BEAMLINE 8-ID

UPGRADE PROPOSAL:

EXTRAORDINARY NEW TIME-RESOLVED CAPABILITIES FOR XPCS AND GISAXS

1. Science

Beamline 8-ID at the Advanced Photon Source (APS) has 2 complementary missions:

1. Develop and apply x-ray photon correlation spectroscopy (XPCS) to the study of nanoscale fluctuations in condensed matter.
2. Develop and apply grazing-incidence small-angle x-ray scattering (GISAXS) to the study of the mesoscale structural properties of surfaces and buried interfaces.

Both techniques, as performed at beamline 8-ID, have had extraordinary scientific impact in recent years. Our upgrade proposal, therefore, is based on significantly extending the scientific reach of each technique without compromising the other. The overall thrust of our requested upgrades is to enable pioneering time-resolved scientific work by extending state-of-the-art multispeckle XPCS-accessible timescales from 10's of milliseconds to microseconds and grazing-incidence small and wide-angle x-ray scattering time scales from several seconds to milliseconds. Among other new capabilities, these major additions to 8-ID's experimental capabilities will allow GISAXS and XPCS to probe nanoscale kinetic and dynamic regimes, respectively, of matter far from equilibrium—a "Grand Challenge" as eloquently described in Ref. [1]. A sampling of the science that the 8-ID upgrade program will enable is described below.

XPCS characterizes the fluctuation dynamics of condensed matter on length scales smaller than can be achieved with optical techniques and on longer time scales than can be achieved via neutron scattering. Despite these idealized advantages, state-of-the-art multispeckle XPCS can only access time scales ≈ 3–4 decades slower than the slowest dynamic time scales measurable via neutron scattering and can not directly access fluctuation length scales less than 10's of nanometers. Our upgrade proposal for XPCS aims to expand these boundaries by extending the range of samples that can be studied over time scales extending from the slowest neutron spin echo measurements [$O(10^{-6}$ s)] to near static [$O(10^{3}$ s)] conditions.

Selected examples of XPCS experiments that will be possible after 8-ID is upgraded but which are impossible or extremely difficult at current facilities include the following examples.

1) Studies of fluctuations about non-equilibrium and far from equilibrium conditions. XPCS is well suited to elucidate the relevant dynamical properties of representative out-of-equilibrium systems. Specific examples include fast localized motion and structural arrest in glasses and gel-forming colloidal suspensions and the nature of spatially and temporally heterogeneous dynamics in nearly jammed systems and driven systems with disorder. But the 8-ID upgrades described in this document are necessary because of the extraordinarily high data quality and extended time scales that are required. Such studies also very naturally complement the kinetic capabilities that will be developed as part of upgrading 8-ID's GISAXS capabilities (see below).
2) Studies of transition metal oxides and other strongly correlated systems that tend to form charge-, spin- and orbital-ordered domains. The expanded capabilities in coherent flux, focusing and large wave-vector transfer XPCS at an upgraded 8-ID will permit resolution of domain wall motions on the order of 10 nm or below and will provide unique insight into self-organization of various types of domains in complex oxides and other correlated materials, shedding light on the relationship between charge, spin, orbital and lattice degrees of freedom. Aside from the basic mesoscale physics that such studies will reveal,

detailed understanding of this class of materials will impact vitally important technology industries such as data storage and power transmission.

3) Studies of dynamics within complex mesoscopically-structured fluids and in biological systems. For example, the enhanced brightness resulting from APS and 8-ID upgrades will permit spatially and temporally resolved measurements of nanoparticles confined within copolymer sub-phases or on the surface of amphiphiles. Such inquiries will be critical in understanding the processes underlying how small particles self-assemble into potentially technologically useful structures. In addition, the higher x-ray-energy operations that will be enabled by the upgrade will facilitate measurements of the dynamics of biologically-relevant materials by allowing x-rays to penetrate aqueous solutions and by minimizing beam damage. Important problems that will be investigated include the dynamics of solid supported membranes, diffusion and surface fluctuations in free floating vesicles, and Brownian motion within concentrated protein solutions. Such studies will provide crucial insight into the central role of thermal fluctuations in biological activity at the cellular scale.

The critical need to understand and ultimately control examples such as these (and the GISAXS examples described below) is strikingly described in both the recent Grand Challenge report [1] and the provocative document by Laughlin *et al.* entitled “The middle way” [2].

GISAXS applies the power and utility of SAXS to study the structural properties of surfaces and buried interfaces. A dedicated GISAXS instrument was recently designed and commissioned at beamline 8-ID. Despite the new instrument’s unrivaled output in the nanoscience research field, it has several limitations that will be eliminated under the proposed upgrades, namely fixed, relatively low x-ray-energy operations and limited ability to study kinetic behavior [time resolution > $O(10\ \text{s})$]. With greatly increased flux, energy tunability and time resolution, a sampling of the science that will be achievable is described below.

1) Self-assembly is a powerful emerging method for fabricating highly ordered nano-structured materials over macroscopic length scales with unprecedented electronic, magnetic or photonic properties. Recent research has revealed that self-assembly facilitated by epitaxy and annealing (solvent or thermal) yields materials with novel hierarchical order [3, 4]. Microscopic structural understanding of the kinetics of such highly non-equilibrium processes is, however, seriously lacking. GISAXS is a powerful technique for studying such kinetic processes especially when they occur *in situ*, in real-time, or at buried interfaces where conventional techniques such as scanning probe microscopy are not readily applicable. Upgraded capabilities at 8-ID will permit real-time measurements of self-assembly processes and ultimately lead to better organizational control of nanoscale building blocks.
2) Dispersions of quantum dots in polymer matrices are of basic and technological interest. Technologically, their unique optical properties garner much attention while fundamentally they comprise a novel system to probe molecular dynamics at surfaces and interfaces. Although the dynamics at micron length scales is largely understood, such information is missing at nanometer length scales. But it is precisely the kinetics and fluctuations at the nanoscale that must be understood and controlled if the technological promise of such materials is to be realized. Recent research performed at 8-ID [5, 6] has established the synergy between grazing-incidence measurements of the kinetics and dynamics in such nanocomposite systems that are fluctuating while slowly evolving

towards equilibrium. Upgrades to 8-ID will provide enough brilliance and flux to probe such phenomena at far smaller length scales and far faster time scales.

2. Added value of the medium term upgrade

We propose to significantly upgrade both the XPCS and GISAXS facilities hosted at 8-ID. We will increase the range of time scales accessible to XPCS by three orders of magnitude and extend XPCS to higher energies where sample radiation damage is diminished. The upgrade will also enable new capabilities in the hitherto under-exploited area of large wavevector transfer XPCS. For GISAXS, the upgraded facility will provide increased intensity by a factor of 20. Also, importantly, the GISAXS upgrade will enable energy tunability over a broad energy spectrum from 6–30 keV. The combination of these improvements will provide increased spatial resolution, interrogation of both submerged and buried interfaces, complementary fluorescence and anomalous scattering measurements and greatly increased time resolution for kinetics studies.

Finally, we discuss how this upgrade positions 8-ID for the future. The proposed 8-ID XPCS upgrade increases the brilliance delivered to the experiment so that it is comparable to that which is promised by a competing facility under development at NSLS-II. But the APS will be unique in being able to extend operations to higher x-ray energies to reduce sample beam damage. Also, with respect to XPCS at future free-electron lasers (FEL's), an upgraded 8-ID will permit sample dynamics to be studied at the nanoscale in the physically important time scale spanning μs to ms—a time scale inaccessible to FEL's because of their low repetition rates and practical limits on pulse delay lines. With respect to the GISAXS facility, our GISAXS capabilities are unique today; the upgrade will provide extraordinary new capabilities to ensure our continued pre-eminence. Table 1 summarizes the expected gains that will be achieved by the 8-ID upgrade.

Upgrade	**XPCS**	**GISAXS**	**Comment**
I_{ring} = 200 mA	2× coherent flux	2× flux	
8 m straight with canted ID's			See next 3 items
5-m 3.3-cm-period planar ID	2.6× coherent flux	N/A	
2.5-m 3.3-cm-period planar ID	N/A	10× flux	Enlarged upstream "pinhole"
Front end upgrade			To support canted ID's
First optics enclosure optics	Diffraction limited side-bounce mirror	Diffraction limited side-bounce mirror	Also dual "pinhole" and beam diagnostics
Large horizontal offset mono	N/A	X-ray energy tunable from 6–30 keV	X-ray energy is fixed at 7.35 keV today
Pilatus detector	N/A	> 1,000× increased time resolution	Custom tiling for SAXS/WAXS data collection
End station multilayer mono and vertical focusing	10–30× coherent flux	N/A	
XPCS optimized detector	1,000× increased time resolution and 10–100× data collection efficiency	N/A	≥ 1 MHz frame rate, 100% efficiency, firmware correlation, small pixels
Ancillary focusing optics	10× increased flux	100× increased flux	2-D GISAXS, 1-D XPCS
Total estimated enhancement factors for an individual experiment	• **50× increased coherent flux** • **Minimum delay times > 1,000× faster**	• **20× increased tunable flux** • **Time resolution 1,000× more** • **Tunable energy**	**Excludes additional gains from horizontal (XPCS) and 2-D (GISAXS) focusing and wide bandpass (XPCS) multilayer mono operation**

Table 1. Enhancement factors for XPCS and GISAXS based on the proposed APS and 8-ID upgrades.

3. Expected user communities

Beamline 8-ID has a large and growing base of General Users from the fields of biophysics, chemical engineering, geophysics, materials science and engineering, nuclear engineering, physics and polymer science and engineering. Our new capabilities developed under the auspices of this proposal will further grow our diverse user community.

4. Enabling technology and infrastructure

The 8-ID upgrade consists of 4 major projects:

Project 1. GISAXS energy tunability and millisecond time resolution
Project 2. XPCS coherent flux maximization
Project 3. 8-ID canted insertion devices (ID's) in an 8 m straight section
Project 4. XPCS detector

The overall idea with respect to Projects 1, 2 and 3 is to extend 8-ID’s straight section, add a canted ID and then add a variety of optics so that the XPCS and GISAXS experiments each acquire full operational flexibility—unlike today where the shared undulator beam severely restricts the choice of operating energies and focusing options. The most challenging part of our proposal (and therefore the item with the greatest cost, cost uncertainties and required contingencies) is Project 4: obtaining a suitable XPCS detector to leverage the beamline upgrades described. The 5 essential features of such a detector are: i) large number of pixels, ii) high speed (≥ 1 MHz), iii) high x-ray efficiency, iv) onboard correlations and v) pixels small enough to resolve speckle. It is outside the scope of this proposal to determine how such a detector should be developed or obtained but it is critical to note that there exist sophisticated proposals originating from Brookhaven National Laboratory and Cornell University to build such detectors. Also, with respect to future XPCS detectors, we point out that existing detector programs within Argonne such as the ANL-Lawrence Berkeley Laboratory Fast CCD project can achieve many of these goals with additional support and staffing. For GISAXS, the relevant detector technology already exists within the framework of the Pilatus detector program hosted by the Swiss Light Source. Custom tilings will be required, however, to support the need to simultaneously collect data at large and small scattering angles. Finally, with respect to required infrastructure, we emphasize the need for sophisticated developments in data acquisition and in scientific visualization and analysis software to ensure productive and efficient access to 8-ID’s resources for the widest possible community of researchers.

5. Partnerships and user interest

Our proposal has been circulated to the 8-ID beamline advisory group (BAG). We have also shared this document with a subset of beamline 8-ID’s regular principal investigators and they strongly support the proposed scientific directions and upgrades described herein. The BAG members and General Users to which this document has been circulated to are listed in Appendix C. We expect these scientists to write or endorse letters of support as required.

6. Industry and technology transfer

XPCS detector development will leverage and extend the capabilities of relevant technology companies in the chip design, the chip fabrication and the high-speed imaging industries.

Appendix A: Estimated budget

The tables below provide our current best estimates of the capital costs associated with the major 8-ID beamline and detector upgrades described in this document. Table 2 is a detailed list of the items that are required to complete the upgrade while Table 3 aggregates the numbers presented in Table 2 and assigns them to logical project units, each of which individually would provide major new scientific capabilities to 8-ID. Many of the items listed in this table, excluding detectors, are relatively standard beamline components or correspond to upgrades that have been contemplated in detail before by various APS entities. As such, the costs of many of the listed items such as monochromators, mirrors and support assemblies are relatively well known and project risks are correspondingly small. The greatest cost uncertainty and project risk corresponds to the required detectors, most especially the ambitious XPCS detector that is required. Cost estimates for this are based on existing funding proposals for analogous detectors.

Item	**Capital Budget (k$)**	**8-ID Project ID**	**Comment**
GISAXS large horizontal offset double crystal monochromator	860	1	
GISAXS downstream station extension (2 × 2 m^2)	113	1	
GISAXS monochomator station extension (2 × 2 m^2)	113	1	No PSS required
GISAXS vertical and horizontal focusing optics	100	1	
Precision GISAXS support assemblies	200	1	
GISAXS time-resolved Pilatus-like detector (9 modules)	1,408	1	9 modules
XPCS vertical focusing assembly	640	2	
XPCS multilayer monochromator	200	2	
XPCS SOE shutter	45	2	
XPCS secondary optics enclosure (SOE)	397	2	PSS required
XPCS SOE mini-hutch	57	2	No PSS required
XPCS enclosure extension	170	2	
XPCS horizontal focusing	75	2	
XPCS precision Motorized Supports	200	3	
8-ID 8 m straight	450	3	
8-ID FE upgrade	500	3	
8-ID canting magnets, BPM, …	100	3	
8-ID 5 m insertion device	720	3	Assume UA used for GISAXS and 2× phased UA's used for XPCS
First optics enclosure (FOE) diffraction limited mirrors	240	3	
FOE dual pinhole	40	3	
XPCS Detector	3,250	4	
TOTAL:	**9,878**		

Table 2. Estimated budget for proposed 8-ID upgrades.

8-ID Project ID	**Project Name**	**Capital Budget (k$)**
1	GISAXS energy tunability and millisecond time resolution	2,794
2	XPCS coherent flux maximization	1,583
3	8-ID canted ID's in an 8 m straight section	2,250
4	XPCS detector	3,250
	PROJECT TOTALS:	**9,878**

Table 3. Estimated budget by project for 8-ID upgrades.

Appendix B: References

[1] *Directing Matter and Energy: Five Challenges for Science and the Imagination*, Basic Energy Sciences Advisory Committee (BESAC) Report, December 20, 2007, (http://www.sc.doe.gov/bes/reports/abstracts.html#GC).

[2] *The middle way*, R. B. Laughlin, David Pines, Joerg Schmalian, Branko P. Stojkovic, and Peter Wolynes, PNAS **97**, 32 (2000).

[3] *Self-directed self-assembly of nanoparticle/copolymer mixtures*, Yao Lin, Alexander Boker, Jinbo He, Kevin Sill, Hongqi Xiang, Clarissa Abetz, Xuefa Li, Jin Wang, Todd Emrick, Su Long, Qian Wang, Anna Balazs, Thomas P. Russell, Nature **434**, 55 (2005).

[4] *Epitaxial self-assembly of block copolymers on lithographically defined nanopatterned substrates*, Sang Ouk Kim, Harun H. Solak, Mark P. Stoykovich, Nicola J. Ferrier, Juan J. de Pablo, Paul F. Nealey, Nature **424**, 411 (2003).

[5] *Real-Time Evolution of the Distribution of Nanoparticles in an Ultrathin-Polymer-Film-Based Waveguide*, Suresh Narayanan, Dong Ryeol Lee, Rodney S. Guico, Sunil K. Sinha, Jin Wang, Phys. Rev. Lett. **94**, 145504 (2005).

[6] *Particle Dynamics in Polymer-Metal Nanocomposite Thin Films on Nanometer-Length Scales*, Suresh Narayanan, Dong Ryeol Lee, Aleta Hagman, Xuefa Li, Jin Wang, Phys. Rev. Lett. **98**, 185506 (2007).

Appendix C: User Support

Jeff Brinker	*Professor of Chemical & Nuclear Engineering, University of New Mexico and Senior Scientist, Sandia National Laboratory*
Mark Foster	Professor and Department Chair, College of Polymer Science and Polymer Engineering, University of Akron
James Harden	Associate Professor of Physics, University of Ottawa
Edward Kramer	*Professor of Materials Science, University of California-Santa Barbara*
Jyotsana Lal	Physicist, Argonne National Laboratory
Robert Leheny	*Associate Professor of Physics, Johns Hopkins University*
Karl Ludwig	Professor of Physics, Boston University
Laurence Lurio	*Associate Professor of Physics, Northern Illinois University*
Simon Mochrie	*Professor of Applied Physics, Yale University*
Oleg Shpyrko	Assistant Professor of Physics, University of California-San Diego
Sunil Sinha	Professor of Physics, University of California-San Diego
Mark Sutton	*Professor of Physics, McGill University*
Pappannan Thiyagarajan	Senior Physicist, Argonne National Laboratory
Ting Xu	*Assistant Professor of Materials Science and Engineering, UCB*

Table 4. 8-ID principal investigators to whom the proposed 8-ID upgrade plans have been circulated to. Italicized entries denote members of 8-ID's beamline advisory group (BAG).