Nuclear Resonance Spectroscopy at the APS in 5 years

1. Past and present

Nuclear Resonance Spectroscopy (NRS) has been practiced at the APS from the very beginning in 1997, mainly at Sector 3-ID beamline, and since 2002 at Sector 16. NRS consists of two separate but related methods: Nuclear Forward Scattering (NFS) and Nuclear Resonant Inelastic X-Ray Scattering (NRIXS). The first method is also known as Synchrotron Mössbauer Spectroscopy (SMS) and latter as Nuclear Resonant Vibrational Spectroscopy (NRVS).

NFS or SMS is the time domain equivalent of traditional Mössbauer spectroscopy. NRIXS or NRVS is a new type of phonon spectroscopy and it is related to inelastic x-ray or neutron scattering, as well as to Brillouin and Raman Spectroscopy in terms of its information content, with some unique and distinct advantages in terms of momentum transfer range covering entire Brillouin zone, isotopic and element selectivity, selection rules that do not require dipole moment formation, and sensitivity both in terms amount of material (nanograms to pico-grams, monolayers), and form (transparent or opaque glass, crystal, nano-cluster…).

Both of these techniques require a specific bunch structure, which is easily achievable at the APS with a large circumference of 1108 m. Currently, NRS technique at the APS is limited to five isotopes, namely ^{83}Kr, ^{57}Fe, ^{151}Eu, ^{119}Sn, and ^{161}Dy. We can project that it can be extended to include yet another six isotopes, namely Ta, Tm, Sm, I, Sb and Ni, in the next five years, provided that more beamlines become interested in practicing this method. There is no special beamline configuration for this method, and therefore, any beamline equipped with high-resolution monochromators can use a limited set of nuclear resonances given above.

In order to project the future, one has to look at the successes and shortcomings of the last decade, and appraise the progress relative to the other major research centers. Currently, there are six operating beamlines worldwide (2 ESRF, 2 APS, and 2 Spring-8) and there are plans to build a very competitive beamline at PETRA III. The major scientific accomplishment has been the introduction and further development of a new vibrational spectroscopy to physics, chemistry, geology and biology. This application has been so successful that it dwarfed all other areas, generating over 170 proposals in the last 5 years, resulting in a beamtime request over three times the capacity, reaching over seven times the available beamtime in 2008-3 cycle. The rich information content of the phonon measurements includes the phonon density of states, Debye sound velocity, vibrational entropy, mode-specific Grüneisen constant, direct access to amplitude of vibrations of all the atoms involved in the molecule or solid with the help of modeling, Debye temperature and its temperature dependence, and finally actual temperature of the sample under extreme conditions of pressure. Furthermore, coupled with modern microfocusing optics, the method has found a unique niche in geophysics and mineral physics by engaging in high-pressure applications. APS-developed data analysis software,

PHOENIX, has become the standard method of data analysis, further establishing our leadership in this field.

In biological applications, NRVS has now become a method of choice when it comes to protein and enzyme vibrational studies. The unusually selective nature of NRVS has led to a renewal of interest in the evaluation of the dynamics of model compounds of porhyrins, and cubanes through the help of modern DFT calculations.

Finally, in materials science, NFS and NRIXS has found a wide range of applications in terms of determination of valence and spin transitions in magnetism of thin films, oxides, sulfides, phosphates, silicates, carbides, nitrates and borides of iron. Again the data analysis software package CONUSS has evolved to become the recognized standard in data analysis of NFS spectra, uniquely able to handle thickness and field distributions frequently encountered in real situations, and applicable to all isotopes.

So what are the shortcomings? Clearly, the need to utilize all of the isotopes in one beamline where the energy ranges from 6-70 keV is a fundamental limitation. Undulators can be optimized over a limited range, and we had chosen 7-15 keV in the first harmonics, to facilitate 9.4 keV Kr, 14.4 keV Fe, 21.4 keV Eu, 23.3 keV Sn and 25.7 keV Dy, plus the 21.6 keV non-resonant IXS spectrometer for the non-timing mode application at 3-ID. This should change, and the following scenario is very realistic:

Move Sn, and Dy program to Sector 30-ID, and use the existing cryo-cooled mono infrastructure and add a second energy choice, which the IXS program also benefits. Move the MERIX program to Sector 9 for full time MERIX beamline. This is consisten with the high energy IXS spectrometer which is designed to work at 1 meV at 25.7 keV, which is not feasible now because the compromised undulator period. If the undulator period is chosen such that 21-26 keV region is in the first harmonics, the flux can be boosted by a factor of 5, and that will then provide an extremely competitive beamline for physics, mineralogy, and materials science.

There are equally important, but less obvious side to the success of the NRS program, and that is mainly in the instrumentation development. Before nuclear resonance studies begun at synchrotron radiation sources, there were no good methods of diagnostics for the perfection of silicon and other crystals, which are used in high-energy resolution crystal monochromators (HRM) at the level of 10^7 or better. With the availability of a nuclear resonant probe, whose energy width is in the range of nano-eV, we have pushed and broke many world records in the last decade, and APS has become the undisputed world-leader in terms of development of science, methodology and instrumentation aspects of high-resolution optics. Starting from 1991, we have developed 4 generations of HRM, starting with nested, channel-cut monochromators. We have developed and patented the weak-link rotary and translational stage technology, and built first “in-line, no-offset” HRM. After this, in an attempt to further improve the efficiency, we have developed the first cryogenically cooled HRM, and extended this into user programs at Sectors 3 and 30.

2. Future

Full realization of the potential of NRS method is limited by the number of beamlines it is practiced, lack of specialized or custom-made undulators and array APD detectors, and inability to develop methodology because of large demand from user programs.

2.1. Beamlines
2.2 Undulator sources
2.3 Methodology development
2.4 Detector development
2.5 Training and software for data analysis

2.1 Beamline

2.1.1 Expand the current NRS activities to Sector 30 for Eu, Sn and Dy, and develop Sm and Iodine there.
2.1.1 Enlarge Sector 3-ID-B station so that 2 new permanent experimental tables can be added, and cryogenic experiments can be perfomed, which is not possible now due to lack of space.
2.1.2 Target 0.1 meV resolution nuclear resonance spectroscopy for Fe as primary activity of this beamline,
2.1.3 Upgrade the 2 meV resolution IXS instrument from horizontal to vertical geometry with 30 analyzers and new detectors.
2.1.4 Add new specialized environmental chambers like “in-situ” deposition system for catalysis and nano-science applications, and a second-laser heatings system for liquids under high pressure at IXS instrument in 3-ID-C. There is demonstrated demand for both activities.

2.2 Undulator source

We propose to switch from the current planar permanent magnet technology to cryogenically-cooled in-vacuum undulator technology, to reduce the undulator magnetic period from the current 2.7 cm to 1.8 cm, and the current undulator gap from 10.5 mm to 7 mm. This itself will provide a factor of 7 gain in flux, with no extension of the current straight section. When coupled with possible increased stored current, extended straight section, we can foresee a gain of 30-50 in the next five years.

2.3 Methodology development

Currently we are involved in developing the cryogenically cooled weak-link technology to further reduce the energy resolution to 0.1 meV level. This current R&D works will have a significant impact on how we configure the beamline, and its scientific priorities.

We have learned how to work lasers, and with high-pressure equipment using DAC technology. We plan to extend the method to study liquids under pressure and this will require to work with multi-anvil apparatus, and external heating.

We would like to extend our work on determining melting point of alloys and compounds of iron, by incorporating more stable fiber-lasers, and in-situ pressure-measurements via a Raman system. Also, we would like to move to dynamicDAC as well as membrane cells for in-situ pressure adjustments. All of this will require subject-matter experts at the beamline.

2.4 Detectors

The detector of choice for NRS is Avalanche Photodiode Detectors. Currently we use single element detectors. However, we would like to have A desired set of specifications could be as follows:

Time resolution:	1 nsec
Area:	10 x 10 mm^2
pixel size:	0.3 x 0.3 mm^2
Thickness:	0.1-0.2 mm Si
Bump bonded ASIC,	
Integrated electronics (amplifier + threshold + time discriminator)	
Count rate:	10^7 Hz
Fast frame readout	< 10 ns

It turns out that such detectors are also needed for intensity fluctuation spectroscopy, thus we see a common ground with them. Similar detectors are being developed as European/Japanese collaboration, and either we take place in such development projects, or we end buying some version of the commercial detectors a few years later. We prefer that our detector group at the APS spearheads an effort and obtain/participate/develop APD based detectors.

2.4 Training and software

Our experience shows that the best way to increase productivity is to have the best personnel at the beamline during the measurement time. This includes beamline personnel as well as users. We believe that training users periodically as well as each time they visit the beamline will increase the productivity, but this will require one additional staff whose focus is getting users up to speed every week, and provide data analysis support during the experiment.

APS Renewal: Small angle x-ray scattering (SAXS)

SAXS has been a crucial technique for a wide variety of materials and in situ studies. With the benefits of high flux and well collimated source like APS, it is getting significant attentions from many fields such as Chemistry and Catalysis, Condensed matter and Material physics, Applied Science, Geological/Environmental Science, Life Sciences, and Surface/Interface/Thin film. Especially in the field of nanoscience, SAXS has been so critical and popular as SEM or TEM. It has been a tool to measure shape/size/density of nanoscale (1nm ~ 1um) objects in any condition (solution, solid, gas, high/low pressure and so on). It has been the most accurate size measurement technique in nanoscience even without destroying samples. Recently SAXS has added chemical sensitivity (ASAXS) and surface/thin film probe (GISAXS).

In five years SAXS capability of APS will be upgraded significantly in terms of available beamtime as well as its performance. While two dedicated beamlines for SAXS-related-techniques, Grazing Incidence Small Angle X-ray Scattering (GISAXS) and X-ray Photon Correlation Spectroscopy (XPCS), have been recently launched and been successful, but SAXS in general has suffered from severe oversubscription. The first dedicated SAXS stations at APS, one of which will be in full operation at 12ID within two years and the other of which serve about 40% for SAXS, and dedicated 5ID SAXS beamline in 7-10 years, will address increasing demands for this technique. Nevertheless having two productive SAXS instruments, USAXS and current SAXS at 12ID-C, at a single line and letting them compete would starve users. Performance of SAXS is significantly affected by stability of beam, collimation, flux, high dynamic range and low background detectors, energy range/resolution, sample environment, and analysis software. High flux and collimation distinguish SAXS beamlines at APS from those at the other facilities. However the other criteria have limited the full use of SAXS capability at APS and the upgrades being planned will address these issues. A dedicated beamline would provide better collimation, more stable beam, and optimized sample environment. Also high performance detectors, such as PILATUS, will provide major breakthroughs for SAXS at APS due to high dynamic range, no dark/readout noise, and fast readout (less than 10m seconds) which are the most critical components for SAXS.

Upgrade plans throughout APS beamlines can be categorized as more beamtime, wider q range (better collimation, SAXS/WAXD, or fast setup change), faster acquisition (down to milliseconds by improved flux and detector), and simultaneous measurement (chemical probe adds). This will cover needs for both efficient routine measurements and extreme frontier science. SAXS group at APS is noting the trend of combining techniques, which can help to overcome limit of SAXS by adding chemical probe or imaging capabilities. An impediment for becoming a leading characterization and analytical facility in nanoscience is the lack of an overall plan for SAXS at APS. For instance, a suite of data reduction and analytical software tools that are common to all SAXS at APS will boost user productivity and promote theoretical studies. Shared sample preparation laboratories and support for engineering of experimental apparatus could be also considered. There are discussions on these issues among SAXS groups and hopefully some types of outcome in five years.

Summary of future capabilities is

Solution SAXS: Solution SAXS for biological samples will be the largest user community benefitted from high performance low noise detector. Fast (greater than 100 Hz) and extremely low background

detectors will enable various real-time monitoring of biological phenomena at the nanoscale. Both SAXS beamlines, 12 and 18ID, will provide improved ASAXS capability that will enhance phase determination for solution SAXS. Significantly reduced sample volume, less than 20ul, will be required.

ASAXS (Anomalous SAXS): With a high resolution monochromator which will be available at 12ID and is designed for spectroscopic purpose, ASAXS could be done in the range of 7-40keV and EXAFS will be measured simultaneously. This upgrade will enable to study various high Z materials: Researches dealing with samples having multi-elements such as nanoparticle alloy, embedded particles, or composites will be benefitted. Chemical status of elements can be resolved after upgrade.

GISAXS: GISAXS has been of interest in self-assembly or growth of nanoparticles on or embedded in interfaces. Higher energy, up to 30~40 keV, will be available at 8ID and 12ID, which will allow studying nanoparticles/nanostructure formation under water and embedded under high Z materials. Tens of micro-second time resolution will be available with higher spatial resolution achieved by focused optics. In addition, GI-EXAFS and GISAXS may be carried out simultaneously at 12ID with mass-spectroscopy which is useful for in-situ catalysis studies, where for example GI-EXAFS can probe oxidation/reduction of nanoparticle catalysts and GI-SAXS determine their morphology at the same time.

USAXS (Ultra small angle x-ray scattering) will be served at 12ID with improved beam stability with faster scanning, 1-2 minutes (Currently 15min). 2D collimation will enable 2D USAXS, which is ideal to study anisotropic materials without any smearing. USAXS imaging will be available routinely.

XPCS : The upgrade plan for 8ID XPCS aims to expand its time scales from the slowest neutron spin echo measurements [$O(10^{-6}$s)] to near static [$O(10^{3}$ s)] conditions with at best 50 times of increased flux.

SAXS/WAXD: 5ID is planning dedicated beamline for simultaneous SAXS/WAXD in the 7-10 year vision. This capability with a PILATUS detector (no dark/readout noise and 3.8 ms readout time) would cover 0.002 to 2 $Å^{-1}$ seamlessly. High energy SAXS/WAXS also has been proposed at 1ID and 11ID, which will then provide inner atomic structure of nanoparticle from pair distribution function as well as shape/size from SAXS simultaneously.

High Energy SAXS: High energy SAXS has been available at 1ID and will be upgraded for faster timing (4 ms readout) with wider area of measurement (400*400 mm^2). In addition this capability is going to be added to 11-ID-B to operate in parallel with PDF measurements. This technique is essential to see embedded nanostructures such as void in high Z thick materials.

MicroSAXS: About 10um size x-ray beam for SAXS will be available at 12ID dedicated SAXS station with the help of a focusing optics. This will help to visualize locally located nanostructure such as self-assembly at interfaces or to better resolve individual cluster of nano-objects of less than 100um in size.

High throughput SAXS: 12ID-B dedicated station is planned to be high throughput SAXS beamline. It will have a detector in a vacuum flight tube, which will allow fast switch of sample-to-detector distance. Some portion of beamtime will be set aside for a rapid access.

Time-resolved SAXS: Down to tens of nanosecond time resolution could be reached with 12ID pink beam and fast detector such as annular Si-based detector. Tens of micro second time resolution will be available at all ID SAXS beamlines.

Status of the APS Time-resolved capabilities in 2013, Eric Dufresne XSD July 29, 2008

The APS is the best machine in the world to perform time-resolved science that uses the bunch structure of its ring. Its 24 bunch and hybrid mode are ideal, since one can isolate x-rays from a single bunch with gated detectors such as Avalanche Photodiodes (APD), Silicon Drifts Detectors and Pixel Array Detectors (PAD). This is ideal from laser-pump/x-ray probe experiments, where the laser can be synchronized to the source and delayed with respect to the x-rays.

In 5 years, the APS will be commissioning its new Superconducting RF crab cavity sector. This new capability will deliver a short one ps duration x-ray pulse having a 6.5 MHz repetition rate and about 1-2 x 10^{11} ph/s. This source will be tunable from the soft to the hard x-ray range and provide some polarization selection. The source will complement the LCLS, as it will provide a significant flux of tunable high-energy x-rays and bridge the time resolution gap between the XFEL (100 fs) and the synchrotrons (100 ps). Since it operates in all the modes of operations of the APS, this new source shall foster the growth of the Ultrafast X-ray community.

With its excellent coherence and brightness, experimenters will push the frontier of nanoscience with the studies of novel materials with 10-20 nm x-ray nanoprobes and 100 ps time resolution. Some of these studies will have demonstrated a few picosecond time resolution with an x-ray sensitive streak camera. Experimenters in the field of atomic physics, material science and chemistry have developed high-repetition rate lasers that best use the source, with some experiments pushing the repetition rate as high as 6.5 MHz. The repetition rate will be limited by the sample damage or heating from the laser source. New high-power Ti:Sapphire laser will have been installed on several beamlines and some groups are using novel fiber lasers, or THz pulsed lasers in their experiments. Fs-laser pulse shaping has been demonstrated in coherent-control experiments.

Ultrafast imaging in full-field mode is now routinely observing irreversible processes with sub-microsecond resolution using the latest CMOS high-speed camera and long specialized undulators. In its 324 bunch mode, with suitable detectors, it could even push the study of irreversible processes to tens of nanoseconds. New communities will gather around these new tools to elucidate non-equilibrium phenomena. Note that in periodic processes, 32ID has already demonstrated imaging with the large hybrid single bunch of the APS achieving a resolution of 100 ps using a white beam, in conjunction with a high-speed chopper.

PADs are routinely used in time-resolved diffraction and spectroscopy having increased the efficiency of experiments by several orders of magnitude. Every time-resolved beamline is equipped with a PAD. The first semiconductor-based ultrafast area detectors are being tested around 2013. They will use arrays of fast diodes to resolve time-dependent phenomena within the bunch structure with a few picosecond time resolution. These fast diodes will start to compete with x-ray streak cameras which have achieved sub-picosecond resolution with x-rays.

With its new upgrade, beamline 8ID will be routinely studying dynamic correlation function with up to a microsecond resolution, and could push down the time resolution to 100 ns in some demonstration experiments with area detector. With a combination of new specialized undulators and long straight sections, the time-resolved community will benefit from flux increases by up to factors of 4-10 which will allow experimenters to push the frontier of time-resolved science.

Enhancement of Medium- and High-Energy-Resolution Inelastic X-ray Scattering Techniques at the Advanced Photon Source in the Near-Term Future (Summary)

Thomas Gog, gog@anl.gov, 630-252-0320

The emergence of inelastic x-ray scattering (IXS) as a set of main-stream experimental techniques has been one of the most significant developments in synchrotron radiation science in recent year. The spectrum of applications reaches from studies of lattice dynamics at the high-energy-resolution (~1 meV) end to correlated electron systems at the medium- and lower-energy-resolution (50…500 meV) end. With substantial improvements in energy- and momentum resolution and greatly enhanced momentum transfer ranges, scientific areas could be addressed that are currently inaccessible to any physical probe, including neutrons and visible light [1, 2, 3].

Upgrade of Existing Instruments

A number of dedicated, state-of-the-art IXS spectrometers are in operation at the APS, including two medium-energy-resolution instruments at 9-ID and 30-ID, one lower resolution, high-throughput instrument at 20-ID, two high-resolution facilities at 30-ID and 3-ID and a few "part-time" instruments in various sectors. In order to dramatically enhance the capabilities of these "photon-flux hungry" instruments within the next 5 years, the following technical challenges have to be addressed:

1) **X-ray Sources**. Long straight sections with additional undulators are necessary to increase the available incident photon flux proportionally. For instruments, that operate at particular fixed energies, optimized undulators need to be considered.
2) **High Heat-Load Monochromators**. With increased incident power, improved high heat-load monochromators based on synthetic diamonds, possibly with cryogenic cooling, need to be developed
3) **Secondary Monochromators**. The next step in the development of secondary, high-resolution monochromators is to advance into the sub-meV regime[3]. At the same time attention has to be paid to multiple bandpass options, to enable coarser orientation measurements at high throughput, easily switchable to high-resolution measurements in the regions of interest.
4) **Focusing Mirrors.** Beam spots at the sample need to be decreased to a few micro-meters to accommodate ever smaller samples and extreme sample environments. A small beam footprint is also important for the precision of modern energy-dispersive crystal analyzer / position-sensitive detector combinations.
5) **Sample Instrumentation.** Small beam sizes necessitate precise micro sample positioning systems, combined with in-situ crystal orientation capabilities.
6) **Crystal Analyzers**. New analyzer geometries and mass fabrication techniques for crystal analyzers have to be developed to equip/enhance existing instruments as well as pave the way for new designs, such as instruments with large numbers of analyzers.
7) **Detectors**. Silicon strip detectors in combination with energy-dispersive analyzers setups have had a great impact in making IXS spectrometers more compact without

loss of resolution while increasing throughput. These detectors need to be further developed, based on cryogenically cooled Germanium chips, to increase detection efficiency for x-rays, especially at higher energies. At the same time, this would reduce noise levels, which limit the applicability of these detectors at the lower end of the energy scale.

Implementation of novel instruments

With the development of new optical components, analyzers and new position-sensitive detection systems, novel instruments can be designed and implemented within the next 5 years, offering access to areas of science which are not accessible today.

Multi-analyzer Spectrometers. Instruments with large numbers of analyzer (> 100) combined with low-noise position-sensitive detectors can increase throughput by orders of magnitude, while being compact without loss of energy resolution. Details for such instruments can be found in [2], and also in [3].

Ultra-high Resolution Instruments. Using novel x-ray optical concepts, a sub-meV monochromator has been proposed [3] and independently, a spectrometer which can achieve sub-meV energy resolution and simultaneously unsurpassed momentum resolution [2].

Both types of instruments are logical extensions of existing IXS spectrometer designs. They would offer substantial improvements in energy-resolution, throughput, momentum resolution and available momentum transfer range and would ensure the place of the APS at the forefront of Inelastic X-ray Scattering research.

[1, 2, 3] See for example
"APS Renewal Medium-Term Proposals: Beamlines 9-ID, 30-ID, 3-ID"

High Energy X-ray Science at the APS in 2013

The Advanced Photon Source offers unique capabilities in the Western Hemisphere for science using high-energy x-rays (defined loosely as photons between 50 keV and 120 keV). The 7 GeV operating energy of the storage ring is very well suited for the production of high-energy x-rays, and it is unlikely that any current or planned storage ring in the U.S. will approach the current high-energy x-ray performance of the APS. Worldwide, however, other facilities (e.g., the ESRF, PETRA-III) are pursuing upgrade paths that expand their capabilities and threaten the APS position as a leading world-class high-energy x-ray source. Fortunately, nearly every aspect of the APS high-energy x-ray operations benefits substantially from relatively straightforward upgrades possible in the five-year time frame. Upgrades in any of the areas will give marked increases in experimental performance for a wide range of scientific applications. Upgrades in all of the possible areas would give an increase in capabilities that is nothing short of spectacular.

Currently, three beamlines are essentially dedicated to high-energy x-ray usage: 1-ID, 11-ID-B, and 11-ID-C. The 6-ID beamline has a side station that is used part of the time for high-energy operations (sometimes parasitically with the low-energy branch and sometimes in a dedicated mode). A few other beamlines (e.g., 5-BM and 13-BM), occasionally operate in the high energy range, but it is not a major part of their operations.

A common factor for high-energy x-ray beamlines is that they are all limited in some way lack of optimization for high-energy x-ray operations. The 1-ID beamline was dedicated to high-energy x-ray usage in 2005, but supports three techniques that mutually limit each other. The 11-ID beamlines share 11-ID with a low-energy beamline, and are compromised in performance by this and their mutual interference with each other. Overall, the dedicated high-energy beamlines are heavily oversubscribed. The only way to address this serious problem without cutting healthy scientific programs is to expand the number of high-energy x-ray beamlines at the APS. The medium-term proposals by APS staff propose a new ID beamline, a new dedicated high-energy bending magnet beamline, and several steps to make current beamlines more independent towards this purpose. With expanded beamline availability, several instruments could be fully optimized, and high-energy x-ray capability would be significantly increased.

The move towards more specialized undulators has already started with the installation of 2.3-mm period devices at 1-ID and 11-ID in the last year. Further use of specialized devices will significantly increase the brilliance of the beamlines, in particular if ID vacuum chambers are modified to allow the closure to at least 9.5 mm. Tremendous gains in flux/brilliance are possible with superconducting undulator technology that is currently in development. Improvement in optics and use of customized beta-functions (w/o this horizontal size limit will be order of 5-10 um) can provide focusing in two dimensions to the submicron level.

The use of large area detectors has revolutionized high-energy x-ray experiments at the APS in the last seven years. The GE amorphous-Si detector is in heavy demand with at least twice as many experiment requesting it as can be accommodated. A similar Perkin Elmer detector is being purchased and this will be of significant help in this respect. However, these detectors are based on technology that is aging, and by 2013 we expect considerable advances, most likely in detector speed and pixel resolution. Improvements in data handling, reduction and analysis are key to fully utilizing detector advances.

Finally, an important factor for high-energy experiments is the penetration possible through furnaces, cryostats, and other sample environmental chambers. Development of such ancillary equipment will continue on all high-energy beamlines. Optimizing and expanding high-energy beamlines will make more of these unique *in situ* experiments possible, including experiments using multiple probes (e.g., diffraction and SAXS, or diffraction and imaging).

Perspective on Status of APS Microprobe in 2013

Jörg Maser, technical coordinator for microprobe (nanoprobe)

July 26, 2008

This is an effort to provide a general perspective; the classifications listed below are therefore somewhat arbitrary, and are not meant to provide a complete list.

A) Status - Summary

The Advanced Photon Source has worldwide renown strength in micro- and nanoprobe techniques and applications. Significant capabilities exist in the following areas:

1) Microprobe work at moderate spatial resolution (> 0.25 μm).
   - Techniques:
     - X-ray fluorescence mapping, X-ray fluorescence spectroscopy
     - X-ray diffraction
     - Combination with techniques such as inelastic scattering, high pressure, magnetic contrast etc
   - Main scientific areas:
     - Environmental/Geo science
     - Materials science
   - High-resolution Optics: mainly KB systems, some refractive lens and diffractive optics
   - Strength:
     - Non-dispersive focusing
     - High flux/large throughput. Makes use of APS *flux*
     - Good flux at high energies (> 30 keV)

2) Microscopy/Nanoprobe with spatial resolution of 0.25μm
   - Techniques:
     - X-ray fluorescence mapping, X-ray fluorescence spectroscopy
     - X-ray diffraction
     - Coherent X-ray diffraction
   - Main scientific areas:
     - Biology, environmental science
     - Materials science, nanoscience, physics
   - High-resolution optics: mainly diffractive optics, some reflective optics, refractive optics
   - Strength: high spatial resolution; makes use of APS *coherent flux (i.e. brilliance).* Throughput is intrinsically limited by coherent flux

Almost all of the micro/Nanoprobe capabilities use x-rays with photon energies between 4 keV and 25 keV, with some important efforts at lower and higher energy.

Outside the area of micro/Nanoprobe, the APS has very significant and world-leading capabilities in development and fabrication of x-ray optics. The APS has a particular competence and leadership in both high-resolution reflective optics and high-resolution diffractive optics.

B) Future Potential

The APS is uniquely positioned to continue pushing the state of the art of its microprobe/ Nanoprobe programs and maintain worldwide leadership.

On the Microprobe level, high flux in small spots allows deployment of photon hungry methods such as fluorescence tomography. Also, higher-energy applications will continue to benefit from large flux. These developments can make use of x-ray optics, namely reflective optics that can be reasonably well fabricated.

On the Nanoprobe level, the APS is in a wordwide unique position to push towards a spatial resolution of 20 nm, possibly below, for hard x-ray energies from 10 keV and below to 30 keV (for higher energies, nanofocusing would be instrinsically limited by small coherent flux, coupled with small cross sections, and microprobe would become the more useful tool). Advancing into the spatial resolution range of below 30 nm will have the potential for large and unique scientific impact: cellular substructures at the level of can be accessed, nanoscale systems be studied at lengths scales that exhibit effects of confinement, and the elemental sensitivity of x-ray nanoprobes will improve into the range of individual nanoaparticles (less than 100 atoms, Zn, 10 keV).

While this author sees strong potential in the improvement of microprobes, the opportunity in the Nanoprobe range appears to be unique and timely. *Unique* in that the APS has worldclass expertise in all relevant technologies: (i) Optics R&D and Fabrication (ii) nanopositioning engineering and metrology (iii) scientific expertise in biology, medical sciences, materials science and nanoscience. *Timely* in that recent instrumentation progress makes sub-30 nm focusing feasible, and that a significant number of scientific applications in the fields mentioned above aggressively request higher spatial resolution,

In order to further strengthen the microprobe programs, in particular with view to throughput, and to make significant progress towards nanofocusing and its potential applications, investment into the following areas would be of significant importance:

1) Efficient, fast, large-solid angle detectors/detector geometries and (relatively low-maintenance) electronics to allow x-ray fluorescence mapping and spectroscopy at high fluorescence counts (e.g. 10 Mhz count rate at 150 eV resolution (Mn Ka)). This will e.g. enable fluorescence tomography in efficient microprobes as user tool by maximizing throughput, and both reduce radiation damage and increase throughput for Nanoprobe methods that must use coherent flux only.
2) Coupled with (1), development of efficient, fast data acquisitions schemes and related data buffering/storage systems.

3) Move towards good vacuum or clean He environment for reflective microfocusing optics. This will reduce contamination and increase (i) lifetime of microfocusing mirrors (ii) coherence preservation of nanofocusing mirrors.
4) Push towards 10 nm x-ray focusing optics for the 5 – 30 keV range, with a 5-year goal of a 2D resolution of 15 nm x 15 nm at good focusing efficiency
5) Increase nanoengineering capabilities. Mechanical engineering is only one leg of nanoengineering. To achieve control at the nanometer level, it must be coupled with metrology *and* proper controls implementation. Integration of advanced positioning concepts with sophisticated but stable metrology systems and related controls is required. This includes metrology capabilities that allow testing of advances positioning.controls systems. Close and effective collaboration across APS groups to create such a nanopositioning capability would appear to be a promising opportunity.
6) Cryo capabilities are required to reduce the effect of structural damage to samples in both biology and materials science. Radiation doses at the sub-30 nm resolution level approach and might exceed 10^{10} Gy, and LN2-based cryo techniques have been demonstrated both electron and x-ray microscopy to be effective up to the 10^{10} Gy level.

Where the APS could be in 2013 for X-ray Absorption Spectroscopies

Matthew Newville, CARS, The University of Chicago

28-July-2008

X-ray absorption spectroscopies (XAS, including EXAFS, XANES, and XMCD) are relatively mature techniques that make use of energy-tunable x-ray sources to study the atomic-scale structure, chemical environment, and magnetic state of selected elements. These techniques do not rely on sample form (for example, crystallinity), and are sensitive to very dilute elemental species in complex systems. These properties make XAS attractive for study of highly disordered systems, including nanoparticles, liquids, molecules, and heterogeneous materials, and so applicable to important research areas for many fields. In addition, the elemental-specificity of XAS is closely related to resonant inelastic x-ray scattering, x-ray Raman spectroscopy, hard x-ray photo-emission, and resonant elastic scattering. At the APS, XAS has been coupled successfully to micro-beams, and used extensively for environmental, chemical, and material sciences. Currently, most beamlines at the APS using XAS as a core technique are CAT beamlines or those taken over by XOR after being fully developed and running successful user programs. However, about one-third of the beamline proposals touch a need for improving, upgrading, or enabling new uses of XAS or related spectroscopies that use core-electron level resonances.

There are a few important ways in which the APS could lead development and use of XAS and related techniques.

First, the APS could develop support infrastructure for sample environments, detectors, and data analysis and interpretation specific to XAS. Examples of these infrastructures are

- facilities to modify high-pressure diamond anvils suitable for low-energies (6-10keV) -- drilled diamonds, and for poly-crystalline anvils to avoid inevitable Bragg peaks from the diamond anvils.

- gas-handling systems for in situ and in operando catalysis work.

- an active Theory group and Analysis Software group that focused on some of the imperfect aspects of XAS Theory and Analysis.

Second, while XAS at the APS is quite strong in micro-XAFS and applications in environmental science, the APS could work to expand support for important applications of XAS that are under-represented at the APS (relative to how XAS is used at other facilities), especially catalysis and life sciences. There is currently no dedicated facilities at the APS for XAS in either of these two important fields.

Third, the APS could more fully support the development of better detector systems for XAS. Detector needs for XAS are primarily for fluorescence XAS, which are currently led by Si-drift fluorescence detectors. With multi-element Si-drift and Ge-drift fluorescence detectors, the APS XAS community could gain an order of magnitude in count rate and sensitivity.
The most efficient approach that the APS could take would be to follow the work of Siddons and Ryan at BNL/ASRP on the so-called 'Maya' detector.

But probably the biggest impact on XAS that the APS could take would be to lead in the development of RIXS / high-resolution fluorescence for more wide-spread application in areas currently using XAS. This technique (measuring the K-edge EXAFS decaying into a ~1eV bandwidth of a specific alpha or beta fluorescence line) has been proven to be able to add two new "knobs" on XAS: spin-selectivity, and species-selectivity, both of which have very high potential for applications in existing areas and expanding our abilities to study new problems. While these techniques are demonstrated, they generally need sources with high brightness, and there are few beamlines in the world dedicated to these advanced techniques. With a fairly small effort, the APS could become the leader in the application of these techniques as an improvement over current XAS measurements.

--Matt Newville <newville at cars.uchicago.edu>

Macromolecular Crystallography APS 5-year Plan

1) Automation – Four of the 8 CATs have operational robots , 2 are in commissioning mode and one is awaiting the delivery of their robots. In the next couple of years all Macromolecular Crystallography (MX) beamlines will have operational robots. The most obvious reason for these automounters are convenience and efficiency. They will become indispensable in screening large numbers of crystals of variable quality to identify a few well diffracting ones. The molecules themselves are becoming larger and more complex and the crystallization more difficult.

2) Remote operations - Three of the CATs use remote operations. Of the three, two of the beamlines are for pharmaceutical companies. SER CAT is the only academic beamline that offers remote operations to its member institutions. This is not to be confused with mail-in service. It is for the use of the beamline by experimenters while at their home institutions. SSRL claims about 80% remote operations. The prerequisites are an operational robot and remote access. A fast, accessible network infra-structure would benefit all beamlines. Computational and experimental methods for quickly and reliably detecting and centering crystals are still being developed and tested.

3) Mini/micro beams and micro crystals – crystals are getting smaller and the sample molecules are getting larger. Mini/micro beams will allow users to reduce background, obtain data from several parts of a crystal or the best parts of large crystals, and to provide a workaround radiation damage. In most crystallographic beamlines, with this mini/micro-beam capabilities, the beam is produced by aperture restricting a focused but larger beam. Further investigation of the use of focused microbeams will determine future directions. This area is growing rapidly in the US and the APS is the best source to take advantage of this. Technical problems that will need to be solved are increased beam stability in the ring, in the beamline optics, and stable experimental equipment.

4) Detectors – During the next five years most of the detectors on the beamlines will be at least 10 years old. The switch over will be to improved CCD detectors or tiled pixel array detectors. The advantages are improved accuracy, lower background, increased sensitivity, and the possibility for shutterless, continuous data collection. The higher dynamic range will allow collecting data to search for smaller (sulfur) anomalous signals. The speed of the detectors is a bonus that will tax both network and storage capabilities. Network infrastructure will need to be upgraded and file handling capabilities will need to keep up with the speed of the detectors.

5) Combined techniques – Development/Implementation of compact in situ spectrophotometers. The spectrophotometer would depend on the system being studied. Possibilities range from absorption, Raman, FT-IR, or fluorescence.

6) Sample environment – This category covers screening crystals in their growth environment, helium chambers and cryostats for low energy, commercial devices that adjust the relative humidity around the sample, and high pressure cells.

7) Laue/time-resolved crystallography – remains a unique branch of protein crystallography that would benefit from the development of fast-readout large-area detectors for pump/probe experiments in the sub-ms time domain.

8) Access to beamlines – Further improvements to streamline general user application to beamlines and to provide easily accessible general information. Cost normalization for bend magnet beamlines versus undulator beamlines may stimulate interest in these beamlines.

9) Crystallographic software support – Automated crystal alignment, scoring of diffraction images, pipelining of images to structure solution, and data bases to handle the increased efficiency of screening and data collection, are areas that will continue to grow in the next five years.

High pressure component in the APS renewal plan
Guoyin Shen, July 25, 2008

Pressure is a principal variable for directing and controlling matter as well as for the synthesis of novel materials. High pressure (HP) has become a major component at synchrotron facilities, because the brilliant synchrotron radiation is an ideal source for small sample volumes in HP environments. At APS, high pressure has been successfully integrated with a number of synchrotron techniques, with significant developments in maximizing brilliance, stability, detecting efficiency, and spatial resolution, minimizing background noise and unwanted signals. The established frontier at many beamlines will be greatly advanced from the overall APS upgrade in x-ray source, beamline optics, advanced detectors, and software controls. In addition, I point out several areas that may be unique to HP and particularly important for advancing to the next generation HP synchrotron research.

Improving depth resolution: While the spatial resolution in a plane perpendicular to x-ray beam has been effectively improved by reducing a probing size, the spatial depth resolution along the x-ray beam remains an under-developed area. Yet, this should be the most important consideration for all HP experiments, because HP samples are always surrounded by chamber materials which often cause strong background signals. Improving depth resolution not only will greatly increase the counting efficiency due to the improvement of S/N ratio, but will make a number of cutting edge projects feasible. The conventional pinhole collimator is often used on the detection side in HP studies, which typically provides a depth resolution of 300-500 μm. The newly developed x-ray lens (e.g., polycapillary x-ray focusing optics) may be used to construct a confocal microscope for collecting scattering signals. Such an x-ray lens can collect a large solid angle (up to 20 degree) with a focus spot as small as 10 μm. The use of x-ray microscope on the detection side will improve the depth resolution by at least an order of magnitude, and will significantly reduce the background from surrounding materials and enhance the detection sensitivity by >100 times. The depth resolution gain through an x-ray microscope is particularly beneficial in HP IXS.

Sub-micron beam size: Since pressure is defined as force per unit area, ultrahigh pressures can be reached by minimizing the area at the tip of diamond anvils. However, this is limited by the minimum available beamsize, which is typically 5-10 μm at HP beamlines around the world. However, beam sizes of a few tens of nm have been realized at several specialized beamlines and the implementation of these sub-micron beams for high-pressure measurements could facilitate x-ray measurements at unprecedented static pressures measured in terapascals. In addition, this rare opportunity for an order-of-magnitude improvement in the spatial resolution of the probe will enable many other new cutting-edge areas, and bring HP synchrotron x-ray diffraction, spectroscopy, and tomography to a new level.

Specialized HP vessels: The diamond anvil cell is almost ubiquitous at HP x-ray facilities around the world. It's success stems from an intrinsically simple conceptual design, yet, for certain specific x-ray techniques, there is a clear demand for a more optimized HP vessel. For instance, for poorly scattering samples, stringent signal to noise limitations will drive the development of a new generation of pressure cells with two primary foci: to minimize the amount of illuminated anvil material and to maximize the sample volume for a given pressure. Innovative pressure cell designs will open up new frontiers in data quality, enabling studies of systems that are currently flux limited, even at 3rd generation synchrotron sources. Significant

effort needs to be spent on developing these optimized HP vessels and integrating these with specialized beamlines and x-ray techniques.

Portable and dedicated systems: In addition to HP vessels, HP experiments often require other ancillary equipment, such as ruby fluorescence system for pressure measurement, laser heating system for heating sample, cryostat for low temperature, and membrane system for precise pressure control. This equipment is often dedicated at HP beamlines. By making them portable, all other beamlines can have access to such equipments for HP studies. The idea is similar to the detector pool at APS. Having a pool of portable systems will boost the HP activities at APS and enable a series of new HP studies with specialized x-ray techniques that are not available at dedicated HP beamlines.

State-of-art central HP facility: Successful HP experiments depend critically upon the quality of sample preparation. Next-generation HP research will require focused effort on micro-machining and micro-manipulating techniques in sample preparation. For example, we are now facing grand challenges to fabricate single crystals of various materials to meet stringent requirements of shape, size, and crystal perfection. Techniques such as high pressure single crystal diffraction, inelastic x-ray scattering and charge/spin-density-wave x-ray diffraction, all require perfect single crystals without background scattering from damaged surfaces. This constitutes a unique challenge when the ideal HP sample is typically plates of several microns thickness and several tens of microns width. Femtosecond (FS) laser micromachining plus ion-milling surface cleaning shows promise, since it provides high quality micromachining of many materials and abilities for minimal damage and precise processing. This will also benefit the strain-free cutting of silicon and diamond single crystals for the Optics Fabrication and Metrology Group at APS.

High Resolution Powder Diffraction at the APS

Science

Definitive knowledge of the crystal structure of a material—inorganic, organic, or biological—is the gateway to understanding its physical properties, its chemical reactivity, and/or its biological functionality. It is the most fundamental aspect of any material. The increasingly complex chemistry and physics of modern materials demands that this structural information be obtained in a routine fashion and with state-of-the-art precision. Because most technologically critical materials only exist as polycrystalline solids, the definitive structural experiment requires high-resolution x-ray powder diffraction. Measurements must also be made with large dynamic range and great sensitivity, so that small signals may be discerned that indicate changes in structural detail or impurity phases – both highly important. Further, measurements must be rapid enough to meet the demand for the experiments and to be used for study of dynamic systems.

Powder diffraction is applied for study of materials of interest to fundamental physics, materials science, mineralogy, and biology. All of these impact DOE missions that are in the national interest including energy storage, remote sensing, environmental remediation, and metallurgical testing and validation. Examples of active problems in condensed matter physics and materials science follow.

The behavior of charge and orbital degrees of freedom is recognized as a powerful organizing principle for understanding the physics in transition metal oxides and chalcogenides. For example, the properties of colossal magnetoresistive (CMR) oxides depend critically on the relative occupation of z^2 and/or x^2-y^2 orbitals on Mn. Recent study of the spinel $CuIr_2S_4$ has revealed a novel charge-ordering motif in which Ir^{3+} octamers interleave with Ir^{4+} octamers. Examples of other systems that manifest charge and/or orbital order are Li-intercalated CoO_2 and spin-crossover compounds (e.g. cobalt oxides). Importantly it is the cooperative ordering of the charge and orbitals on either short- or long-range length-scales that determines the rich physics of manganites and other related systems. The examples given above demonstrate that the full breadth of charge order and orbital order remains to be discovered and understood. Anomalous scattering at the appropriate x-ray absorption edge for the oxidation states of interest is a powerful method to study charge ordering.

Commonly, developments in materials science arise from synthesis and subsequent characterization of materials with certain properties of interest (conductivity, thermal expansion, magnetoresistance, catalytic effect, etc.). Understanding of these materials originates from precision structure determinations coupled with parametric studies (T, pO_2, composition, etc.). As most of these materials rarely come as single crystals, high-resolution powder diffraction is essential for structural work. A recent example comes from the field of dielectric ceramics, in which complex phase equilibria in the $LaCa_{0.5}Zr_{0.5}O_3$—$SrTiO_3$ pseudobinary have been studied. Phase diagrams and structure-property relationships were determined for several samples quenched from synthesis

conditions. The proposed instrument would dramatically increase the rate at which the phase space in such systems can be mapped, as well as offering *in situ* monitoring of reactivity and phase transformations at elevated temperatures.

High resolution powder diffraction likewise is a key research tool in many other fields, including structural biology, geosciences, catalysis, pharmaceuticals, and many others. For exploration of new pharmaceuticals materials, powder diffraction is vital for exploring polymorphic forms – now required by the FDA, since crystalline form affects bioavailability. Likewise it holds promise as a high throughput screening tool for drug binding studies using macromolecular powder diffraction crystallography. The applications in other fields are too numerous to list. It comes as no surprise that all new 3rd+ generation sources have new powder diffraction capabilities.

Powder Diffraction Instrumentation

To obtain ideal resolution in powder diffraction a highly monochromatic beam is used. Detection typically is done with an analyzer crystal, which serves to limit the acceptance of the detector to parallel rays originating from the sample, while rejecting fluorescence and Compton scattering that deviates in energy. These pseudo-parallel beam optics allow resolution to be decoupled from the sample size.

Linear and area detection offers potentially many orders of magnitude greater efficiency. However, pixelated detectors have higher background, since collimation and energy analysis is not possible. Further, in a typical instrument, resolution is limited both by the pixel size of the detector and the finite size of the sample. Decreasing the sample size diminishes the number crystallites that scatter, which decreases the accuracy of the measurement. New instrument designs, which introduce focusing optics close to the sample, coupled with new designs for detectors have promise for overcoming this problem as this allows a large beam on the sample which is focused to match the detector point spead function. New generations of detectors may also offer energy discrimination, giving further favor to use of pixilated detectors. The development work for new powder instruments at Diamond and NSLS-II is also prompting new designs for analyzer-based detection.

The APS is fortunate that our bending magnets have a spectrum and brightness very well matched to the needs of powder diffraction. However, use of focusing to subtend the large divergence of the source causes minor degradation of resolution. A bending magnet instrument will suit most experimental needs, but there will be a small number of experiments that can only be performed with an insertion-device source.

XOR currently has three beamlines that are used for high-resolution powder diffraction, 33-BM, 1-BM and 11-BM. 33-BM is a general purpose diffraction station that is migrating towards surface-interface scattering applications. 1-BM is a highly versatile instrument that currently is deployed in specialized diffraction measurements and fuel-spray imaging. 11-BM is a recently commissioned high-throughput and high-resolution instrument. It has a best-in-world 12-analyzer detection system that provides the alignment flexibility of a discrete detector system but with better than an order of

magnitude improvement in throughput. The current scope of the 11-BM instrument is limited to samples of a specific mounting geometry and measurements in the temperature range from 80 K to 500 K.

Development Plan

This proposal seeks to expand the utility of the 1-BM instrumentation through modernizing it. It seeks to expand the capabilities of 11-BM and develop an instrument along a similar design to 11-BM that would share beam on an insertion device station.

1-BM needs an upgrade of optics, since the components are now over 10 years old and well below the state of the art. The relevant optics to be improved are a water-cooled flat white-beam mirror, a flat vertically focusing mirror, and a sagittal bending second monochromator crystal. The mirror tanks and mirror supports (including the bending mechanisms) will be kept and reused. The monochromator at present requires very high level of training to tune. A more advanced design has the potential to be more user friendly and allow automation. It can also extend the energy range of the station. With this upgrade, 1-BM will again be among the highest flux bending magnet beamlines in the world. Many experiments would benefit from an advanced detector system, for example a tiled array of amorphous silicon detectors. Others would benefit from a multiplexed analyzer system, such as the 12-crystal system in use at 11-BM or perhaps the 64-detector system that is has been discussed for Diamond.

11-BM optics are limited by the poorly designed mirror mounts, which make beamline alignment difficult and will prevent easy access to the planned energy range. While the initial proposal for 11-BM envisioned a wide range of sample environmental support, only the Oxford 700+ cryostream device remained in the final program scope. In contrast, note below the planned suite of ancillary equipment that will be available on the Diamond I11 powder diffraction station when it begins operation in August 2008:

- Stoe capillary furnace (T = 300 – 1700 K)
- MRI flat plate furnace, (T=300 - 2000 K)
- Bruker Humidity chamber (-5°C up to 75°C dew-point, T= 25 – 90°C)
- Linkam DSC (T = 77 – 870 K)
- Cyberstar Hot air blower (T = RT-1300 K)
- PheniX He Cryostat (T = 11 – 300 K)
- ASI cryostat (T= 4 -300 K)
- Oxford Cryosystem 700+ cryostream (T = 80 – 500 K)

All of these capabilities are vitally needed by US scientists. Additional engineering will be required to adapt the goniometer to support such equipment and to allow it to be shared with 1-BM.

With the envisioned improvements to 1-BM and 11-BM, we envision that most beamtime on 11-BM will be dedicated to high-throughput diffraction measurements. A

large fraction will be mail-in samples that are run under routine conditions. In general, experiments that require considerable setup time will be run on 1-BM, which is a more flexible station. As mentioned, however, there are some experiments that require the best possible resolution and sensitivity. These measurements can only be performed on an ID line. Since the demands of such an instrument are modest both with respect to beamtime shifts and physical space, this is can share with any number of beamlines and does not need a dedicated source. Such an ID line may also prove to be much better for automating resonant scattering experiments, since focusing is not required and the 11-BM analyzer design has proven to be easy to align when changing energy.

User Community and Partnerships

The user community for synchrotron powder diffraction at the APS can be expected to grow to well over a hundred academic groups. It will certainly include a number of industrial laboratories, including UOP, Chevron and General Electric, who are already active in the brief time since 11-BM became operational. Government researchers in military and the DOE can also be expected to major users. Key partners may be:

- John Mitchell (MSD)
- Angus P. Wilkinson, Chemistry, Georgia Institute of Technology
- Yan Gao, General Electric
- James Kaduk, INEOS technology
- Ken Poeppelmeier, Chemistry Department, Northwestern University
- Clare Grey, Chemistry, Stony Brook
- David Bish, Geology, Indiana University

Estimated Budget

The budget for this project will be on the order of $400 K for 1-BM, $500 K for 11-BM and $2M for a new ID instrument. A total of additional 6-10 FTEs will be needed to properly staff the instruments, depending on the scope of the intended user program.

Where APS Full-field x-ray imaging can be in 5 years.

A: 'Conventional' absorption/phase x-ray imaging with spatial resolutions ~ 1 micron

1. Larger field of views (~ 25 mm H x 10 mm V) to accommodate mouse-sized samples.
2. Improved spatial resolutions to below 1 micron (from current 1.2 micron).
3. Improved image quality for phase-contrast due to smaller angular source size; achieved via a long beamline and/or accelerator horizontal beta improvements.
4. Complete 3D tomography data set in ~ 1s
5. Improved S/N > 40 for single-shot (100 ps) exposure by optimized insertion device and cooling of scintillators.
6. Tomography of extended objects: either by local tomography or stitching
7. New contrast agents will be developed for animal physiology studies.
8. Increasing integration of simultaneous x-ray imaging to other techniques (eg., optical microscopy or x-ray scattering).
9. Tomography reconstruction times reduced to ~ 1 minute for 2Kx2K images.
10. Use of larger detectors (4K and larger).

Types of experiments:
Ultrafast imaging of supersonic liquid jets/sprays and crack propagation in materials.
High resolution real-time studies of fluid flow patterns in small animals, eg, intra-arterial blood flow or intra-gut mixing.
Very high spatial resolution CT of an extended object eg., 1 micron resolution of an internal crack on a 1 cm piece of material.
Almost-real-time (~ 1 Hz) 3D evolution of material dynamics such as fatigue-induced cracks and corrosion.
Simultaneous measurements in real and reciprocal space eg., to look at local strains in fatigue-induced cracks.

B: Full-field transmission x-ray microscope with spatial resolutions < 100 nm

1. 20 nm spatial resolution at 8-18 keV with ~ 50 micron field of view
2. Hardware/software developments to enable true automated tomographic data collection and reconstruction. (Current system severely limited by mechanical systems).
3. Increased imaging throughput to ~ 20 frames per second allowing for sub-second dynamic studies.
4. New targeted contrast agents will be developed for cellular imaging studies.

Types of experiments:
3D mapping of individual tagged neurons in very small animals (eg. fruit fly).
3D imaging of subcellular structures or organelles.
3D investigation of hard materials under in-situ environmental control
Almost-real-time nanoscale radiography of dynamics.