beam coherence length and flux density to requirements for specific samples. We propose to determine the optimum focusing requirements and trade-offs for the LCLS facility through measurements at APS. Responsibility: Lurio and Sutton.

Subtask G. XPCS Split-Pulse technique and analysis development: In order to determine specifications for the final area detector and prepare analysis techniques for Split-Pulse XPCS experiments at LCLS, we propose to understand the requirements for the Split-Pulse technique using measurements at much slower time scales at APS. For example, methods will be developed to accurately discriminate the number of photons in each pixel and to extract correlation functions from the decay of contrast, in real time. Responsibility: Mochrie and Lurio.

Team Organization

Organization of the multi-institutional team will involve central coordination and reporting through the spokesperson (Stephenson), defined sub-tasks with subgroup leaders as described above, regular meetings both of subgroups and the full team, and integration of subgroups with appropriate LCLS project staff. Schedules and milestones will be established and progress reported. Construction management, procurements of beamline components, and formal DOE MIE project reporting will be handled by LCLS.

Preliminary Budget:

The proposed funding source for this endstation is DOE Basic Energy Sciences.

This budget is based on a five-year proposal schedule, three years before initial operations (FY06-8) and two years of commissioning and early experiments (FY09-10).

These cost estimates are inclusive of benefits, overhead, etc.

	Cost Estimate ($K)
Personnel	
4 Postdocs (Subtasks B,C,E,G) for 3 years (FY06-08) at $100K/year	1,200
2 Graduate students (Subtasks F,G) for 4 years (FY06-09) at $40K/year	320
2 Postdocs at LCLS commissioning for 2 years (FY09-10) at $100K/year	400
Staff/Professor effort for supervision, summer salary, sabbaticals at LCLS, 0.5 FTE/yr for 5 years at $200K/year	500

Subtotal	2,420
Travel	
11 PI's for 5 years at 4 trips/year, $1K/trip	220
Materials and Supplies	
10% of effort cost	242
Permanent Equipment	
Monochromator	300
Focusing Optics	200
Slit Systems	150
Sample Chamber	300
Pulse Splitter	1,000
Area Detector	4,000
Streak Camera	300
Laser Optics	200
Electronics, Control	200

Subtotal	6,650
Total	9,532

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