A strategy to position the APS as the World's Premier Synchrotron Facility for Engineering Applications and Applied Research

1. Executive Summary

We propose a strategy to make the APS the leading synchrotron institution in the world for engineering applications and applied research. This proposal includes hardware and software upgrades that will make the APS accessible to a wider range of engineering and applied research scientists and will allow for unprecedented experimental capabilities. The suggested upgrades exploit unique capabilities of the APS that enable fundamentally new characterization tools and offers a huge return on investment that will stimulate American competitiveness and create new opportunities in areas of critical national interest including energy independence and national defense.

The proposal leverages off special properties of the APS that make it ***the key x-ray source in the United States for engineering applications and applied research.*** In particular the APS high brilliance at short wavelengths creates unprecedented opportunities to study ***real materials in real environments and in real time***; with ultra-brilliant hard x-ray beams, materials can be characterized ***nondestructively*** in situ, during service and deep within actual parts where materials behavior is fundamentally distinct from the behavior in near-surface regions. For example, ***spatially-resolved*** measurements with high-energy x-rays are a transformational class of synchrotron science with the potential to revolutionize engineering and applied sciences; spatially-resolved characterization of stress distributions, texture and particle size distributions that evolve near structural inhomogeneities can test and guide models and dramatically improve the performance of parts. Nondestructive spatially-resolved measurements will also allow for characterization of process driven grain growth and phase evolution including the driving forces of anisotropic or transient stress states. Other approaches will allow for ultra-high-resolution characterization of engineering materials at the mesoscale level of defects such as grain boundaries, dislocation walls and phase boundaries to test emerging mesoscale and multiscale theories of materials behavior. In short the emerging potential to image scalar and tensor distributions of chemistry, structure, orientation and defects in three dimensions will transform our ***understanding of the hierarchical structures that control materials behavior***.

An area of almost unexplored potential will be the detection and characterization of minor phases that emerge at interfaces of real materials including complex phases at cladding interfaces in nuclear fuel rods, phases at the fiber/matrix interface in composite materials and in service measurements of corrosion including elastic stresses and studies of stress corrosion cracking. In addition to spatial-resolved measurements, ***time-resolved*** measurements –made possible by high source brilliance and efficient x-ray sensitive area detectors- will allow for the discovery of transient phases that evolve during welding, heat treatment, concrete solidification, amorphization and other phase evolving processes.

Finally, high-brilliance, high-energy beams offer unique advantages for ***nondestructive imaging***. For example phase-contrast high-energy radiography and tomography can resolve structures in low Z or low-density materials that would otherwise be invisible to x-rays. Phase contrast imaging can also resolve internal structures in specimens comprised of mixed Z components or inside environmental chambers, and can characterize sprays and jet distributions that are difficult to study by alternative methods. With advanced instrumentation, imaging can also be used to follow the evolution of cracks, voids, precipitates and other phenomena in near real time.

To realize the immense scientific potential of the APS for engineering and applied research, ***dedicated instrumentation*** is essential, including sophisticated ***hardware, software and high-performance computational facilities***. To address urgent near-term needs and emerging scientific opportunities we propose the following steps which will significantly increase user access and experimental capabilities:

1. Dedicate an additional beamline to high-energy x-ray measurements of aggregate stress/strain/texture from materials.
2. Complete the 1-ID Phase II upgrade.
3. Build a dedicated polychromatic nanoprobe hutch and install canted undulators on 34-ID to allow for simultaneous and independent use of polychromatic mesoscale and nanoscale probes.
4. Optimize at least one bend-magnet beamline for high-energy energy-dispersive diffraction.
5. Develop a dedicated high-energy tomography station with phase contrast sensitivity.
6. Develop a range of environmental chambers for powder diffraction and SAXS targeted for catalysis and other applied studies.
7. ***Develop user-friendly "expert" software for all engineering stations that insures users walk away from experiments with data sufficiently processed for analysis at their home institution.***
8. Coordinate combined techniques/characterizations to follow materials evolution.
9. Develop user-friendly mail-in capabilities in anticipation of the growing difficulty of travel with high energy costs.

2. Introduction

A fundamental multiscale understanding of the origin of materials behavior is vital to the design and development of new and improved materials for structural, energy and other engineering applications. Similarly, applied studies of fuel spray turbulence, dendritic growth, bubble formation, chemical segregation/transport and other processes are essential to optimize and guide the development of important systems like gas-engines, electrodeposition systems, fuel cells, toxic barriers and batteries. These kinds of studies can be classified as engineering applications and applied research and represent an area where the APS can significantly impact technologies of critical national interest. The unique properties of APS beams make it urgent that work in this area be supported by upgrades to undulators, beamlines, techniques, and supporting infrastructure. Indeed much of the science proposed here and discussed in APS mid-term upgrade proposals, http://www.aps.anl.gov/Renewal/mt_beamlines.html, simply cannot be done at other synchrotrons in the Americas. Even the ultra-low emittance projected for the new NSLS II cannot produce *high-energy* beams with the brilliance possible from the APS (Fig. 1).

The importance of the APS for engineering applications and applied research is clear from recent growth and facility oversubscription in these areas. For example, polychromatic microdiffraction developed at the APS[1,2,3] is now being instrumented on beamlines around the world- but the infrastructure and capabilities of the APS station 34-ID-E remain unmatched with a >400% over subscription for available beamtime and with outstanding proposals unable to secure beamtime. Similarly the development of high energy diffraction microscopy[4,5] programs and high-energy powder, small-angle-scattering, and other diffraction methods[6,7,8] have developed large communities and available beamtime is highly oversubscribed for these powerful probes of engineering materials. Other techniques like high-energy imaging have been demonstrated but have not been properly instrumented for routine user operations.

The explosive growth of these research areas is driven by the high-energy brilliance of the APS, which allows for nondestructive imaging of systems during processing or in service. The combined ability to *nondestructively probe* volumes deep inside complex environments with *spatial resolution*, *time resolution* and density, crystal and chemical sensitivity requires ultra-brilliant high-energy x-rays. There is simply no source in the Americas with the high-energy brilliance of the APS.

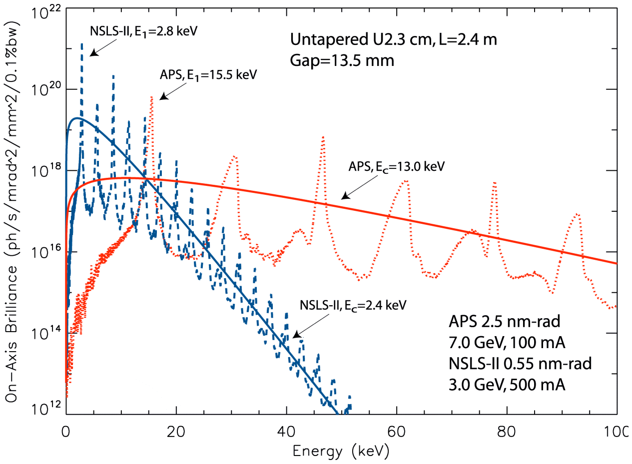


Fig. 1. On-axis brilliance for the *un-tapered* undulator U2.3 cm (APS23#1) installed on the APS and the NSLS-II storage rings. The undulator is 2.4 m long and the undulator gap is 13.5 mm. The first harmonics and the critical energies for two cases are labeled. The solid lines show the wiggler approximation using the corresponding critical energies

Although there are no comparable sources in the Americas, the ESRF in France, Spring-8 in Japan and the new PETRA III facility in Germany are international competition with similar potential to the APS and with strong engineering and applied programs. Synchrotron research for engineering materials is a particularly active research area in Europe with strong ties to major European industries like Airbus and Volkswagen and with close collaborations with universities.[9] Nevertheless, the proposed upgrades will provide scientists in the United States with a suite of tools for engineering applications and applied research that is unmatched anywhere else in the world. Beyond these upgrades however, there is a need to develop closer collaborations with industrial and university researchers to best exploit the capability of synchrotron research to transform engineering and applied research and make the U.S. more efficient and more competitive. In order to realize a meaningful collaboration with non-synchrotron specialist university and industrial scientists, the control and data analysis software for engineering and applied research must reach a new level of sophistication that makes experiments at the APS more user friendly. This is a realistic goal, which together with a suite of sample environmental chambers will make efficient use of precious engineering and applied research capabilities.

3. Key Science Drivers

The ability to *nondestructively* image materials inhomogeneities with unprecedented *spatial*, *chemical*, *crystallographic* and *defect* sensitivity opens up fundamentally new opportunities for engineering and applied research. Similarly, the ability to detect and characterize ultra-small sample volumes and transient phases will transform our understanding of materials behavior during processing and in-service and will bring understanding to nonlinear phenomena where average properties are insufficient to understand materials behavior. Below we briefly touch on a few of the many long-standing engineering and applied research issues where new instrumentation and software can have a major impact.

Fracture: Fracture is a long-standing issue in engineering materials with broad implications. Lack of control and understanding of fracture necessitates over design of parts that adds cost and weight and lowers performance. Fracture is inherently a spatially inhomogeneous phenomenon with atomic level interactions near the crack tip and with mesoscale strain fields both near the crack tip and in the post cracked region. For this reason nondestructive spatially-resolved measurements can provide essential new information to understand crack behavior. Of course, beautiful TEM experiments can image crack propagation in real-time, but dislocation motion is recognized to be influenced by the small sample size required for TEM; X-ray studies are essential to study cracks in samples with true bulk behavior.

This is an area where research has already begun, but where improved instrumentation and software can have a huge impact. For example, ***energy dispersive*** studies of fine-grained materials have detected unexpectedly large plastic deformation "wake fields" behind propagating cracks.[10] In addition, studies of Ni-Ti alloys have revealed the extents of both plastic and shape-memory zones around stress concentrations.[11] The APS has the brilliance to push the spatial resolution of these measurements by orders of magnitude and with time resolution. This will require high-performance achromatic focusing optics to achieve small beams and ultra-fast banks of energy-resolving detectors to characterize the local plastic and elastic strains. In addition, special techniques implemented at the APS -like ***polychromatic microdiffraction***- and ***high energy diffraction microscopy*** allow for sub-grain-resolved studies in samples with typical grain sizes of 0.5-20 microns. Ultimately an improved understanding of fracture physics will have a direct and widespread impact on part design and materials.

Related areas of research include, fatigue cracking[12] and stress corrosion cracking[13] that impact industry, transportation and energy production. An example in this area includes thermo-mechanical studies of alloys used for nuclear reactor applications. Recent HE scattering studies have provided strain and texture evolution of non-irradiated Zr-based alloys[14] used as cladding materials for current and future reactors. Future studies on radiated and composite materials, under realistic conditions of thermo-mechanical deformation, are anticipated to provide unique spatially-resolved microstructural information and improve life-cycle predictions.

Stress, residual stress and deformation: Understanding mesoscale structural evolution in materials is one of the great challenges for engineering materials research. Recent work at the APS has begun to characterize defect self-organization in systems where the starting conditions and processing conditions are sufficiently well defined to allow for direct comparison to models. This is an area of enormous importance where better spatial resolution, faster detectors and more brilliant sources can move experiments from demonstration and proof-of-principle to transformative. Measurements on single crystals[15,16], bicrystals[17] and polycrystals[18] are beginning to provide new guidance for how dislocation behavior differs near surfaces and interfaces from the behavior within a grain (Fig. 2) This information is essential for multiscale models of engineering and other materials. The ability to nondestructively study local crystal rotations with submicron spatial resolution and with good angular sensitivity now allows for the first nondestructive measurements of the Nye or dislocation tensor and has stimulated the development of a formalism to cleanly separate plastic from elastic deformation in complex stress states.[19] This will enable critical tests of plasticity models that invoke the Nye tensor to explain evolving materials behavior.

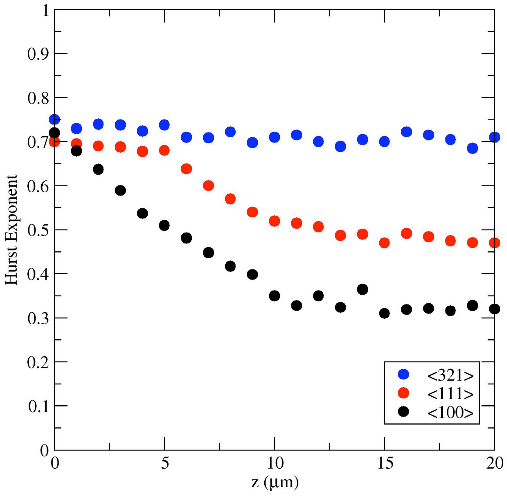


Fig. 2. Hurst coefficient for correlated lattice rotations in a deformed single crystal shows crystallographic and depth-dependence from Pang et al. In the 321 crystal only a single slip system is activated and the near surface behavior extends deep into the crystal. In the 111 and 100 orientations, multiple slip systems are activated and the near-surface behavior is confined to a few micron region.

Residual stress measurements (lattice strains) are another important application where HE x-rays can transform studies of engineering materials. This essential area for industrial research has long been the domain of neutron scattering due to the deep penetration of neutron beams into real parts. High-energy x-rays offer the potential for good sample penetration with much faster measurement speeds and with much better spatial resolution.[20] Since residual lattice strains ultimately arise from defect distributions in materials, the improved resolution, and faster time scale offered by a dedicated high-energy beamline will both guide designers and resolve specific issues with engineered structures, and will tie directly to model simulations of defect evolution in materials. The need for such improved structural assessments is anticipated to grow, for example, in the nuclear industry where planned plant lifetime extensions will place emphasis on limiting damage to critical components such as welds.

Deformation in novel materials: The APS renewal also provides opportunities for transformational studies of novel structural materials including nanomaterials and amorphous materials. Recent in-situ HE PDF studies have demonstrated the ability to characterize strain in amorphous bulk metallic glasses[21,22]. These first experiments show a surprisingly strong dependence on local chemically-specific atomic correlations. Extensions of this technique will provide unique insights into the role of local structure and stress in amorphous materials. Similarly, the unique deformation properties of nanoscale materials, has begun to be understood with the aid of HE scattering studies.[23] Finally, high-energy studies of biomaterials have revealed important information about the micromechanics of bone deformation, and extensions of these studies towards biomaterials constructs such as implants in bone and teeth are anticipated.

Dynamic deformation: Materials behave fundamentally differently under dynamic deformation, defined at timescales on order of hundreds of nsec and below. Applications here are primarily defense / national security related and post-deformation analysis can provide information about activated slip systems and other deformation mechanisms.[24,25] Ultimately however real-time measurements are desirable and possible studies include (i) high-energy x-ray scattering with large detectors capable of ROI readouts (nsec fast) and (ii) high-speed radiography. The APS recently hosted a dynamic compression workshop, which advocated a separate CAT based on the special needs of the dynamic deformation, community; information can be found at http://www.aps.anl.gov/News/Conferences/2008/DC_Workshop/

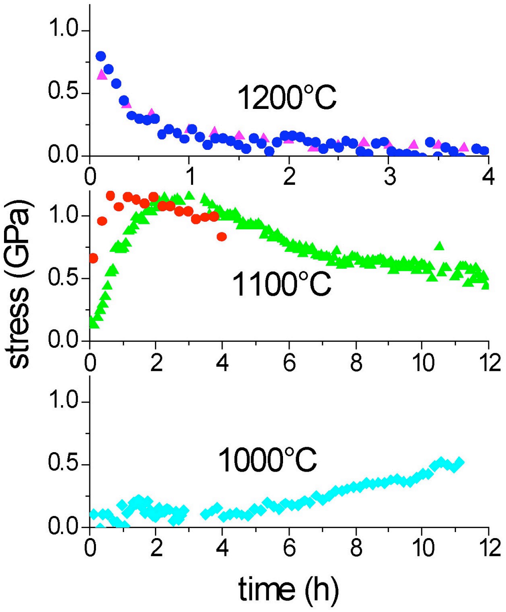
Imaging: For all these cases, elemental maps and 3D tomographic visualization can support emerging diffraction research. For example, 3D density maps can help identify positions of greatest interest in crack samples and can follow the crack tip to guide diffraction studies. Similarly in some materials minor elements are known to drastically influence intra-granular cracking. For example, boron and other elements are known to ductilize by segregation to grain boundaries in NiAl superalloys and Ti[26,27] and hydrogen is believed to embrittle these same alloys. If surrogate elements with higher Z can be found for these behaviors, then 3D fluorescence tomography can provide nondestructive analysis of elemental segregation before, during and after fracture.

Nucleation, grain growth and texture evolution: Another place where the APS can transform engineering research is by providing new insights into nucleation, grain growth and texture evolution. Much of materials processing is related to attempts to control grain size, competing phases and texture in materials. Heat treating, rolling, extruding and other processing steps affect grain size, phase fractions and texture. Yet the basic driving forces for grain nucleation, growth and texture evolution remain controversial and largely untested. Recent synchrotron-based research has begun to address this area and suggests a path forward toward greatly enhanced understanding of these critical phenomena. For example, Budai et al. [28] have used polychromatic microdiffraction to study 3D grain growth with submicron spatial resolution and sensitivity to small angle grain boundaries that would be difficult to detect by other means. Their results support a key role of defect density in grain boundary evolution (grain refinement).

Similarly Poulson et al.[29,30] have studied texture evolution and grain growth using a high energy diffraction microscope that is functionally equivalent to the microscope on 1-ID. They find rough agreement between the simple Sachs and Taylor models for texture evolution, with discrepancies arising near crystallographic symmetry lines. These discrepancies may arise from near neighbor effects, but further research is needed. In addition, they find highly discontinuous growth of grains that is not explained by current models. Work at 1-ID will systematically study grain growth in a series of samples starting with high purity and progressing toward higher alloying element concentrations. This work should shed light on recent theory as well as the role of grain boundary segregation in determining the nature of boundary motions.

These pioneering studies will become even more useful with improved spatial resolution and with larger sample volumes. These competing needs demand fast x-ray sensitive area detectors, powerful data analysis software and high performance computational facilities as well as advanced x-ray optics.

Transient and evolving phases: Transient and evolving phases are everywhere and play a major role in engineering materials. For example, surfaces exposed to O_2 develop oxides that can either be tenacious or fragile. In-situ measurements at the APS have recently revolutionized our understanding of oxide coatings on high-temperature steels (Fig. 3). This is only one small example of the general corrosion issue which represents a huge cost to our developed infrastructure and costs about 3% of the GDP/year[31,32]. High-temperature oxidation is a particular issue in energy generating parts for coal, gas and nuclear power plants.[33] Stress corrosion cracking is a similarly important problem in nuclear and oil industries.



Transient tensile stress in Fe-Cr-Al-Y

Fig. 3. Oxide stresses in a high-temperature steel at temperature. Because the lattice expands during oxidation, the oxide coating was assumed to be under compressive load. In-situ experiments at the APS found that the coating is actually in tension, which is critical for cracking and failure of the protective oxide coating.[ref]

Similarly thermal processing is essential to develop multiphase materials microstructure needed to combine strength and ductility. Yet the evolution of phase fractions is only now being investigated by real-time measurements.[34,35] Recent measurements of steel components during heat-treating can follow the evolution and dissolution of phases during processing. This new information can test theories and help clarify paths toward ideal microstructures.

Boundary layer phases and buried interfaces

An area of almost unexplored potential is the study of minor phases that develop at the boundary layer of engineering materials. For example, in nuclear cladded fuels, a rich progression of phases can exist at the fuel/cladding boundary. Similarly in fiber-reinforced composites, minor phases at the fiber/matrix boundary can significantly enhance or degrade performance. Finally, knowledge of phase stability at the electrode/electrolyte interfaces in fuel cell stacks is crucial for assessing performance and durability of these systems. The high brilliance of the APS can be used to perform powder and single crystal diffraction on very small volumes, without the need to section the sample. Improved spatial resolution and new in-situ capabilities that simulate realistic operating conditions can target key issues including oxide layer evolution in thermal-barrier coatings[36], deformation and thermal evolution during tribological contact of low-friction coatings and depth-resolved measurements throughout the anode, electrolyte and cathode layers of operating fuel cells.[37] New instrumentation however is needed to make such measurements practical.

Two specific examples:

1. *Lightweight materials*

Lightweight materials for automative applications is a topic of critical national importance that can greatly benefit from new and emerging capabilities at the APS. The Energy Independence and Security Act of 2007, SEC. 651. LIGHTWEIGHT MATERIALS RESEARCH AND DEVELOPMENT calls upon the DoE to conduct research "to determine ways in which the weight of motor vehicles could be reduced to improve fuel efficiency" and specifically mentions the introduction of new Al alloys and high strength steels.

Transportation consumes 69 % of the petroleum used in the U.S. and is responsible for 33 % of all U.S. CO_2 emissions. *Reducing this dependence upon non-domestic oil is the most critical factor in achieving U.S. energy independence and energy security.* Attention has largely focused on alternative fuels and power systems; however, regardless of which engine technology ultimately prevails, the weight of a vehicle plays a major role in its energy efficiency. A recent study by the National Research Council[38] identified weight as the most critical factor in automobile fuel efficiency and showed that reducing the weight of automobiles and light trucks by just 10 % would reduce U.S. oil imports by approximately 8.5 %. Reducing the vehicle body weight by substituting lighter-weight materials such as aluminum alloys and high-strength steels is particularly effective since a lighter body requires a less massive suspension, transmission, engine, etc., thus multiplying the

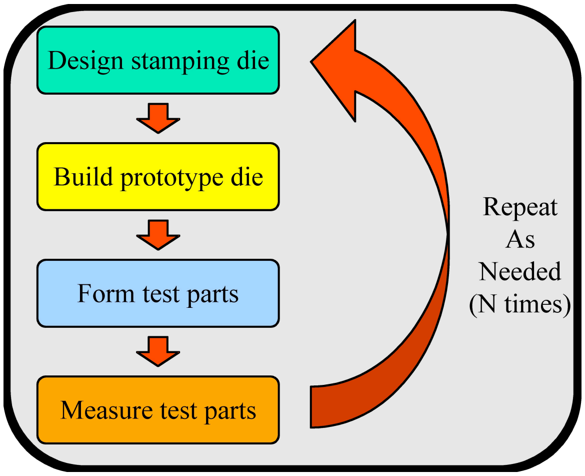


Fig. 4. Because deformation "spring-back" cannot be predicted, dies must be iterated until they form the correct shaped part. Although this process is manageable with soft steels, the iteration process is unacceptably long for high-strength steels and low density alloys that could drastically reduce the weight of vehicles.

weight reduction without compromising vehicle safety.

Large-scale introduction of advanced lightweight alloys is limited by their complex behavior during production. The central technical issue is the inherent multiscale nature of materials behavior, where small length-scale processes govern the behavior at the macroscopic level. The most intractable problems reside at the mesoscopic length scale where long- and short-range interactions between millions of mobile and immobile dislocations produce complex dislocation structures that are responsible for the observed macroscopic behavior (see Fig. 4). As shown in Fig. 7. Synchrotron X-rays are uniquely well suited to studies of dislocation structure evolution since they can penetrate deeply into macroscopic samples and provide quantitative measurements from sample volumes smaller than the size of typical dislocation structures. The APS is already at the forefront of research in this field with groundbreaking studies published using depth resolved microbeams on sector 34-ID and high-energy, high-angular resolution diffraction on sector 1-ID. If the APS is to retain their leading position in this field and continue producing the groundbreaking research results that drive engineering advances, investments are required in facility upgrades such as new high-performance focusing optics, area detector arrays and real time data analysis using parallel processing software and hardware. These upgrades would provide higher spatial resolution, faster data acquisition, and real time access to data allowing the researchers to make informed decisions during their experiments, all critical factors for successful research in this field.

2. *Cement*

Cement is another example of the critical importance of the APS. Cement has a global environmental impact through the large amounts of carbon dioxide released during production. Nevertheless, cement finds wide-spread use in concretes, mortars and grouts and more Portland cement is produced than any other man-made material. Synchrotron X-ray techniques, including diffraction, small angle scattering and imaging are essential for studies examining the early hydration of cements to understand how mechanical property development relates to the mix chemistry and how the long term degradation of cement based materials can be better controlled. Access to high energy x-rays is essential for cement studies of samples that are large enough to represent "bulk" material and to make use of special sample environments.

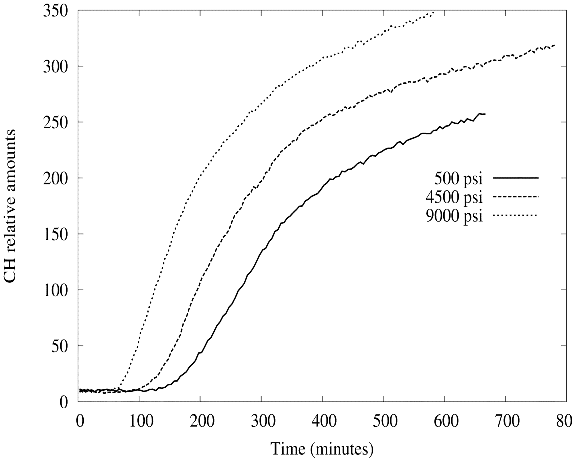


Figure 5. a) Kinetics for the formation of $Ca(OH)_2$, the major crystalline hydration products of Portland cement at low temperature, as slurries are hydrated at different pressures. Pressure dramatically reduces the observed induction time

Studies of cement grouts illustrate the importance of sample environment. Cement grouts (slurries) are used to seal the space between the bore holes in oil/gas wells and the steel pipe that is placed in the bore hole. As oil wells can go to great depths, these cement slurries can experience extreme pressures (> 1 kbar in some cases) and temperatures during their hydration. The hydration process and final properties of the cement are controlled by the use of a wide variety of additives to the cement slurry. High energy synchrotron scattering offers a powerful window for examining the crucial early hydration stage of these slurries under realistic oil well conditions. 30 – 60 keV radiation from APS bending magnets has already been used for studies at pressure of up to 600 bar and show dramatic effects of pressure on cement chemistry (see Figure 5).

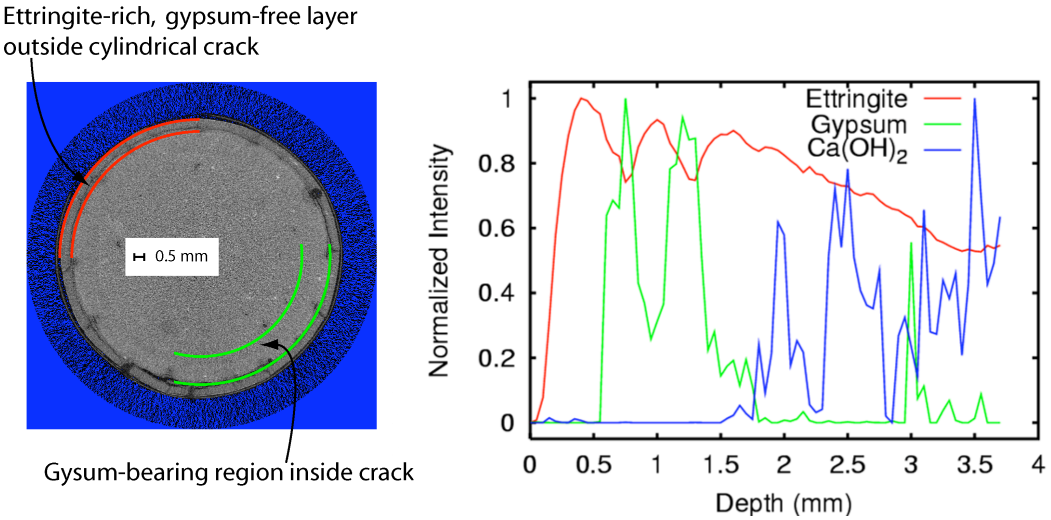


Figure 6. (left) Microtomograph of a cement paste cylinder that has been exposed to sulfate solution showing subsurface crack formation. The corresponding depth profile for some of the crystalline phases in the same specimen (right), generated from EDXRD data.

Environmental degradation of cement-based materials is another area where APS capabilities can clarify the underlying science. Cement-based materials are subjected to a wide variety of chemical agents that accelerate degradation and reduce service lifetime. This is a significant economic and environmental issue. Synchrotron radiation can be used to both image microstructural changes in specimens and generate corresponding maps or depth profiles of how crystalline phases are distributed within the specimens leading to a better understanding of damage mechanisms (see Figure 6). High energy synchrotron methods complement more traditional analytical tools as experiments can, in many cases, be done nondestructively and without concern over artifacts arising from specimen preparation. Early phase mapping experiments at the APS made use of energy dispersive diffraction capabilities at 1-ID, which are presently not available but would be with the proposed high-energy bending magnet

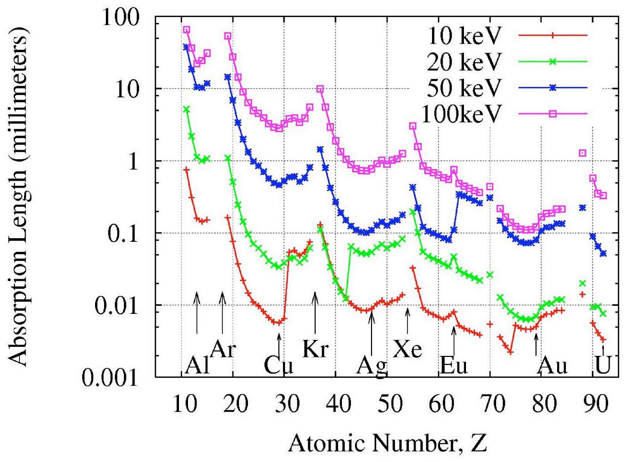


Fig. 7 Absorption length in millimeters as a function of atomic number, Z, for 10, 20,50 and 100 keV x-rays.

beamline. Furthermore, higher spatial resolution studies could be conducted on these materials using high energy diffraction microscopy.

4. Significance of APS

Among sources in the Americas, the APS is uniquely suited for engineering applications and applied research. The ***penetration depth*** of x-rays scales roughly as λ^{-3} and only hard x-rays (short wavelengths) can travel millimeters to centimeters through samples for studies of bulk properties and processes (Fig. 7). In addition, a minimum wavelength is required for access to a reasonable volume of reciprocal space and hard x-rays have low absorption but modest real-scattering factors for phase contrast imaging applications on low Z, mixed Z, or for samples inside environmental chambers. These factors make hard x-rays essential for engineering applications and applied research.

With this understanding, ***high-energy x-ray brilliance*** is the figure of merit for virtually all engineering and applied experiments. The APS is uniquely brilliant at hard x-ray energies and will remain the premier U.S. source for high-energy x-rays for the foreseeable future. For example, the deflection parameter, $K=0.934B_0(T)\lambda_u$, of an undulator is a unitless constant that depends on the undulator period, $\lambda_{u,}$ and on the maximum magnetic field strength B_0, but is independent of the electron energy. To compare the performance of a state-of-the-art undulator on two rings, it can be assumed that the minimum period and maximum field strength are about the same. With this assumption, the maximum K and minimum undulator period are fixed by technology. As shown in Fig. 1, at high energies, brilliance for a comparable undulator on the APS is significantly above even that of the ultra-bright NSLS II. One figure-of-merit, exception is for high-energy full-field imaging (radiograph/tomography) and energy-dispersive measurements, where a high flux and large beam size are often desired. Due to the high APS ring energy, bending magnet beamlines provide a unique source nationwide for such studies.

5. Scientific Community

The scientific community that will use these facilities is immense.

1. Leading academic institutions include U. Michigan, Northwestern, Carnegie-Mellon, Cornell, U. Tennessee, UIUC, Vanderbilt, M.I.T, Stanford, Georgia Tech., and SUNY.
2. Industrial laboratories including Alcoa, Ford, Chevrolet, Dodge, Boeing, G.E., John Deere, Martin-Marietta, Pratt and Whitney.
3. Government laboratories belonging to DOE, DOD, NRC, NASA, NIST, DARPA, Navy, Air Force, and Army.

6. Requirements and Capabilities

X-ray Optics: As described above, spatial resolution is a key driving force for engineering applications and applied research and the ability to study increasingly small sample volumes will continue to revolutionize engineering and applied research into the forseeable future. In order to lead in this area, the APS beamlines must have access to the world's best focusing and imaging optics. Past progress at the APS has resulted in promising x-ray optics with demonstrations a record smallest one-dimensional hard x-ray focusing device[39]- a Laue Zone plate lens with 16 nm FWHM- and demonstration of the world's smallest polychromatic probe, a Kirkpatrick-Baez total-external-reflection mirror system with 80 nm-D probe size.[40] Yet these optics are not widely available at the APS ***and a major effort is needed to approach these performance numbers routinely and for x-rays above 40 keV***. Indeed, stable optical environments require new levels of engineering to achieve the promise of emerging optics. Major efforts at the ESRF have produced sub 50 nm achromatic optics and a tremendous program at Osaka/Spring-8 has recently set a new record for the smallest hard (20 keV) x-ray beam at 15 nm. Refractive-based optics provide efficient, source-size limited focusing of monochromatic high-energy x-rays to the µm-level, with relatively low divergence as required for high q-resolution studies of e.g. strain.

Detectors: There is no question that advanced x-ray sensitive area detectors can revolutionize synchrotron science in general and engineering applications and applied research specifically. This is an area that is rapidly developing with dedicated synchrotron-driven area detectors under development around the world, and with the continued evolution of more powerful area detectors driven by electron science and visible light cameras. In the near term, ultra fast detectors with readout times of 100-10000 frames-per-second, fps, will efficiently use intense x-ray beams and extend 3D microscopy methods. Such cameras will also allow for monitoring of increasingly short-lived transient phases, and stresses.

Also over the next 5 years, x-ray cameras will become available with ~1% energy resolution/pixel. These cameras will greatly improve signal-to-noise in virtually all classes of diffraction experiments, will resolve orders in reflections and will revolutionize fluorescence spectroscopy including the development of differential aperture methods to accelerate fluorescence spectroscopy. X-ray sensitive area detectors with crystal-spectrometer-class energy resolution (0.01%) will further revolutionize synchrotron science in a number of ways that will directly impact engineering applications and applied research. Two clear examples- out of the numerous possibilities- are for polychromatic microdiffraction and for grain-average challenged powder diffraction.

In polychromatic microdiffraction, a broad bandpass beam intercepts a small crystal volume and generates a characteristic Laue pattern. The angles between the Laue reflections determine the average crystallographic orientation of the sample volume and the deviatoric strain (distortion of the unit cell shape). This information is incredibly useful, but even more information can be recovered if the energy spectra of each pixel in the area detector pattern can be analyzed. For example, the average energy spectra of even one Laue spot will determine the unit cell volume and therefore the hydrostatic strain of the unit cell. More practically, in virtually all engineering materials, Laue spots are streaked by dislocations and strain gradients. With energy resolved area detector patterns, the signal-to-noise of the pattern is vastly improved and a detailed map of plastic and elastic strain is possible including diffuse scattering (sensitive to defects) that would be difficult to resolve with an energy integrated map.

Similarly in powder diffraction, as the sample volume decreases, particle statistics become insufficient for high-precision measurements of lattice parameters and strains. In particular, reflections are nearly specular with respect to the lattice planes and a grain even off the ideal Bragg angle will scatter through the Lorenzian tail of the reflection. Because sample volumes with low particle statistics often do not include grains oriented near the ideal Bragg condition, a false peak position can be observed. If however, a white beam is used to probe the small sample volume, there are ~10^2-10^4 more chances for grains to satisfy the Bragg condition. The particle statistics problem is therefore vastly improved and the overlapping Debye rings from the different energies can be separated according to the encoded energy at the detector.

Finally, high spatial resolution cameras will improve resolution for tomography and remove some of the burden on point probe techniques that are inherently slow compared to full field imaging of samples. For example, with more efficient cameras and more optimized insertion devices 100 ps exposures are possible at the APS for radiography and tomography on "few millimeter" thick steel samples. With very high frame rate cameras (1 Mhz) this opens up the potential to track the evolution of a crack propagating in excess of 1 km/s.

With renewal funding, the spatial resolution available with transmission x-ray microscopy can also be greatly improved and the technique can be pushed to much higher energies. This is an area that will require combining advanced x-ray optics and special small pixel detectors to push resolutions with hard x-rays. Existing developments have achieved ~40 nm resolution, but only for long wavelengths. The promise of this technology is indicated in Fig. 8, which shows a reconstruction of a solid oxide fuel cell interface including the grainy Ni anode in a matrix of Yttrium-stabilized zirconica and electrolyte.

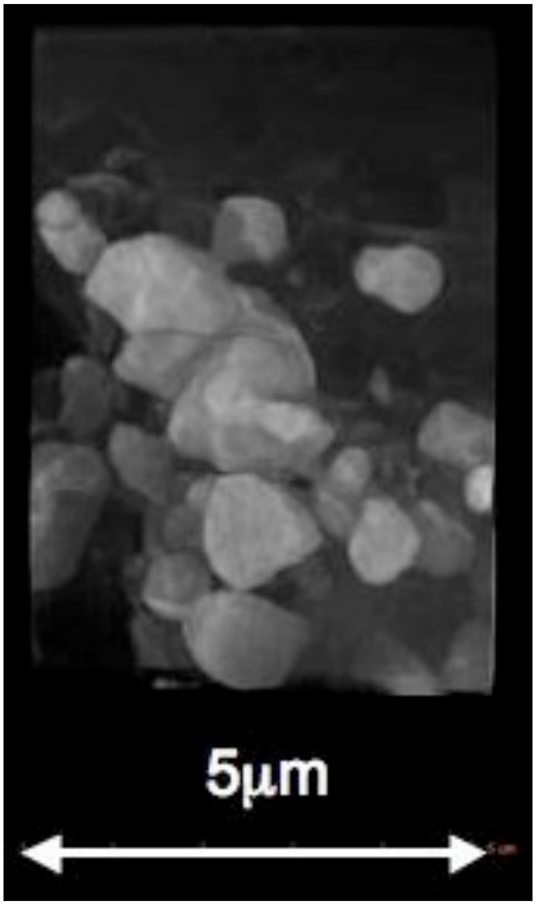


Fig. 8. Transmission x-ray tomographic image of a solid-oxide fuel cell volume.

Software and experimental control: Although instrumentation builds experimental capabilities, beamline control and data analysis software is the key to widening the experimental community and is a ***deciding factor in facility productivity***. Some techniques lend themselves to user-friendly operations. For example, absorption tomography, powder diffraction, EXAFS, and SAXS are comparatively easy to automate and users can be efficiently trained. Other experiments are more difficult to automate, and may require data analysis that is still evolving. For example, high energy diffraction microscopy and polychromatic microdiffraction are emerging methods with no history of data acquisition or analysis software. It is essential that high performance computations are accessible during data collection so that users see microscope output in time to guide further processing steps. The enormous potential of these methods is challenged by the need to develop user-friendly data acquisition and analysis packages.

Undulators: The 7 GeV electron energy of the APS makes possible ultra-brilliant high-energy undulators that are simply not possible at any other source in the Western Hemisphere. Indeed specially optimized undulators can vastly improve beamline brilliance and flexibility, at high energies. Canted undulators offer the prospect of individual undulator control on stations that would otherwise be forced to work with compromised shared spectral qualities. Consider for example the energy spectra above ~20 keV. Fig. 1 compares the theoretical performance of an undulator on the APS, to the calculated performance of a similar device on the NSLSII. As can be seen, an optimized undulator on the APS can deliver about an order of magnitude more brilliant x-rays above 20 keV, and roughly 3 orders of magnitude greater brilliance over 40 keV.

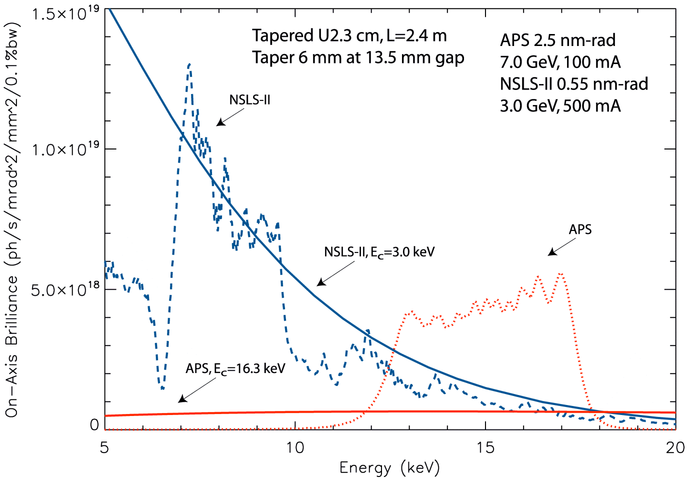


Fig. 9. On-axis brilliance for the *tapered* undulator U2.3 cm (APS23#1) installed on the APS and the NSLS-II storage rings. The average undulator gap is about 13.5 mm and the gap taper is about 6 mm over the full length of the device (2.4 m). The critical energies for two cases are labeled. The solid lines show the wiggler approximation using the corresponding critical energies. Note the much higher brilliance over the range of 12-18 keV on the APS.

For polychromatic microdiffraction, there are also important opportunities to optimize the source properties. Polychromatic microdiffraction utilizes broad-bandpass radiation and studies materials behavior with submicron to nanometer resolution. The geometry pioneered at the APS has the detector at 90° to the incident beam path which allows for good spatial resolution in all three dimensions. X-ray beams of around 12-18 keV are favored by this geometry; this energy range allows for many Laue spots, with reasonable integrated reflectivity in the high energy reflections. At lower energy and narrower bandpass, there are too few reflections for general purpose characterization of materials. At higher energy, the integrated reflectivity falls rapidly and a transmission geometry is favored, which reduces spatial resolution along the beam. There are interesting possibilities of moving to more penetrating high-energy beams for polychromatic microdiffraction, but even in the 12-18 keV regime, the APS offers major advantages for polychromatic microdiffraction. As illustrated in Fig. 9, even in the 12-18 keV range, the bandpass-integrated brilliance (photons/sec/mm^2/mrad^2) is about 5 times higher for an undulator on the APS than for a similar device on the NSLSII. Indeed, tapering an undulator, working on or working off axis can all broaden the undulator spectra and allow for spectra with ~2-3 orders of magnitude more brilliance than from 2nd generation sources but with wide bandpass.

Specific recommendations:

1. Dedicate an additional beamline to high-energy x-ray measurements of aggregate stress/strain/texture from materials. This should include state-of-the-art ancillary equipment for sample environments and mechanical testing. Complementary x-ray imaging and small-angle scattering capabilities should be integrated into the beamline for simultaneous measurements. The current aggregate materials program on 1-ID will be moved to this new beamline. For more information, see: www.aps.anl.gov/Renewal/Proposals/1-ID_upgrade.pdf.

2. Complete the 1-ID Phase II upgrade. With the completion of #1, this upgraded beamline will have a dedicated high-energy diffraction microscope (HEDM) station and a dedicated station for high-energy microfocused PDF and powder diffraction experiments. The HEDM station will include tomography capability. For more information, see: www.aps.anl.gov/Renewal/Proposals/1-ID_upgrade2.pdf.

3. Build a dedicated polychromatic nanoprobe hutch and install a canted undulator pair to allow for simultaneous and independent use of polychromatic mesoscale and nanoscale probes. Station 34-ID-E is instrumented with the only 3D diffraction microscope in the world with submicron spatial resolution. To meet the already overwhelming and growing demand for beamtime, the coherent diffraction station should be relocated to a long beamline where it can benefit from larger sample to source distances and the polychromatic microprobe should be given a dedicated canted undulator. The canted undulators will be optimized as described above to produce ultra-high brilliance beams between 12-18 keV. A second canted undulator would be dedicated to a nanoprobe station that can be located beyond the current microprobe station. This nanoprobe would have an initial polychromatic focal spot of ~40 nm and with further development beams of ~15- 20 nm should be achievable in the near future.

4. Optimize *at least* one bend-magnet beamline for high-energy energy-dispersive diffraction. As noted throughout the document, the APS is well suited to produce high-energy x-rays. Compared to ID beamlines, bending-magnet beamlines have received little attention for high-energy development despite their strong potential. More information can be found at http://www.aps.anl.gov/Renewal/Proposals/HE_BM_midterm.pdf.

5. Develop a dedicated high-energy tomography station with phase contrast sensitivity. This station could be developed in addition to, or integrated with, the above beamline.

6. Develop a range of environmental chambers for powder diffraction and SAXS targeted for catalysis, in-situ thermomechanical loading and other applied studies.

7. ***Develop user friendly "expert" software for all engineering stations that insures users walk away from experiments with data sufficiently processed for analysis at their home institution***. This will require an investment in computational infrastructure, both in the form of personnel and hardware. Data need to be automatically pipelined to appropriate machines for analysis during data collection. In some cases, this may require getting data through firewalls to TeraGrid machines or other off-site systems.

8. Co-ordinate combined techniques/characterizations to follow materials evolution.

9. Develop user-friendly mail-in capabilities in anticipation of the growing difficulty of travel with high-energy costs.

7.0 References

1. J. S. Chung and G. E. Ice, “Automated Indexing for Texture and Strain Measurements with Broad-Bandpass X-ray Microbeams,” *J. Appl. Phys*. **86**, 5249-5256 (1999).

2. G.E. Ice, "3D Micron-Resolution Laue Diffraction" pg. 353 *Neutrons and Synchrotron Radiation in Engineering Materials Science*, Ed. W. Reimers, A.R. Pyzalla, A. Schreyer, H. Clemens, Wiley-VCH Weinheim Germany (2008).

3. B. C. Larson, W. Yang, G. E. Ice, J. D. Budai, and J. Z. Tischler, “Three-Dimensional X-Ray Structural Microscopy with Submicrometre Resolution,*” Nature* **415**, 887 (2002).

4. H.F. Poulsen, Wolfgang Ludwig and S. Schmidt, "3D X-ray Diffraction Microscope", pg. 335 *Neutrons and Synchrotron Radiation in Engineering Materials Science*, Ed. W. Reimers, A.R. Pyzalla, A. Schreyer, H. Clemens, Wiley-VCH Weinheim Germany (2008).

5. B. Jackobsen, H. F. Poulsen, U. Lienert, J. Almer, S.D. Shastri, H. O. Sorensen, C. Gudlach, HF Poulsen, W. Pantleon, "Formation and subdivision of deformation structures during plastic deformation", *Science* **312** 889-892 (2006).

6. A. Wanner and D.C. Dunand, "Synchrotron X-ray Study of Bulk Lattice Strains in Externally-Loaded Cu-Mo Composites", Metallurgical and Materials Transactions **31**A, 2949-2962 (2000). Copyright: TMS, Warrendale, PA. and ASM International, Materials Park, OH., 2000.

7. A.A. Kulkarni, H. Herman, J. Almer, U. Lienert, and D. Haeffner, "Depth-resolved porosity investigation of EB-PVD thermal barrier coatings using high-energy X-rays", *J. Am. Cer. Soc.* **87** 268 (2004).

8. J.-S. Park, P. Revesz, A. Kazimirov, and M. P. Miller, "A methodology for measuring in situ lattice strain of bulk polycrystalline mateial under cyclic load", *Rev. of Sci. Inst*. **78** 023910 (2007).

9. Neutrons and Synchrotron Radiation in Engineering Materials Science, Ed. W. Reimers, A.R. Pyzalla, A. Schreyer, H. Clemens, Wiley-VCH Weinheim Germany (2008).Wake field plastic NSLS

10. M. Croft, N. Jisrawi, Z. Zhong, K. Horvath, R.L. Holtz, M. Shepard, M. Lahshmipathy, K. Sadananda, J. Skaritka, V. Shukla, R.K. Sadangi and T. Tsakalakos, "Stress gradient induced strain localization in metals: High resolution strain cross sectioning via synchrotron x-ray diffraction", *J. of Eng. Mat. and Tech.* **130** 021005 (2008).

11. M.R. Daymond, M.L. Young, J.D. Almer, D.C. Dunand, "Strain and texture evolution during mechanical loading of a crack tip in martensitic shape-memory NiTi", *Acta. Mat.* **55** 3929-3942 (2007)

12. R.I. Barabash**,** Y. Gao, Y. Sun, S.Y. Lee, H. Choo, P. Liaw, D. Brown, G. Ice, “Neutron and X-ray diffraction studies and cohesive interface model of the fatigue crack deformation behavior” *Philosophical Magazine Letters* (2008) **88**, No.8, 553-565.

13. P.M. Scott, *Corrosion* **56** 771 (2000) and http://www.esrf.eu/news/spotlight/spotlight66

14. Kerr, M., M.R. Daymond, R.A. Holt, and J.D. Almer, “Strain evolution of zirconium hydride embedded in a Zircaloy-2 matrix”. *Journal of Nuclear Materials*, **380**(1-3), 70-75 (2008).

15. L.E. Levine, B.C. Larson, W. Yang, M.E. Kassner, J.Z. Tischler, M.A. Delos-Reyes, R.J. Fields, W.J. Liu, "X-ray Microbeam measurements of individual dislocation cell elastic strains in deformed single-crystal copper", *Nat. Mat*. **5** 619-622 (2006).

16. J.W.L. Pang, G.E. Ice, and W. Liu, "The role of crystal orientation and surface proximity in the self-similar behavior of deformed Cu single crystals", Submitted to *Ultramicroscopy* (2008).

17. T. Ohashi, R.I. Barabash**,** J.W.L. Pang**,** G.E. Ice, O.M. Barabash, “X-ray Microdiffraction and Strain Gradient Crystal Plasticity Studies of Geometrically Necessary Dislocations Near a Ni Bicrystal Grain Boundary”, *International Journal of Plasticity*, (2008), doi:10.1016/j.ijplas.2008.04.009.

18. J.W.L. Pang, R. Barabash, W. Liu and G.E. Ice, " Determination of Deformation Inhomogeneity in Polycrystalline Ni under Uniaxial Tension with 3D X-ray Microscope", *TMS Letters*, **1** 5 (2004).Pang polycrystals

19. B.C. Larson, A. El-Azab, W. Yang, J. Z. Tischler, W. Liu, and G. E. Ice, “Experimental Characterization of The Mesoscale Dislocation Density Tensor,” *Philosophical Magazine*, **87**, 1327-1347 (2007).

20. D.R. Haeffner, J.D. Almer, U. Lienert, "The use of high-energy X-rays from the Advanced Photon Source to study stresses in materials", *Mat. Sci. and Eng. A* **399** 120-127.

21. H. F. Poulsen, J.A. Wert, J. Neufeind, V Honkimaki and M. Daymond, *Nature Mater.* **4** 33-36 (2005).

22. Hufnagel, T.C., R.T. Ott, and J. Almer, “Structural aspects of elastic deformation of a metallic glass”. *Physical Review B* **73**(6): p. 64204-1-8 (2006).

23. R. Wang, H.W. Chen, Z.L. Lu, S.H. Qiu and T. Ko, "Structural transitions during aluminium leaching of $NiAl_3$ phase in a Raney Ni-Al alloy", *J. of Mat. Sci*. **43** 5712-5719 (2008).

24. J.M. McNaney, et al., “High-Pressure, Laser-Driven Deformation of an Aluminum Alloy,” *Mettal. Trans. A*, **35A**, 2625–2631 (2004).

25. B.A. Remington, et al., “Materials Science under Extreme Conditions of Pressure and Strain Rate,” *Mettal. Trans. A*, **35A**, 2587–2608 (2004)

26. E.P. George, R.L. Kennedy, D.P. Pope, "Review of element effects on high-temperature fracture of Fe0 and Ni-base alloys", *Phys. St. Solidi A-Applied Res*. **167** 313-333 (1998).

27. E.P. George, C.G. McKamey, E.K. Ohriner, E.H. Lee, "Deformation and fracture of iridium: microalloying effects", *Mat. Sci. and Eng. A* **319** 466-470 (2001).

28. J.D. Budai, *Mat. Sci. Forum* **467**, 1373-1378 (2004).

29. L. Margulies, G. Winther, H.F. Poulsen, "In situ measurement of grain rotation during deformation of polycrystals", *Science* **291** 2392-2394 (2001).

30. H.F. Poulsen, L. Margulies, S. Schmidt and G. Winther, "Lattice rotations of individual bulk grains-art I: 3D X-ray characterization", *Acta Materialia* **51** 3821-3830 (2003).

31. http://www.nace.org/content.cfm?parentid=1011&currentID=1045

32. Materials and Coatings to Resist High Temperature Corroision, Ed. D.R. Holmes and A. Rahmel, Applied Science Publishers ISBN 085334784 (1978).

33. S.M. Bruemmer, G.S. Was, *J. Nuclear Materials* **216**, 348 (1994).

34. S. S. Babu, E. D. Specht, M. L. Santella, G. E. Ice, and S. A. David, “In-Situ Observations of Oxidation and Phase Stability in Cast Nickel-Based Intermetallic Alloys,” *Metall. Mater. Trans. A* **37A**, 195 (2006).

35. H.J. Stone, M.J. Peet, H.K.D.H> Bhadeshia, P.J. Withers, S.S. Babu, and E.D. Specht, "Synchrotron X-ray Studies of Austenite and Bainitic Ferrite," *Proc. R. Soc. London,* Ser. A **464** 1009-1027 (2008).

36. Almer, J., E. Ustundag, G.A. Swift, J.A. Nycha, C.C. Aydiner, and D.R. Clarke, *In situ synchrotron measurements of oxide growth strains. Materials Science Forum* **490-491**: p. 287-293 (2005).

37. Almer, J.D., D.J. Liu, B. Harder and C. Weyant, “High-energy X-ray measurements of layered systems”, *Proceedings of the 29th Risoe Symposium on Materials Science: Energy Materials*, p. 207-214 (2008).

38. National Research Council

39. H.C. Kang, J. Maser, G.B. Stephenson, C. Liu, R. Conley, A.T. Macrander, S. Vogt, " *Phys. Rev. Lett*. **96** 127401 (2006).

40. W.J. Liu, G.E. Ice, J.Z. Tischler, A. Khounsary, L. Assoufid, A.T. Macrander, "Short focal length Kirkpatrick-Baez mirrors for a hard x-ray nanoprobe", *Rev. Sci. Inst*. **76** 113701 (2005).