Hard Condensed Matter and Materials Physics

A white paper on science requirements for NSLS and NSLS-II

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1. Summary Overview

The workshop on Science Requirements for Hard Condensed Matter and Materials Physics took place at Brookhaven Lab on February 5th and 6th of 2008. Approximately 75 individuals representing 25 institutions were in attendance. The workshop presentations surveyed science directions relevant to 3rd generation light source, and those directions serve as the foundation for this report. These science directions span most of the current problems in condensed matter physics: nanomaterials, ferroelectrics, multiferroics, graphene and 2D electron systems, magnetism, spin transport and dynamics, strongly correlated electron systems, superconductivity, competing orders, quantum criticality, nanoscale inhomogeneity and disorder, photonic bandgap and meta-materials, organic conductors, etc.

Much of the research strives to understand the relationship between electronic properties and a material's structure, and synchrotron radiation serves as both a spectroscopic and structural probe of materials. In particular, this community utilizes the very wide range of synchrotron radiation photon energies to explore behavior from transitions involving core electrons to pair breaking in low temperature superconductors. The wavelength range extends from atomic dimensions out to the macroscopic dimensions associated with electrical transport, and corresponds to momentum transfer spanning the entire Brillouin zone for most crystalline solids.

One of the forefront activities in condensed matter research is to understand the nature of materials under extreme conditions. The creation of such conditions in the laboratory usually involves small volumes or restricted sample geometries where optical throughput is an issue. High brightness synchrotron radiation overcomes these throughput limitations. Another activity is the science of spin dynamics, especially spin-carrier interactions in layers, nano-structured and synthetic materials with an eye on applications in spintronics.

This report takes into account that other workshops have focused on specific synchrotron radiation measurement techniques and beamlines that are expected to play a significant role in the study of hard condensed matter and materials, and details on those techniques and scientific drivers can be found in the reports for those particular workshops. We naturally expect that NSLS-II beamlines will include the standard spectroscopic and scattering/diffraction methods available at most synchrotron radiation facilities. Though we have developed a list of beamline techniques that this community recommends for development at NSLS-II, a detailed prioritization in terms of schedule and cost is beyond the scope of this particular workshop report. We have chosen instead to include source and facility requirements that are not necessarily beamline-specific, and in some cases their development can begin at the existing NSLS. Our recommendations fall into 3 general categories: a) endstation development to enhance our ability to study materials in strong magnetic fields and other extreme environments, b) considerations for producing transient non-equilibrium conditions and utilization of the NSLS-II source time-structure for studying dynamics and c) the development of unified software for beamline control, data collection and analysis, especially in light of the large quantity of data that results from many imaging-type measurements. Programs for a) and c) can be started at NSLS.

2. Scientific Community and Research Themes

The hard condensed matter and materials physics communities provide the research foundation for the discovery and fundamental understanding of new materials. It consists mostly of researchers in university and national laboratories where the goal is to explain the electronic properties of materials and their relationship to material structure as well as the degrees of freedom expressed in thermodynamic variables. This has been a reasonably stable community for synchrotron radiation research, yet has the potential for growth as new material structures are created in the laboratory. Technologically relevant materials are identified for development by the applied sciences and engineering communities that exist in both academic and industrial laboratories. Thus, the growth of industrial involvement follows from investments in basic condensed matter research.

In many cases, the electronic behavior of a material results from a complex interplay or competition among various excitations. The competition between different ordered states can drive structural changes in a material, cause intrinsic inhomogeneity, or lead to dynamical conditions … any of which could give rise to new electronic phenomena. Exploring this complexity has become practical with the development of a variety of synchrotron radiation methods and instruments. We outline some of the research activities where synchrotron radiation is a well established tool and is likely to play an important role over the next 10 years.

2a. Magnetism, spin transport and spin dynamics.

The spin of an electron gives rise to a host of magnetic behaviors in solids: ferromagnetism, antiferromagnetism and ferrimagnetism being examples where electron spins are ordered on a macroscopic scale resulting in domains. Present interest is focused on the manner in which domains grow (morphology) and change orientation. Imaging methods (*i.e.* LEEM/PEEM, scanning & full field transmission x-ray microscopy and coherent diffraction imaging), particularly when combined with polarization selection (*e.g.* linear vertical, linear horizontal, circular) among the synchrotron methods for studying domain morphology. Such microscopic techniques are ideally suited to high brightness sources and they will be invaluable in understanding many phenomena at the nanoscale, including spin transport and domain wall motion in artificially patterned nanostructures. Spin dynamics at the boundaries between magnetic and non-magnetic regions also affect electrical transport and the intrinsic potential speed of read/write heads in memory devices. The basic mechanism for ferromagnetism in dilute magnetic semiconductors is also not understood. Antiferromagnetism in metal oxides form the basis for a class of complex oxides, including the colossal magneto-resistance system La(Sr,Ca)MnO_3. Synchrotron radiation probes the local magnetic environment around each atom type through element-specific xray magnetic circular and linear dichroism (XMCD and XMLD). Many magnetic materials consist of compounds and alloys of diverse elements such that a wide range of x-ray energies is necessary to probe a large section of the periodic table. Spin dynamics can be probed using combinations of static and time-dependent magnetic fields. Characteristic time scales are determined by the spin relaxation conditions and the applied field strength. High frequency spin resonance can be measured in the very far-infrared with sufficiently strong magnetic fields ($B > 5T$). The local magnetization state for a material can be driven into oscillation while the amplitude and phase lag of the response followed by circular dichroism techniques. The amplitude and phase then provide details on coupling mechanisms between different magnetic moments, which is expected to shed light on several long-standing controversies involving loss mechanisms in mangon scattering. Time resolution is determined by the electron bunch length and is presently limited to around 100ps. The goal is to reach about 1ps.

2b. Ferroelectric and multiferroic materials

Ferroelectrics are systems that develop a spontaneous electric polarization below a critical temperature, usually associated with phonon softening in transition metal oxides. They are of great practical value and effort continues to find materials that can switch more quickly and at lower fields or with less loss. These materials also can have very high dielectric constants that may prove useful in electronic devices. A more recent discovery is materials that show both ferroelectric and ferromagnetic behavior. These *multiferroics* may allow one to control the magnetic state of a material through an applied electric field and vice versa. As with many systems where two ordered states compete and attempt to co-exist, questions about homogeneity remain. Synchrotron probes that sense the valence state, atomic positions in the unit cell, and the magnetic environment for each atom may clarify this issue.

2c. Low dimensional, nanomaterials, artificially structured material

Examples of nanomaterials include quantum dots, carbon nanotubes and graphene. These materials possess novel electronic and other properties, typically due to quantum effects and the relative importance of the surface relative to the bulk of the material. Magnetic quantum dots are expected to display dynamical properties different from their bulk counterparts and domain wall pinning and motion in magnetic nanowires is an active area of research. XMCD, soft x-ray microscopies and hard x-ray nanoprobes are relevant SR techniques. The band structure for graphene is especially interesting due to the nearly massless behavior of electrons at the Fermi energy. Small energy shifts can give rise to enormous changes in the electron (quasiparticle) properties. Like nanotubes, these carbon sheets may allow for the construction of novel layered materials (similar to intercalated graphite). The band structure can be probed by ARPES and other types of spectroscopies (infrared and magnetospectroscopy). Another system of layered materials is the diborides, which includes the two-band superconductor MgB_2. The competition between these two superconducting reservoirs may give rise to the so-called Leggett collective mode. Useful probes include IXS, ARPES and infrared spectroscopies.

2d. Correlated electrons, competing orders

The complex metal oxides have become an important laboratory for the study of strong electron correlations in solids, where the standard Fermi liquid and quasiparticle picture for electrons in solids breaks down. Regarded as doped Mott insulators, their properties depend on the type of metal oxide (cuprates, manganites, ruthenates, etc.) and vary strongly as a function of doping. In the case of the high-T_c cuprates, many aspects of their rich phase diagram are now reasonably well understood, however the role of magnetism and the nature of the pseudo-gap is still not clear. IXS is expected to play a major role by providing information on the q-dependent (momentum transfer) aspect of the electronic response (i.e., away from the q=0 limit of conventional optical spectroscopies). There is still ARPES work to be done in filling out as much of the Fermi surface as possible as a function of doping and temperature. The phase diagrams for other transition metal oxides also show a variety of phenomena, believed to be due to the competition between different types of ordering (charge, orbital and spin). This competition is believed to cause phase separation, where different regions adopt a particular order to lower their energy. These nanoscale inhomogeneities can be often detected by synchrotron methods such as XAS microscopy, XRD, diffuse x-ray scattering and measurements of the pair distribution function (PDF). Spin, orbital, and charge ordering can be detected with resonant x-ray scattering, particularly with incident beam polarization control and polarization analysis of the scattered photons. Like many systems in this class (including the heavy fermion materials), large single crystals can be difficult to grow, this (plus the need for very low temperatures) has been an impediment to their study. The high brightness of NSLS-II will be beneficial even for basic XRD of these materials.

2e. Materials in extreme environments.

The physical behavior of a material is a function of its thermodynamic condition, and most materials can show a rich phase diagram in terms of temperature and pressure. Being able to control these parameters allows one to explore a wide range of phenomena in a single sample and test theoretical models for fundamental issues such as the insulator-to-metal transition (as a function of inter-atomic or intermolecular spacing). Changing the environment can cause a system to switch to a different state, with the potential for revealing new phenomena (e.g., quantum critical points at low temperatures). Dynamical effects associated with a (sudden) change in environment also of interest from both a fundamental science viewpoint but also for practical reasons concerning components used in information processing (memory, processors). There are other practical implications in the areas of dielectric breakdown and the strength of materials. Extremes of pressure and temperature can be achieved in diamond anvil cells and their variants, and allow both x-ray scattering and optical / infrared spectroscopies to be performed. We mention it here and note that more detail can be found in the Materials Science and Engineering workshop report. Inelastic x-ray scattering is expected to become an increasingly important technique for covering a wide range of phenomena from energy bands and dispersion, to bandgap phenomena (e.g. excitons) and vibrational properties (phonons). Less has been done to address extreme field conditions. An electron’s spin will precess in an applied magnetic field and, depending on its environment, will change orientation assuming it is energetically favorable. This may involve the movement of domain walls in ferromagnetic materials, or a competing order (such as orbital order) in a complex metal oxide. Magnetic fields up to 15T can be achieved with superconducting coils, but higher fields are typically available only at specialized laboratories (e.g. NHMFL). Higher fields can be achieved using pulsed magnets, although the duty cycle is low so their use is limited to high sensitivity measurement techniques.

2f. Electronic materials

Most electronic devices consist of different materials arranged in layers to form things like the gate in an FET or the 2D electron gas region in a high electron mobility transistor. The layer interfaces are usually detrimental to performance (due to defects) but can sometimes be an advantage (mobility improvements due to strain in Si). New materials, such as organic crystals of rubrene and pentene, may serve as low cost replacements to silicon. The nature of charge transport (polarons, hopping) is a current subject for study. Synchrotron radiation tools will be needed to characterize existing semiconductor materials and interfaces as well as new materials. XRD will continue to be important for studying crystalline quality, but tools such as XPS and XAFS are necessary for studying details of defects in the bulk and at surfaces and interfaces (as used by NIST at their existing NSLS beamlines). Grazing incidence x-ray reflectance is also important for monitoring growth behavior. The Materials Science and Engineering workshop report has additional details, including beamline requirements.

3. Requirements for Hard Condensed Matter and Materials Physics

3a. Synchrotron Measurement Techniques

The multiple competing phenomena combined with spatial inhomogeneity in today's complex materials mandates that a wide range of spectroscopic and structural probes be available, often combined with the ability to probe at a microscopic scale and even produce images when inhomogeneity is a key feature. Thus the suite of synchrotron radiation beamline facilities needed for their study spans a broad range of measurement techniques. We list them here:

Spectroscopies: for probing energy scales from intraband transitions related to transport and cyclotron resonance (below 1 meV) through interband & valence electron transitions and on to the atomically specific core levels (keV). The combination of ARPES, IXS and conventional IR and optical spectroscopies provide a remarkable combination for exploring different facets of the same condensed matter system. Direct optical spectroscopies have high sensitivity and spectral resolution, can explore both crystalline and non-crystalline materials, but are typically limited to very small momentum transfer ($q \approx 0$). ARPES enables studies of a broader range of momentum space in crystalline materials and can be directly compared with theoretical calculations for band structure. IXS provides the ability to perform spectroscopy over a large range of momentum transfer ($q \neq 0$) and can be made sensitive to specific electronic arrangements through resonant scattering. Element specificity is also available from more conventional soft and tender x-ray spectroscopies involving core level transitions. Relevant spectroscopies include XAS/XANES. When combined with polarization control (*e.g.*, horizontal versus vertical linear, or left and right circular), sensitivity to the symmetry of charge distributions or the direction of electron spin is achieved. A wide range of photon energies are needed to span the majority of the periodic table, with special interest to the transition metals where the electron's spin manifests itself in a variety of magnetic phenomena (ferromagnetism, ferrimagnetism, antiferromagnetism).

Structural probes: Diffraction and scattering methods for sensing the local atomic environment in solids. Hard x-rays for diffraction and scattering to determine the average structure in novel crystals or evidence for ordering (including magnetic scattering in an applied field). The photon energy range necessary to cover all the relevant absorption edges can be very large. EXAFS and Pair Distribution Function (PDF) measurements to determine the local atomic structure and order, especially in inhomogeneous materials. X-Ray reflectivity of nanopatterened or layered materials in the soft x-ray and hard x-ray energies.

Microprobes and Imaging: Both spectroscopic and structural probes can sometimes be combined with microprobes or imaging methods to extract spatial information from a nominally heterogeneous system (e.g., as may occur in phase separation from competing orders). Useful in mapping the domain structure of magnetic and ferroelectric materials. LEEM/PEEM, scanning transmission x-ray microscopy (STXM) & full-field transmission x-ray microscopy (TXM), and both soft and hard x-ray coherent diffraction imaging (CDI) are two particularly relevant imaging techniques.

Themodynamic Environment: How a material changes when a thermodynamic parameter (such as temperature, pressure, electric field, magnetic field and particle number) is varied has always been an important tool for understanding materials. The range of interesting temperatures reaches down to 0.1K for the highly-correlated heavy fermion materials. Magnetic field strength over 20T is also needed. Pressures reaching into the multi-megabar range. Electric field strengths reaching to MV/cm and above. The particle number in a system can also be varied by chemical doping, by shifting the Fermi level in an applied field, or by creating particle-hole excitations with light (from a laser).

Dynamics (Ultrafast): When a thermodynamic parameter is varied as a function of time, the system responds and its dynamics can be studied – including the recovery of the system from a non-equilibrium condition. The next generation of light sources – based on linacs – will focus on the time range from 1 ps to ~10 fs. Storage ring sources nominally reach to about 100 ps, but efforts are underway at several SR facilities to bring this down to ~ 1 ps and thereby fill this gap. A number

3b. Source and Beamlines

The workshop participants did not make detailed recommendations for the distribution of NSLS-II beamlines and source types. This is likely to be determined by particular beamline advisory teams with an interest in particular scientific problems and measurement techniques. Some of those were specified in beamline specific workshops. Instead, the HCM&MP workshop surveyed a variety of scientific activities to illustrate the variety of techniques necessary to make a complete facility. There are general expectations that the facility will include a complete complement of IXS beamlines … low and high energy, moderate and high spectral resolution, and compatible with microprobes and extreme environments. ARPES for directly sensing bandstructure and electron self energy effects. Spin-resolved PES for magnetic materials.

The NSLS-II storage ring has been optimized to deliver an extremely low emittance beam for high performance insertion devices. The low emittance is also highly beneficial for three pole wiggler and bending magnet sources for low throughput techniques. The proposed suite of IDs (SCW, DWs, U20, EPU45, QEPU100) provides a complete capability for high energy x-ray scattering and diffraction through various types of IXS (high energy, high resolution, medium energy and resonant), XMCD, ARPES and coherent imaging, while 3 pole wigglers and the standard bends serve for XAFS, XPS, some of the microprobes and all of infrared spectroscopies.

The beamline techniques for this community include:

- Inelastic X-ray Scattering (IXS). Operating at low and medium energies to access resonances and core levels, plus having sufficient q to span the Brillouin zone. At least one high resolution IXS beamline for access to low energy electronic transitions and phonons.
- Combination XPS, XAS, XRD, EXAFS. For electronic materials and devices, layers and interfaces in support of NIST programs. New electronic materials.
- XRD and XRS designed specifically for small crystal specimens. Endstation should have cryogenic sample capability.
- XRD and XRS in strong magnetic fields (XMS, DMS)
- General purpose powder diffraction.
- Soft x-ray spectroscopy and scattering. Coherent imaging.
- SX-MCD and time-resolved.
- Tender x-ray scattering. To reach heavier elements in the periodic table as found in some complex oxides. Time-resolved potential.
- ARPES – possibly with time-resolved.
- Soft x-ray microscopy: LEEM/PEEM, STXM, full field TXM, all with time resolution.
- IR magnetospectroscopy
- IR time-resolved and microprobe.
- XPCS (time-resolved)

3c. Time-resolved capabilities

This measurement and technique list of last section indicates ones where time resolved studies are anticipated, and the number is expected to grow as the potential for short synchrotron radiation pulses becomes recognized. This potential depends on the ability of the NSLS-II source to produce shorter pulses for a modest set of time-resolved users without seriously degrading performance for other. Therefore, consideration should be given in the storage ring design to ensure optimal flexibility (through the type of operation modes or future upgrades) to achieve a synchrotron light pulses capable of 1 ps time resolution. This is being actively pursued at other 3rd generation facilities such as APS, ALS, BESSY and SPRing-8. The operational modes for improved time resolution often involve performance trade-offs that affect other users of the facility. Beam instability thresholds can be difficult to predict. The APS has developed a design to employ so-called crab cavities (or RF deflecting cavities) that lead to a bunch where the vertical motion or position is correlated with the longitudinal position along the bunch. Synchrotron radiation methods that can vertically resolve (spatial or angular) the dispersed bunch can select a short time slice using a system of slits.

We expect that the hard condensed matter and materials physics community will want the capability for short bunches at many of its beamlines. The NSLS-II storage ring will have intrinsically shorter bunches (estimate 30 to 50 ps FWHM) than either the NSLS VUV/IR or Xray rings. Shorter pulses could be produced through adjustments to the storage ring dispersion, but instabilities typically limit the beam current to rather low values. Crab cavities can work with more typical beam currents, but are operated in pairs with only the beamlines in-between able to extract short pulses. These cavities also take up precious space in the straight sections, so having them optimally deployed will require careful consideration. These decisions will also depend on how well the electron bunches can be deflected in the NSLS-II lattice without introducing instabilities, beam loss and emittance reduction for other users. These issues should be addressed sufficiently prior to commitment on a detailed NSLS-II accelerator lattice to ensure a reasonably degree of compatibility.

3d. Endstation Facilities

A significant conclusion of this workshop was the need to develop advanced beamline endstations. These can be developed at NSLS and then moved to NSLS-II when appropriate beamlines are available to accept them. As a community, the development of endstations that allow for a wider range of fields, temperatures and dynamics (time-resolution) are high on our list of priorities.

- *Magnets*: DC and possibly pulsed. Superconducting coils have been developed for static fields of 20T, but SR maximum is around 15T. Potential for >20T? More manageable in terms of ring operations and EM interference. Consistent with low throughput, low S/N methods. Pulsed magnets are capable of >30T. Limited to pulses with low duty cycle. Smaller coils? (Japan)
- *Cryostats*: One or two systems capable of reaching below 1K. For studying heavy fermion systems, interplay between metallic conduction and disorder, quantum critical points. Perhaps best suited for long wavelength spectroscopy, but scattering methods may also be useful.
- *Spectrometers*: The NSLS regularly makes improvements in beamline spectrometer systems. However, a spectral range that is not properly covered at the moment is at very long wavelengths (200 μeV to 2 meV range), a range that is critical for studies of magnetism (spin / cyclotron resonance & Landau level transitions), heavy fermion materials (carrier dynamics and energy gaps) and metal-insulator transition. This is a range for which the VUV/IR ring is among the best in the world.
- *Lasers*: Synchronized ultra-fast laser systems serve in a number of potential methods for studying material dynamics. The standard use is for a photoexcitation source in pump-probe type time-resolved measurements. But these lasers can also serve as ultra-fast switches for generating current and voltage pulses and also have the potential for laser

"slicing" to create a short x-ray pulse from a long electron bunch. This latter technique requires an appropriately tuned undulator (modulator), and the effective photon flux (when duty cycle is considered) limits the feasibility of many measurements. Though a system could be implemented on the NSLS VUV ring, the limited amount of time before that ring ceases operations makes it a questionable decision. The much higher electron energy for NSLS-II presents greater challenges for laser slicing, but some performance analysis should be done before discarding the idea completely.

- *Diamond anvil cells and presses*: This is covered in more detail in the Materials Science and Engineering workshop report. Though diamond cells are not necessarily expensive, they require expertise for sample loading and accurate pressure calibration. Thus the ability to pursue high pressure studies is limited more by staffing that the availability of a particular DAC.
- *In situ materials growth and analysis*: The hard condensed matter and materials physics community would benefit from this capability, but the main focus is from the Materials Science and Engineering. We expect that a group with particularly strong ties to that community (e.g. NIST) will drive this particular effort..

3e. Supporting Facilities

The development of infrastructure for supporting the expected high-demand for NSLS-II beamtime can begin at NSLS. These include:

- Controls and automation for both beamline operations and data collection.
- Acquisition and installation of Large Area Detectors plus the necessary software and hardware for data handling and processing.
- Improved soft x-ray dectectors (faster response, improved sensitivity and energy resolution).
- Development of software and hardware standards and implementation at NSLS beamlines to be transitioned to NSLS-II.
- Defining a common platform for code development.
- Sample characterization such as optical miscroscopy, atomic force microscopy, possibly transport measurements and magnetic charcterization (e.g. MOKE and SQUID magnetometry). These facilites are available to a larger or lesser degree at other 3rd generation sources.

We also expect that the development of higher performance beamlines for NSLS-II will in itself require R&D, and NSLS should be able to play an important role in instrumentation development, such as the characterization of components, or testing of new ideas for optics, monochromators, high-resolution systems, etc.

Appendix I: NSLS-II Beamline Table

Beamline Method	E Range [eV]	Source	Transition from NSLS?	Primary or Secondary	TR Future
Low energy IXS		EPU45			(TR)
Medium energy IXS		U20			(TR)
XRD Small Xtal	6000 to 16000	U20			TR
ARPES, SR-ARPES	10 to 1000	QEPU100	yes	Primary	(TR)
XAFS/XRD/HardXPS (NIST/Materials)		3PW	yes	Secondary	(TR)
LEEM / PEEM	15 to 1000	U100	yes	Primary	TR
LEEM/PEEM	200 to 2000	EPU45	no	Primary	
IR Magneto	0.0001 to 1	LGBM	yes	Primary	TR
IR Time-resolved	0.0001 to 1	LGBM	yes	Primary	TR
XRS, Magnetic field		3PW	yes		
Soft XMCD	200 to 2000	EPU45		Primary	TR
STXM	200 to 2000	EPU45	no	Secondary	TR
Full Field TXM	200 to 2000	SBM	Possible develop @NSLS	Primary	TR
Hard X Coh. Imaging	6k to 16k	U20	no		TR
Soft X Coh. Imaging	200 to 2000	EPU45			TR
Project					
V Hi res'n IXS (PB)		U20		Primary, Project	
XPCS (PB)				Primary, Project	
EXAFS (Chem/PB)		SBM		Project	(TR)
Powder Time-resolved		DW		Secondary, Project	TR
Hi Pressure					
XRD Hi Press		SCW		Hi-P	(TR)
IR Hi Press	0.001 to 1	LGBM	yes	Hi-P	(TR)
Hi-P IXS		U20		Hi-P community	