Argonne Scattering and Imaging Institute
A White Paper

Raymond Osborn, James W. Richardson, and Gabrielle G. Long
Argonne National Laboratory

The Department of Energy operates a range of major facilities that enable over 10,000 scientists each year to perform world class science using x-rays, neutrons, and electrons. These facilities have developed, and continue to develop, state-of-the-art instrumentation that exploits the dramatic improvements in source flux and brightness produced over the last two decades. However, there has not been a comparable investment in the theoretical and computational tools that are required to optimize the scientific productivity of these instruments, nor has there been a mechanism to integrate scientific resources across facilities to address grand challenges beyond the reach of individual users. The goal of the Argonne Scattering and Imaging Institute is to address the grand challenges in materials science performed at large-scale facilities and, in so doing, build a theoretical and computational infrastructure that will benefit the entire scientific community.

Introduction

In January, 2005, the BESAC Subcommittee on Theory and Computation recommended in their report *Opportunities for Discovery: Theory and Computation in Basic Energy Sciences* that "BES should undertake a major new thrust to significantly augment its theoretical and computational programs coupled to experimental research at its major facilities." The background to this report is that facilities such as the Advanced Photon Source and the Spallation Neutron Source, and the new generation of aberration-corrected electron microscopes, provide unprecedented opportunities for scientific discovery that promise to generate new understanding of materials and new technologies to address societal needs. The dramatically enhanced source flux and brightness at the x-ray and neutron sources has led to novel techniques exploiting beam coherence and polarization and increasingly complex instrumentation with multi-detector systems designed to maximize the efficiency of data collection. The ability to control aberrations in electron microscopes has broadened the range of materials systems that can be analyzed and led to an increase in achievable resolution. However, the scientific promise of these instruments will only be fully realized when there is a comparable improvement in our ability to visualize, analyze, and understand the data that they produce. Fortunately, there have been parallel advances in both the sophistication of theory and the power of computation, so the time is right to respond to the challenge laid down by the BESAC report. The Argonne Scattering and Imaging Institute (ASI^2) is designed to do just that by focusing on well-defined scientific challenges that can only be accomplished by combining the most advanced experimental, theoretical, and computational expertise.

A distinctive feature of ASI^2 is its organization. Through a mixture of permanent staff and temporary visitors, the institute will contain professional expertise in scattering and imaging theory, experimental techniques, simulation, numerical analysis, algorithm development, data visualization, and software engineering. Under the guidance of a Science Advisory Council, the institute personnel will be assigned to limited-term projects that are chosen to address grand challenges in scattering and imaging science.

The scientific community will be invited to propose these projects, which will then be selected using three criteria:

1. The importance of the scientific problem.
2. The need for the multidisciplinary support provided by the institute.
3. The size of the community that would benefit from the resources developed.

Consideration will particularly be given to projects that enable data from multiple facilities and multiple techniques to be combined within a coherent data analysis framework to address a grand scientific challenge.

We believe that this project-oriented structure is essential to maintaining a high level of interaction between the different disciplines and to ensuring a strong focus on the achievement of scientific, rather than purely technical, goals. After project completion, the strong body of professional software engineers will ensure that the resources developed are user friendly, robustly designed and well maintained to benefit the wider scientific community in the long term.

In this White Paper, we will give some examples of the scientific challenges that the Argonne Scattering and Imaging Institute could tackle, and the resources that would be needed. We then describe in more detail the matrix structure of the institute and the composition of its groups.

Science

In the following sections, we give a few examples of the type of scientific question that could be tackled by ASI^2 project teams. They meet the broad criteria described in the introduction, but only represent a small fraction of the potential science that ASI^2 could generate. It will be the task of the Science Advisory Council to prioritize and select the actual projects from those proposed by the community. We envisage about six to ten projects being active at any one time, each one lasting from three to five years. In the first year of the institute, before the project proposal system is fully operational, we shall select three projects after consultation with the management of major DOE facilities.

Condensation Energy

One way of determining the origin of high temperature superconductivity (or any other novel ground state) is to answer the question, "Where do the energy savings come from?" In classic superconductors, it is from the lowering of the ion kinetic energy. In cuprates, however, others have been suggested, such as a lowering of the exchange energy of the electrons, their kinetic energy, or their Coulomb repulsion. Testing these alternatives requires a knowledge of various response functions: the electron momentum distribution (measured by photoemission, Compton scattering, and positron annihilation), the spin-spin response (measured by polarized neutron scattering), the density-density response (measured by inelastic x-ray scattering), and the dielectric function (measured by ellipsometry and electron energy loss spectroscopy). The condensation energy, though, is so small that very precise measurements are necessary over a large range of (four-dimensional) momentum and frequency space. Moreover, various effects such as matrix elements, backgrounds, instrumental resolution, noise bias, and other excitations (phonons, etc.) must be factored out to obtain meaningful results. This is a real challenge not only in data acquisition for a variety of spectroscopies, but also in data analysis,

modeling, and theory. Meeting this challenge will solve some of the most fundamental questions facing materials science with profound implications for future technologies.

Dynamic Structure of Self-Assembled Fluids

Gaining a fundamental understanding of the relaxation process due to thermal motion in micelles, colloidal suspensions, lamellae and microemulsions is one of the grand challenges in polymer science. Light photon correlation spectroscopy accesses length scales centered around 500 nm, but this is larger than the characteristic size of typical self-assembled structures, which is around 10 nm. x-ray photon correlation spectroscopy enables measurement of the dynamic structure factor on this length scale, but there is an urgent need to develop the numerical analysis and modeling to support the correlations uncovered in the data. Furthermore, quasi-elastic neutron scattering probes similar length scales, requiring the development of methodologies that utilize the complementary nature of measurements in the time and frequency domain.

The Nano Problem

Nano-materials have properties that differ significantly from their larger, but chemically identical, counterparts. A key driver for these differences is often a difference in structure, but standard crystallographic analysis techniques do not apply to nano-materials since they lack translational invariance. The use of multiple techniques (diffraction, imaging, and spectroscopy) can help to constrain, and therefore, determine structure solutions. We need to develop algorithms for taking local structural data, obtained through Pair Distribution Function analysis and XAFS, electronic structure determined from three-dimensional electron energy loss data, and real-space imaging, so that they can be combined to generate unique solutions to the structure of nanoparticles.

Multiscale Modeling of Plasticity

The distribution and evolution of elastic strains (and stresses) at the submicron length scale within deformed metallic structures has broad implications for understanding mechanical behavior, transport of dislocations, and changes in properties as a function of loading. Since the effects of small length scale processes often propagate all the way to the macroscopic level, a fundamental understanding of plastic deformation can only be obtained through multiscale modeling. While no single experiment can provide the entire range of information required, transmission electron microscopy, ultra-small angle x-ray scattering and x-ray diffraction imaging have been developed to a level where they are capable of providing the experimental input required to guide dislocation dynamics codes and to produce reliable benchmarks for multiscale simulations. The grand challenge in plasticity is to derive physics-based constitutive laws that govern the behavior of materials, rather than the purely empirical rules that we have today, so that the development of processing protocols for new materials is not prohibitively costly.

Complex Disorder

Complex disorder in solids, characterized by short-range correlations that extend up to 100 nm, plays a significant role in many phenomena of current interest, including colossal magnetoresistance, relaxor ferroelectricity, negative thermal expansion, quantum spin liquids, and high temperature superconductivity. In transition metal oxides and related compounds, the enhanced response to external fields is intimately linked to the existence of strong short-range order (stripes, ladders, checkerboards, phase separation) and the highly ramified energy landscape that underlies it. Understanding the novel ways

in which materials respond and self-organize on a variety of length scales is a key challenge in condensed matter physics. There are substantial computational and theoretical challenges in modeling complex disorder and short-range correlations. Analyzing large multi-gigabyte datasets requires the development of efficient algorithms for extracting the diffuse intensity, refining the average structure, and visualizing the real-space deviations from the average structure. A powerful way of constraining defect models is to combine complementary data from multiple techniques: neutrons to give the time-averaged structure, x-rays to give the instantaneous structure, and electrons to provide real-space images of defect distributions. This multi-pronged approach is an important advantage of an institute that contains expertise in multiple scattering techniques.

Nanomagnetism

The artificial manipulation of magnetic materials at the nanoscale has led to the observation of new physical phenomena relevant to a number of key technologies where confinement and symmetry breaking at surfaces and interfaces, for example, are key contributors. Competing interactions at the nanoscale result in novel, inhomogeneous magnetic states wherein the magnitude or direction of magnetic moments vary in depth away from surfaces and interfaces. While magneto-optical techniques spatially-average over the near-surface region, and magnetometry averages over the sample thickness, polarized neutron reflectometry and x-ray resonant magnetic scattering provide depth-resolved magnetization profiles through measurements and analysis of the neutron and x-ray intensity and polarization. A grand challenge lies in developing fundamental models of depth-resolved magnetization profiles in artificial nanostructures with special emphasis in probing the nature of the inhomogeneous state within individual layers.

Coherent Imaging

Coherent x-ray imaging is a fast developing field with applications in the study of phase transitions, charge density waves, and the dynamics of polymers and colloidal particles, using speckle patterns and the creation of three-dimensional images of the interior density of particles from the micrometer down to the nanometer scale using lensless coherent x-ray diffraction. The development of the field of coherent x-ray imaging depends strongly on the development of high-performance data analysis. For example, a crucial aspect in coherent lensless x-ray imaging is the development of efficient and generally available codes that allow the inversion of diffraction patterns into three dimensional images. The underlying algorithms rely on oversampling for the retrieval of the phase that is lost in the diffraction process, but essential in the reconstruction of the three-dimensional image. The creation of robust and user friendly inversion codes combined with the development of optimal measurement strategies that allow researchers to deal with large quantities of data is crucial for the success of coherent imaging as a tool at synchrotrons and x-ray free-electron lasers, such as the LCLS.

The Structure and Function of Biomembranes

Biological cell membranes are highly selective barriers, predominantly formed from impermeable lipid bilayers, whose selectivity comes principally from the specific membrane proteins embedded in and spanning the lipid bilayer. These membrane proteins form complexes that control transport through the lipid bilayer and are responsible for most of the communication between intracellular compartments and

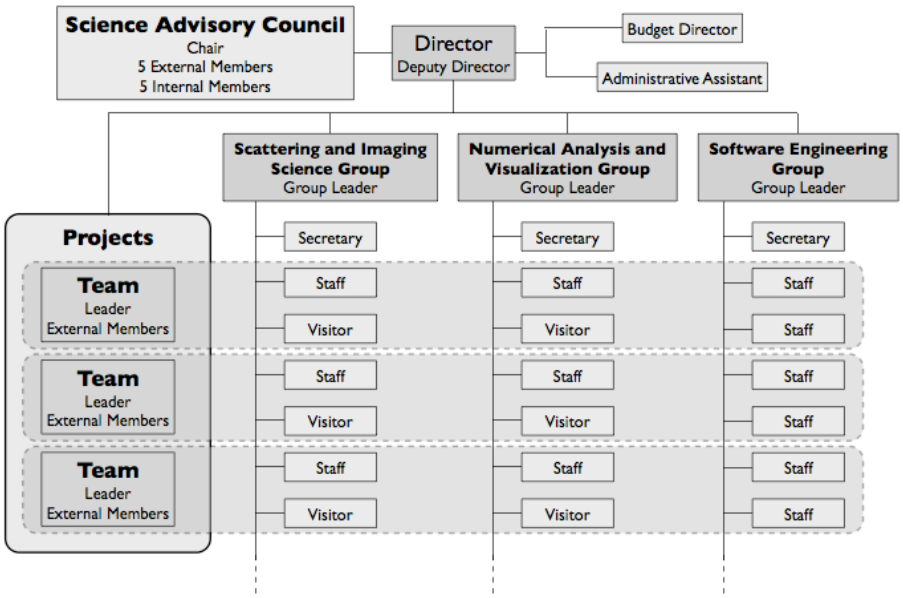

Figure 1: Organization chart of the Argonne Scattering & Imaging Institute.

between the cell and its environment. It is clear that lipid composition and other associated membrane components, like cholesterol, provide the membrane with specific structural characteristics that enable this function. Insight into the structure-property relationships of realistic cell membranes and their bioactivity requires the use of multiple probes over a wide range of length and time scales, combined with multiscale simulations of macromolecular interactions.

Organization

Group/Project Matrix

The organization of the institute is illustrated schematically in Figure 1. The institute will contain both permanent staff and temporary visitors who are organized into three groups, defined by broad areas of expertise. The group leaders will ensure that the composition of their groups is well balanced and meets the needs of the institute as a whole. The group structure enables the staff and visitors to benefit from interactions with their peers and helps them collectively to keep abreast of the latest developments in their fields.

In addition to their group membership, each member of staff or visitor is assigned to one or more of the projects that have been selected by the Science Advisory Council. This assignment is designed to match the particular needs of each project, and will last as long as required by the project leader up to a limit of 3 years with the possibility of renewal for 2 years. This is analogous to the evolving needs of, for example, a building project, where different skill sets may be needed at different stages of construction, and gives the projects tremendous flexibility in achieving their scientific goals. Another important benefit of the matrix approach is that interactions between group members will help them to identify overlaps in project needs, and so prevent unnecessary duplication.

A strong visitor program will also contribute to the scientific range and flexibility with which ASI^2 accomplishes its goals. They will ensure that gaps in the in-house expertise

are filled by highly qualified scientists both within Argonne and in the broader scientific community. Those from neighboring institutions can have regular interaction with the rest of the project team for the duration of the project, whereas those from more remote institutions might visit Argonne for shorter periods during critical phases of the project.

Scattering and Imaging Science Group

This group will consist of scientists with particular expertise in different fields of theory, simulation, and experimental techniques. The U.S. does not have any theory groups to match the size and scope of those attached to European facilities such as the European Synchrotron Radiation Facility. ASI^2 will correct this imbalance with experts in, *e.g.,* the theory of dynamical diffraction, density functional theory, lattice dynamics, spectroscopy, resonant scattering, molecular dynamics, quantum Monte Carlo and multiscale simulation. In order to attract scientists of the highest quality, they will be allowed to spend a fraction of their time on independent research related to the overall goals of the institute. This will also give the institute some flexibility in responding rapidly to new scientific directions, which could then generate new project proposals.

Numerical Analysis and Visualization Group

It is critically important that ASI^2 takes advantage of the latest developments in the techniques of numerical analysis and advanced visualization. At present, there is no established interaction between the major DOE facilities and the vibrant academic community responsible for developing new algorithms, optimization techniques, advanced statistical analysis, methods of automated data mining, multi-dimensional histogramming, and data inversion. There is also an urgent need for advanced visualization techniques to facilitate real-time analysis of experimental data, and develop novel methods of, *e.g.*, identifying significant scattering events in sparse data, or highlighting dispersion surfaces and Brillouin zone boundaries embedded within multidimensional data volumes. By recruiting experts in these fields, the Numerical Analysis and Visualization group will enable facility science to benefit from the latest advances in their respective communities. Argonne already has leading researchers with relevant expertise in the Mathematics and Computer Science division, and ASI^2 will draw on their experience in setting up this group as well as benefit from close interactions in the future.

Software Engineering Group

One of the criteria by which ASI^2 projects will be chosen is the benefit to the wider scientific community of the resources developed in meeting their scientific goals. These resources will mostly be in the form of "community codes", software tools that may be applied to a wide range of scattering and imaging problems. The third group in ASI^2 will consist of a group of professional software engineers that will be responsible for developing and maintaining these community codes.

Historically, most scientific software have been developed by the scientists themselves, but this paradigm is unlikely to be sufficient in ASI^2 for two reasons:

1. The software developed through ASI^2 projects will often be computationally intensive, requiring advanced code parallelization or grid computing techniques, beyond the usual skills of even computationally literate scientists.

2. It is essential that the software is developed within a coherent framework using the best practices of software engineering so that it is easy to maintain, compatible with modules developed by other projects, platform-independent, user friendly, and well documented.

This group will ensure that the computational infrastructure developed by ASI^2 will serve the needs of the overall U.S. scattering and imaging community, and not just the immediate needs of each project. The group will also contain experts in GUI design and web-based documentation so that the software is widely adopted by the wider community, as well as experts in software project management to oversee testing, release schedules, and long-term software maintenance.

Relation to the DANSE Project

The first investment in the kind of community codes that ASI^2 will develop is the NSF-funded DANSE software development project. This is a collaboration between a network of universities and neutron scattering facilities, led by Caltech, to develop more sophisticated neutron data analysis tools and make them accessible to the wider scientific community through a distributed computing architecture. DANSE has been awarded $11.97M over five years starting in FY07. ASI^2 will coordinate its activities with those of the DANSE project for its duration and cooperate on areas of strong overlap.

In our proposal, ASI^2 will have considerably greater resources than DANSE, but mutual cooperation will benefit both projects. DANSE will provide neutron scattering expertise and experience in developing neutron data analysis frameworks. ASI^2 will provide professional numerical analysis and software engineering expertise, and will broaden the scope of DANSE software to include x-ray and electron microscopy science. When DANSE is complete, ASI^2 can assume responsibility for the long-term maintenance of the DANSE project's legacy.

Conclusion

The Argonne Scattering and Imaging Institute will play a vital role in enhancing the scientific productivity of the major DOE x-ray, neutron, and electron facilities. It will address the grand scientific questions that can be answered by scattering and imaging when coupled to advanced theory and computation, and, in so doing, generate a body of robust community codes that will enable the U.S. scientific community to maximize the effectiveness of their facility experiments. It will also encourage the use of multiple techniques within a coherent data analysis framework, which should greatly enhance the reliability of the resulting scientific conclusions.

We propose that Argonne host such an institute because it has a long history and strong existing base of x-ray, neutron, and electron scattering and imaging expertise, strong programmatic divisions in the physical, chemical, and biological sciences, and leadership computational research expertise in the Mathematics and Computer Science division. It is also located close to a number of educational institutions of exceptional quality and will make full use of the resources that they offer to supplement those at Argonne when necessary.