High-field magnet development for x-ray scattering and spectroscopy studies.

This proposal presents plans for developing both steady-state and pulsed-field magnet instrumentation for high-field scattering and spectroscopy research. For scattering studies requiring a continuous field, a new state-of-the-art 15 Tesla magnet with 300mK capabilities specifically designed for x-ray experiments is envisioned. For studies of materials at even higher fields, a modular 1MJ capacitor bank capable of producing pulsed fields of up to 60 Tesla for 30 ms is proposed. Both these instruments would aide in nucleating a user base for a future beamline dedicated to research at extremely high fields.

Scientific Justification

The primary motivations for subjecting materials to extreme conditions fall into two general classes: (1) from basic science point of view, novel states of matter that are crucial for developing a correct theoretical understanding of materials can be realized by changing external conditions, which would otherwise not be observable; (2) from applications point of view, inorganic and organic materials can be treated or manipulated by magnetic fields to change their functional behavior. Unlike the two most commonly exploited thermodynamic variables, namely pressure and temperature, and chemical manipulation, magnetic field is unique in that it couples directly to the spin and orbital degrees of freedom of materials. Furthermore, in both relativistic and non-relativistic treatments of classical and quantum mechanical problems the magnetic field becomes an integral part of kinetic motion of elementary constituents of matter via the so-called canonical momentum. In other words, the magnetic field is a key *fundamental* parameter that enters directly into the energy expressions (*i.e.* the so-called Hamiltonians) that govern all properties of materials. As such, a magnetic field acts as a contact-free and versatile experimental "knob" for tuning properties and states of matter under extreme conditions.

The problems which can be addressed utilizing high magnetic fields are varied and span both equilibrium and non-equilibrium phenomena. First, using a superconducting DC magnet such diverse equilibrium phenomena as field-induced structural responses in energy-related functional materials (magneto-caloric, magneto-electric, etc.), competing charge and/or spin order with superconducting ground state in high- T_c cuprates, long-range order in geometrically frustrated magnets, interplay of various degrees of freedom in multi-ferroic oxides, and field-induced novel order in spin-gap compounds, can be studied. Although such studies have been in the domain of neutron facilities for decades, the advent of modern synchrotron sources has created an unprecedented opportunity to reveal high-resolution structural, orbital, and magnetic structures by exploiting unique features of photon-atom scattering cross section. X-ray studies of materials in a 15 Tesla (T) superconducting (SC) magnet with very low-temperature (<300 mK) capability will go a long way in obtaining information on the atomically resolved structural and magnetic responses leading to a comprehensive understanding of phase diagrams of numerous materials.

Almost all studies of equilibrium phenomena in magnetic fields beyond 15T, on the other hand, have been limited to transport, thermodynamic, and some spectroscopic measurements. This is primarily because of the complexity involved in generating high fields using SC-resistive hybrid magnets coupled with high-resolution structural probes such as

synchrotron x-rays. Recent developments in SPring8 and ESRF and current activities at the APS utilizing high-field pulsed magnets offer a complementary approach. High-field pulsed magnets are not the solution to *all* problems in materials physics; but it is the *only* solution available to many that are of great interest requiring fields well beyond 15T. Design flexibility of coils to take advantage of specific scattering cross section and the possibility to produce fields beyond 40T make pulsed magnets quite desirable as well. Furthermore, by combining pulsed field with the natural timing structure of synchrotron radiation, structural and magnetic effects should be detectable on the microsecond to sub-millisecond levels with the possibility of pushing the temporal resolution to a faster timescale, ushering in studies of field-induced *non-equilibrium* phenomena which lie outside the reach of DC magnets. We propose to expand our ongoing pulsed-magnet initiative to generate long pulse (30 ms or more) high-field (30-60 T) magnets with a fast and efficient detection capability.

Added Value of the Mid-term Upgrade

We propose to upgrade our existing in-field scattering capabilities by (1) replacing the 12-year-old 13T magnet with a new 15T SC magnet incorporating a very low-temperature (<300mK) insert; (2) installing a new 1MJ bipolar capacitor bank for generating long-pulse (30 ms) ultra-high (30-60T) magnetic fields; and (3) procuring an APD array for fast 2D detection of scattered photons in pulsed fields.

The development of such a high-field instrumentation is very timely. In 2005, the National Research Council (NRC) enthusiastically recommended that "New instruments for studying the neutron and x-ray scattering properties of materials in high-magnetic fields should be developed in the United States". More recently, this position was reaffirmed in an NRC committee report on future directions in condensed matter and materials physics [CMMP-2010]. We note that both committees recognized the need for complementary structural probes and endorsed that high-field research instruments be placed at both neutron and x-ray facilities. Further, as a part of the strategic plan for future beamlines, the APS has proposed a large DC magnet (35-40 Tesla) user facility within the next decade, which has been endorsed by the SAC. In particular, SAC has recommended the establishment of a magnetic field facility in the near term to operate at ~13 Tesla that would satisfy current user demands and could later be incorporated into a high-field magnet beamline. The proposed set of instrumentation, i.e. a 15T SC magnet and long-pulse extremely high-field system along with a fast 2D detection system is, therefore, very appropriate and will act as stepping stones for building and expanding user community in high-field science at the APS and as well as be a catalyst for securing external funding for higher-field DC magnet facility in the future. Finally, we point out that both SPring8 and ESRF have SC and high-field pulsed magnet instruments for scattering studies, which have been in operation for several years. The proposed instruments will give the APS indisputable lead in the area of high-field science at synchrotrons for years to come.

Expected user communities

The expected user community of high-field instruments consists of scientists from various divisions here at Argonne National Laboratory (e.g. MSD, XSD) and researchers from universities (e.g. Stanford, MIT, McMaster, UCSD, Notre Dame, ISU) who require synchrotron techniques for studies of materials in high magnetic fields. Some specific examples of studies that will greatly benefit from the use of such a magnet are given below:

- Reduction in local rotational symmetry in spin-gap materials (Stanford)
- Competing order in high- T_c superconductors (MIT, APS)
- Release of frustration and long-range order in frustrated magnets (McMaster, MIT)
- Origin of electric polarization due to fields in functional materials (Rutgers)
- Magneto-striction at the vortex lattice melting in high- T_c superconductors (ND)
- Melting of polarons and charge order in magneto-resistive manganites (MSD)
- Heavy-fermion physics (UCSD)
- Quantum phase transitions and novel order in complex meta-magnetic systems (ISU)
- Field enhanced magneto-caloric effect and structural transitions (Ames Laboratory)
- Field tuning of precursor phenomena in ferromagnetic shape memory alloys (ANL)

Enabling technologies and infrastructure

Three key equipments are proposed to enable high-field science.

(1) 15T SC magnet with a low temperature (< 300mK) insert (\$1400K)

A dedicated 15T SC magnet with low-temperature capability for x-ray scattering studies will be essential to make progress in high-field science at the APS. The existing 12-year-old 13T magnet would require significant investment in efforts and costs to make it operational with a doubtful long-term reliability. In order to have a reliable intermediate-field SC magnet for general users that can be incorporated into a future high-field beamline a new 15T magnet is required.

(2) 1MJ bipolar capacitor bank (\$1250K)

Current pulsed magnet system at the APS generates ~1ms pulse with the peak field (30T) lasting for ~10µsec, restricting experiments primarily to structural effects. Since the duty cycle and repetition rate are inherently low for pulsed magnets generating as long a field pulse as possible is essential. A modular-designed bipolar 1 MJ capacitor bank will generate at least 30 times longer-duration pulses in half-sine mode and peak fields lasting for at least 15 ms in flat-top mode, respectively. Having a peak field duration some 1500 times longer will enable measuring low-count rate effects (*e.g.* scattering from CDW, magnetic and orbital order) in extremely high fields (40-60T), vastly expanding the number of problems that can studied in pulsed fields. A large bore enables very low-temperature sample inserts in split-coil geometry and provides large optical access in Voigt geometry, which is crucial for scattering experiments. Bipolar nature allows for magnetic dichroism measurements to be performed as well. The system is modular with three subunits, 100kJ base unit (\$350K), 400kJ (\$300K), and 500kJ (\$600K), respectively. We should emphasize that these power supplies are modular therefore the initial unit could be used for immediate experiments before all three are installed.

(3) 2D detection with fast temporal resolution (\$250K)

A large APD array will enable data collection over a significant region of reciprocal space. This is critical for pulsed-field experiments in order to monitor the location of the Bragg reflection through the entire pulse duration.

Partnerships and user interest

Prof. P. Abbamonte, Dept. of Physics, University of Illinois Urbana Champaign

Dr. G. Boebinger, Director, National High Magnetic Field Laboratory

Prof. P. C. Canfield, Dept. of Physics and Astronomy, Iowa State University

Dr. C. Detlefs, European Synchrotron Radiation Facility

Prof. M. R. Eskildsen, Physics Department, Notre Dame University

Prof. I. R. Fisher, Department of Applied Physics, Stanford University

Prof. B. D. Gaulin, Dept. of Physics, McMaster University

Prof. A.I. Goldman, Dept. of Physics and Astronomy, Iowa State University

Dr. J. P. Hill, Physics Department, Brookhaven National Laboratory

Dr. E. Isaacs, Center for Nanoscale Materials and Professor, University of Chicago

Prof. V. Kiryukhin, Department of Physics, Rutgers University

Prof. Y. S. Lee, Department of Physics, Massachusetts Institute of Technology

Prof. Z. Mao, Physics Department, Tulane University

Prof. B. Maple, Department of Physics, University of California, San Diego

Dr. Y. Narumi, Institute for Solid State Physics, University of Tokyo

Dr. C. S. Nelson, NSLS, Brookhaven National Laboratory

Prof. H. Nojiri, Institute for Materials Research, Tohoku University

Dr. Y. Ren, Argonne National Laboratory

Dr. J. Singleton, Los Alamos National Laboratory

Prof. S. K. Sinha, Department of Physics, University of California, San Diego

Dr. D. Vaknin, Iowa State University and Ames Laboratory

Dr. U. Welp, Materials Science Division, Argonne National Laboratory

Industry and technology transfer

None is foreseen at this time.

Budgetary Profile

| Total: | | \$2900k |
|---|--------|---------|
| 2D APD pixel array detector | | \$ 250k |
| 500kJ expansion slave unit | \$600k | \$1250k |
| 400kJ capacitor expansion unit | \$300k | |
| 100kJ base unit with control electronics | \$350k | |
| 1MJ bipolar capacitor bank | | |
| 15T SC magnet with a low temperature (< 300mK) insert | | \$1400k |