Condensed Matter and Materials Physics (CMMP)
APS Midterm Renewal Science Case

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1. Executive Summary

The 21st century has ushered in many new societal challenges, the greatest being to develop alternative energy technologies. This is an area where CMMP can play and is playing a major role. CMMP is an engine for innovation emanating from its quest for new materials, new properties, and a new understanding of the world around us. CMMP also provides new instrumentation to enable many diverse fields of endeavor from information technology to healthcare to tools for exploring the cosmos. However, the U.S. is at a turning point where its leadership in the field of CMMP is not to be taken for granted. New strategic investments are necessary. DOE major facilities, including the APS, need to continue to play a central role in making progress toward significant national scientific and technological goals. The charge is to identify where major breakthroughs are likely to occur in the coming decade, and to utilize this perspective in order to outline upgrade investments needed to enable the APS to perform optimally to uncover, explore and understand the new science in CMMP.

This report presents an examination of the grand scientific challenges in CMMP in order to provide sets of *hot* questions that can be addressed in the future within a laboratory setting. These questions are then generalized to provide a clear indication of where it is most worthwhile to invest in APS upgrades in order to be able to perform the science of the next decade or so. From six grand challenges considered, dozens of hot questions are posed within the report. These questions provide a roadmap for a fundamental set of upgrades. The challenges include the interdisciplinary scientific and applications realms of emergence, energy, biophysics, non-equilibrium phenomena, nanoscience, and information technology (IT). The hot questions focus on opportunities to create new materials, new structures and morphologies, new properties and functionalities, and new theoretical and computational understandings that have an advanced level of predictive ability that spans multiscale spatio-temporal regimes.

Such transformational scientific advances require a new look at how synchrotrons are equipped and organized. There is need for new operational philosophies, new end-station configurations, sharply focused x-ray optics, and novel source properties in order to provide the following, which would have to be prioritized in independent studies:

- multiple extreme environments, such as high fields and/or pressures, extreme temperatures, and facilities for reactive or radioactive samples, and

- the presence of multiple, complementary probes in order to determine structure and properties simultaneously, and
- sample preparation and processing *in situ* needed to study growth mechanisms and the onset of new properties, and
- small spatial resolution in imaging and spectroscopy to complement structural investigations, and
- short time resolution to probe rapid dynamics and non-equilibrium phenomena.

In addition, a paradigm change is needed in the way synchrotrons are utilized, to actively drive new science within the user community rather than merely satisfying needs of disparate user groups. This would be a first stage to meet the scientific needs of a world faced with global instabilities that require a concerted effort to address. *The Gathering Storm* report of the National Academies has captured the nature of the challenge. This new approach to synchrotron utilization requires surveys to determine which 20th century beamlines and programs need to be cleared away in order to accommodate the science of the 21st century. This report is the beginning of a dialogue of deep introspection that will bring about sweeping changes to advance a critical scientific agenda of renewal to ensure U.S. leadership in scientific and technological goals that are shared globally. More detailed recommendations will require workshops to further examine the ideas set forth herein.

2. Introduction

CMMP is an engine of innovation for society at large, leading to industrial applications especially in areas of high technology, and thus stimulating the economy, helping to ensure our defense and homeland security, and most importantly, striving to improve our energy independence. CMMP helped to initiate the IT revolution that is changing our world in so many ways, from the Internet to the *iPod*. CMMP is also an enabler of scientific ventures outside of its own domain because of the materials and instruments that it creates and perfects, such as solid-state lasers, transistors, fiber optics, magnetic resonance imaging, and scanning probe techniques, that energize fields from healthcare, to space exploration, to particle accelerators. Thus, CMMP is amongst the most exciting areas of science - discovering and investigating the emergence of new materials and phenomena.

CMMP embraces “the science of the world around us.”[1] It involves the creation of advanced materials, the exploration of their properties, and an understanding in terms of both the theoretical underpinnings of the phenomena of interest, and the structure-properties relationships exhibited. CMMP is a broad field that encompasses mission-oriented research, such as for advanced solar cells, and largely curiosity-driven research, such as in the fractional quantum Hall effect. It encompasses bench-top science as well as major facility research, independent investigator research, as well as team research - experimental physics, theoretical physics and computational physics. It can involve hard materials, like superconductors, or soft matter, like polymers, organics and biomaterials. And it can involve different forms of these, such as solids, liquids, glasses, crystalline, amorphous, nanostructured, and/or composite systems. The structure and properties can be intriguing in their equilibrium states, or in metastable or dynamically driven environments as a function of state variables and/or spatio-

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[1] *Physics 2010 CMMP Report*, Nat’l Acad. Press 2007

temporal constraints. Thus, there is a richness and diversity to the field that makes for a lively internal culture characterized by a mix of virtuosity, inquisitiveness and unpredictability, a culture shared by many scientific disciplines.

There have been many efforts to encapsulate what the 21st century will represent scientifically. The BES Grand Challenges Committee[2] coined it the *Age of Control*. Physics Nobelist Robert Laughlin[3] dubbed it the *Age of Emergence*, where we delight in the study of the complex organizational structure that grows out of simple rules. The journalist Thomas Friedman recently alluded to an *Era of Energy and Technology*.[4] These are all apt descriptions that predict exciting possibilities within our grasp.

As we move forward in our task to foresee how to best prepare for the future of our science at the Advanced Photon Source, we know that we will necessarily do an imperfect job. The best we can try for is to keep an open mind, strive for good scientific taste and judgment, and to change course to adjust to the future as new and unexpected opportunities present themselves.

3. Key Science Drivers

The scientific case for the beamline upgrade at the APS must begin with the grand challenges in CMMP. These were already identified in the CMMP 2010 Report. (The BES Grand Challenges report emphasizes similar themes, encompassing additional scientific areas as well.) Thus, our focal point is the six challenges identified in the CMMP 2010 Report:

1. How do complex phenomena emerge from simple ingredients?
2. How will the energy demands of future generations be met?
3. What is the physics of life?
4. What happens far from equilibrium and why?
5. What new discoveries await us in the nanoworld?
6. How will the information technology (IT) revolution be extended?

These six overlapping challenges point to where we anticipate the most exciting breakthroughs over the next decade and beyond: Emergence, Energy, Biophysics, Non-equilibrium, Nano and IT. The committee fleshed out these challenges with lists of hot questions that make the challenges more tangible in a laboratory setting. We outline below examples of these hot questions. The questions reveal patterns indicating that similar approaches are needed to address the different challenges. This allowed us to identify fundamental types of experiments that are needed to advance the field. Many of them could benefit from the hard x-rays that the APS provides, if there were suitable upgrades, as will be outlined in Section 6 of this report.

In summary, we will see that the lists of hot questions that follow have a healthy amount of redundancy to them, and lend themselves to experiments that require:

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[2] G. Fleming and M. Ratner, *Physics Today*, July 2008, pp.20.
[3] R. B. Laughlin, *A Different Universe*, (Basic Books, 2006)
[4] *New York Times*, July 30, 2008

- multiple extreme environments, such as high fields and/or pressures, extreme temperatures, and facilities for reactive or radioactive samples, and
- the presence of multiple, complementary probes in order to determine structure and properties simultaneously, and
- sample preparation and processing *in situ* in order to study growth mechanisms and the onset of new properties (in, for example, single crystals, films, foams, glasses, proteins, polymers, heterostructures, and/or nanostructures,) and
- small spatial resolution in imaging and spectroscopy to complement structural investigations, and
- short time resolution to probe rapid dynamics and non-equilibrium phenomena.

1. How do complex phenomena emerge from simple ingredients?

Is it possible to develop multi-scale spatio-temporal methods to theoretically or computationally predict desired properties, structures, dynamics of transitions between structures, and synthetic pathways? Theorists have developed powerful codes to simulate materials properties at the atomic scale, at the mesoscale, and in the continuum limit. A challenge is to tackle the weak-coupling limit where such codes can be meaningfully integrated into a hierarchy so that the properties of macroscopic systems can be simulated from elementary constituents, including organic, molecular and biological building blocks, as one traverses vastly different time scales. This involves challenging issues associated with the interface at each hierarchical step, in order to reconcile assumptions, pass meaningful parameters, and retain physically significant and accurate results and insights. The strong-coupling limit will require entirely new theories and approaches.

How does one describe materials whose electrons/atoms are neither perfectly localized nor fully itinerant (*i.e.* correlated electron/atom systems)? Fermi liquid theory has enjoyed great utility, but there are important classes of materials that defy description and require going beyond present day theoretical understanding of complex matter. Traditionally, the high-temperature superconductors and narrow-band metals, such as the heavy Fermion systems, fall into the category that poses major challenges to understand. The 5*f*-electron systems are relatively unexplored, but might hold the key to understanding because their *f*-bands and bonds are more itinerant than are the 4*f*'s, but tend to be more localized than 3*d*-states. Can we identify and understand materials with quantum critical points? Today another important facet of the problem is to understand correlated atom behavior in ultra-low temperature Bose-Einstein condensates, such as have recently been discovered and explored using laser cooling methods.

Can we integrate materials whose properties tend to be mutually exclusive, such as ferromagnetic and superconducting, or transparent and conducting? There is always the challenge to search for elusive single materials that embrace diverse properties, such as novel multiferroics. This quest to create hybrids has more recently captured the imagination of the scientific community. For example, combining soft and hard matter could bring the physics community closer to understanding intriguing issues associated with the life sciences. What is the origin of such contra-indicated properties? What are the limits of their performance, and why are they limits?

2. How will the energy demands of future generations be met?

Fundamental advances in CMMP are central to all technologies currently being eyed as contributors to meeting future energy demands, from renewable energy such as solar energy or biofuels, to carbon-neutral energy sources such as nuclear energy, to energy efficient technologies such as solid-state lighting. The diverse roles of materials in these technologies raise an equally diverse set of scientific questions.

Can we create high efficiency, low cost photovoltaic devices? Is there a self-assembly strategy that can produce highly efficient solar energy conversion? Can we solve the materials problems in wide bandgap semiconductors (Group III-Nitrides and ZnO) for green, blue and UV light emitters? Can we improve wide bandgap materials/devices (GaN, SiC, *etc*.) for high power and high temperature applications (*e.g*. electric transmission, electronics for improved efficiency of automobile and aircraft engines)? A key to progress might be a new understanding of materials that enable rapid searches of phase space to gain a wide variety of information. In developing thermoelectrics, which convert heat into electricity, the need is to balance the properties of phonons *vs* electrons. How can we advance the search? For nuclear technologies we need to study fundamental properties of transuranic systems, fuel rod designs, environmental remediation, and stewardship of materials. In summary, we need to discover new classes of materials and combinations with outstanding properties, *e.g*. efficient light emission, photovoltaic conversion, superconductivity, catalytic efficiency, electrodes for fuel cells, *etc*.

What is the source of the uniqueness of catalytic sites on a surface? What is their structure? How can we distinguish “spectator” sites from “player” sites? Are there new catalytic properties of nanostructured elemental or composite materials that are not the canonical ones in use today? Are there classes of energy conversion reactions that share similarities in their transition state? Can we identify the structures and similarities and use them to create new insights, sets of design rules, and to tune chemical intuition?

3. What is the physics of life?

How does one distinguish, characterize and differentiate living from inert materials? What is the chemical language by which cells communicate? What are the differences between right- and left-handed DNA and what is the basis for chiral asymmetry in life? Does this question relate to the chemistry and physics of how molecules form in extreme environments? Can we observe and record the structural changes in arbitrary biomolecules when they interact with each other or with external stimuli? How does protein folding influence the properties of active sites? What can we learn from biosystems in order to culture and harvest the materials we need, from biofuels to self-repairing materials, to computer subsystems?

What are the “design rules” governing the 3D architecture and properties of soft materials in specific environments (temperature, radiation, chemical, *etc.*)? What is the origin of “mistakes” in structure during synthesis? Can we avoid these “mistakes” to get compositionally and structurally pure material? What governs the limits of charge transfer in organic/polymer materials? How can we optimize this property for particular needs?

4. What happens far from equilibrium and why?

Is there a general way in which collective behavior emerges or disappears? Phenomena such as magnetism, superconductivity and ferroelectricity can be created out of non-equilibrium states, *i.e.* by pumping with light to a higher excited state, or by pumping an ordered state into the disordered phase. Can the glassy state be described theoretically?

Is there a universal description of quantum transport, one that can embrace electrons, spins, molecules and fluids? Non-equilibrium quantum transport equations need to be examined within the broadest possible perspective to encompass phenomena as diverse as, for example, superfluid systems, organic spintronics, single electron transistors, and ultra-fast dynamics of optically excited semiconductors. Can all ranges of frequency, mobility and spatial scale be meaningfully covered? Is there a universal origin to 1/f noise? Can we witness the birth of an elementary excitation in CMMP systems?

5. What new discoveries await us in the nanoworld?

Nanomaterials offer the promise to enable us to continue to develop technologically as we face that present-day materials and processes are reaching their limits. The questions concern what is beyond Si? Can we transcend microlithography with bottom-up approaches? Can we harness spintronics, near-field photonics, and other advanced concepts and approaches to meet the needs of tomorrow? Will quantum computing become a reality? Will we learn to utilize bio-inspired approaches to address our many societal concerns?

As objects shrink in size, are new methods available that are needed to measure what we make, thus enabling the study of the new physics that emerges? Can we achieve the level of control needed to fabricate integrated nanosystems with the same level of reproducibility as found in today’s integrated circuits? What are the limits to controlled fabrication of integrated nanosystems with reproducible properties? And what governs these limits? Can one control and manipulate organic-inorganic interfaces? Can nanophotonics be used to make lasers in the mid-IR and to find new detectors that extend farther into the IR?

What are the rules that govern phase separation? What are the rules that govern self-organization in general? Self-assembly, defined as the autonomous organization of components into patterns or structures, is rapidly emerging as a topic of major interest. Self-assembly occurs on length scales that extend from the molecular to the cosmic. Self-assembly provides pathways to create new nanoscale materials from the bottom up that transcend the spatial limits of lithographic patterning, and is implicated as a universal feature in the physics of many complex systems. The thermodynamic and kinetic rules that govern self assembly, and the underlying structural and electronic properties, need to be understood in order to harness self assembly to create a new generation of advanced materials and hierarchically organized systems, as well as to get a better physical understanding of living systems. For example, how does surface topology influence morphology? Can porous membrane materials provide a new platform to catalyze heterogeneous chemical reactions while separating reactants and products?

6. How will the information technology (IT) revolution be extended?

As we reach the end of Moore's law in Si technology, advances in CMMP will have a crucial role in extending the IT revolution. Can we solve the materials issues related to the convergence of III-V semiconductors with Si - integration of light emitters/detectors with Si ICs? Can we engineer new substrates for semiconductors that are not lattice matched to bulk materials and thereby exploit strain to tailor properties? Can we heterogeneously integrate different materials on one integrated circuit chip, with multiple layers of devices (3D integration), overcoming problems of differential thermal expansion and stress? Can we tackle the problems of advanced packaging/heat extraction from circuits? What lies beyond the semiconductor roadmap? Can we understand and develop alternatives to Si electronics, *i.e.* to determine the limits of opto-electronics, photonics, plasmonics or spintronics. Can we utilize the wide range of functionalities of complex oxides, together with their high susceptibility to external perturbations, to develop wholly new types of (Mottronic) technologies?

Modern semiconductor-based electronics utilize the charge of the electron, but ignore its spin. Harnessing the spin can add value and functionality, such as in non-volatile electronics that retain stored information when the power is off, and in the creation of ultra-low heat dissipation circuits that communicate via pure spin currents without the flow of charge. Can we image a pure spin current? Can we find materials for spintronics devices (for injection of spin polarized electrons into semiconductors)? Can one find nearly 100% spin polarized electron sources and retain interfacial control of the polarization? Can one create a spin transistor with gain to enable spintronic applications? Can one find magnetic semiconductors that operate well above room temperature? A corollary challenge is to integrate new spintronic materials with conventional electronic materials and processes in order to add value and evolve the technologies in a continuous fashion.

How does one store and address information with light? As interest in optical materials and phenomena migrates to the sub-wavelength and near-field realms, new challenges arise. Exploring and understanding the relevant issues will advance the emerging areas of photonics and plasmonics. The key requirement of addressability, the ability to direct the movement of energy or signals within a useful architecture, is a difficult challenge. Future developments in photochemical energy transduction, opto-electronics and optical computing might hinge on such breakthroughs. A corollary challenge is to address the question of what material parameters drive the coupling and diffusion of thermal energy on length scales shorter than a phonon wavelength.

Can we create solid-state devices that operate at 80 K or higher for quantum computing? What are the ramifications of quantum entanglement and coherency in computation? The goal ahead is to explore new paradigms that can enable computations that cannot otherwise be achieved.

A challenge is to develop arrays of quantum coherent objects that would form the quantum bits (qubits) and the logic gates of a quantum computer. What materials systems and configurations permit sufficiently long quantum coherence times for computation?

4. Significance of APS

The Advanced Photon Source is one of the world premier hard x-ray facilities offering a crucial set of tools to attack these forefront scientific areas. These powerful hard x-ray synchrotron techniques, such as high-resolution scattering, imaging, and spectroscopy, offer unique ability to penetrate materials and provide an element-resolved picture to address the scientific grand challenges and associated hot questions outlined above. However, a new paradigm will need to be embraced for the APS to realize its full potential to keep our Nation at the leading edge of the scientific frontier. Presently, the APS is largely organized to serve independent user communities. In the future the APS will provide greater value by leading and focusing the user community in order to make sure that a critical mass of talent is focused on grand-challenge goals. This means that the APS will have to reorganize to act as a single organic entity to address a hierarchy of scientific questions in a coherent manner. The APS will need to make room for new 21st century science, expanding some of its lab modules into full-service laboratories, of which the Center for Nanoscale Materials can be considered as a first model. End-stations on rolling platforms may be moved in place or x-ray beams may be brought to semi-permanent installations, through collaborations from contiguous institutes where the hot questions are pursued 24/7. User agreements will span over periods of years, rather than days, in order to get serious work accomplished, as outlined below.

5. Scientific Community

The CMMP scientific community will benefit, as well as our Country and the Planet as a whole.

6. Requirements and Capabilities

This proposal encompasses a transformational change in the way experiments are undertaken at major synchrotron facilities. Rather than having the x-ray studies conducted separately, a complete suite of experiments would be performed *in-situ* on the beamline in real time by combining a variety of probes and processing tools. The evolution of static and dynamic properties can, thusly, directly connect with information provided by the x-rays. We will outline several types of experiments and capabilities. Key to this approach will be a strengthening of ties to existing external users as well as generating new users who would partner to perform long-term (1-3 yr) dedicated scientific programs. The new types of combined experiments fall into general categories:

- expansion in facilities to bring widely applicable, powerful hard x-ray synchrotron techniques such as high resolution scattering and spectroscopy to bear on a greater variety of materials and processes.
- an appropriate balance of facilities for specialized, high performance, niche techniques (*e.g.* inelastic scattering, magnetic scattering, ultra-fast or coherent x-ray techniques).

- the presence of multiple, ancillary and complementary probes. The ability to follow material properties in real time is crucial in order to correlate the x-ray measurements with changes due to transitions between allowed phases. This would be accomplished by enabling beamlines to directly integrate laboratory-based measurements of electrical, magnetic and optical properties. This also encompasses the ability to externally perturb systems away from equilibrium with electrical and optical stimulation, which is crucial to study phase transitions and systems both near and far from equilibrium.

- combinations of extreme environments (to alter temperatures, pressures and fields, and to accommodate reactive chemistry, radioactive samples, and ultra-short electromagnetic pulses and shock waves.) Such measurements are suited to x-ray probes that can handle small sample volumes because high pressures and pulsed magnetic fields are more readily created over confined spaces. While today such experiments can be undertaken in environments with one extreme condition, in the future multiple extreme conditions imposed simultaneously will be used to track the behavior across a multidimensional phase space.

- *in-situ* processing and synthesis. Understanding, for example, catalytic phenomena requires the ability to alter reaction conditions in real time. Obtaining rate laws and transition state information, and being able to extract general insights depends on following reactions dynamically. Similarly, *in-situ* studies of film growth processes provide a unique opportunity to understand the formation of materials in real time. Via such studies much of the art of materials growth can be transformed into a science, where intuition will succumb to sets of rules to guide discovery. It is envisioned that major efforts can be launched to provide a synthesis institute with an initial array of grow environments that include, for example, different types of growth chambers, such as molecular beam epitaxy, as determined by workshops that identify the optimal goals in synthesis and processing.

- where possible, portable systems offering the above capabilities to the user community that can be transported to multiple beamlines, optimized for different x-ray studies. With an integrated laboratory space such systems could then be harnessed when off-line in preparation of experiments.
- where needed for complex, non-portable apparatus, facilities and peer-reviewed access would allow building and operating experiments for extended periods (2 years) in multiplexed hutches where an x-ray beam can be shared between multiple experiments.

- a range of spatial resolutions extending to ~1 nm for imaging and probing with various x-ray contrast mechanisms (absorption, phase contrast, diffraction, fluorescence, spectroscopy, polarization).

- a range of temporal resolutions extending to ~1 ps to probe rapid dynamics and non-equilibrium phenomena.

The above bullets can be considered as a first generation of recommendations to be discussed, altered and/or refined via a process that is open to stakeholders in the community. The need for changes in the structure/organization of APS beamlines, highlighted only in very general terms herein, will also need to be discussed by a broad community. Some constructive suggestions below can help to get the dialogue launched.

- A two-step process is needed to determine a prioritized strategic plan and future portfolio of beamlines:
    - Evaluate the existing beamlines in the light of the scientific needs identified by the reports of the APS Science Teams, and
    - empower a single blue-ribbon committee over the next year to critically assess all APS scientific and technical needs (including but not limited to CMMP issues), balance the number of sectors needed in each area, and determine which beamlines to upgrade and which sectors should become available to be rebuilt into new 21st century facilities.
- Implement this prioritized strategic plan as the APS Renewal. In some cases this will lead to the formation of new research communities as stakeholders at APS.
- Looking further, some grand challenge areas will nucleate new institutes to be formed around the beamline facilities.
- Finally, there will be a need to nurture staff to pursue breakthrough science as well as interact with external users at these enhanced beamlines.