Intermediate Energy X-ray Beamline at the Advanced Photon Source

1. Science Drivers

One of the grand challenges for condensed matter physics in the 21st century is to understand the physics of materials that exhibit competing interactions. We refer specifically to materials in which the energy scales of valence bonding, Coulomb repulsion, and the kinetic energy of mobile electrons are similar in size, and the resulting ground state is a subtle compromise between these effects. Such materials exhibit a startling array of electronic phases and a heightened sensitivity to extrinsic perturbations such as changes in temperature, applied electric or magnetic fields, pressure, or nanopatterning. Two common examples are the manganese-oxides, in which small changes in applied magnetic field or temperature lead to large changes in magnetization or resistance, and the copper-oxides, which with increasing carrier density evolve from an antiferromagnet to a spin glass to a *d*-wave superconductor to a regular metal. These effects are driven, it is believed, by a competition between valence bond order and the kinetic energy of valence electrons. Other examples of such systems are the spin ladder compounds, which have a spin gap and show charge ordering but become superconducting under pressure, and the triangular cobaltates which show frustrated ferrimagnetism and charge order but superconduct with the intercalation of water. In most of these cases low-dimensionality plays a crucial role by amplifying the quantum fluctuations.

Two conspicuous traits of materials with competing interactions are (1) a complex electronic excitation spectrum, resulting from the emergence of low energy collective modes, and (2) the emergence of new electronic length scales. To investigate these two categories of phenomena we propose to construct an intermediate energy beam line at the Advanced Photon Source (APS) to operate in the regime from 0.2 to 2.0 keV. This facility, called Intermediate Energy X-ray Collaborative Development Team (IEX-CDT), will serve two experimental techniques – "high" energy angle-resolved photoemission spectroscopy (ARPES), which is a *bulk* probe of the low energy excited states in condensed matter, and resonant soft x-ray scattering (RSXS), which is a direct probe of electronic ordering. ARPES and RSXS complement one another scientifically, are of interest to the same scientific community, and in particular have *highly compatible source and beam line technical requirements*. For this reason the PIs (Juan Carlos Campuzano and Peter Abbamonte) have formed a partnership to investigate the physics of these materials, and others yet to be discovered, and more broadly to bridge the realms of synchrotron scattering and spectroscopy.

ARPES determines electronic structure by measuring the kinetic energies of electrons following irradiation with monochromatic x-rays. Such experiments aim at elucidating the low energy electronic excitations in solids, which govern most of their physical and chemical properties. To date, the energy resolution requirements of ARPES (10-30 meV) have required the use of relatively low energy x-rays (<100 eV). But unfortunately, the inelastic mean free path of the electrons is only ~0.5 nm at these energies, so the surface makes a major contribution to the spectra. By increasing the incoming photon energy, however, the mean-free path of electrons increases proportionally. For example, for 1 keV photons, the mean free path of electrons reaches 2-3 nm, making it possible to study bulk properties of materials. To be more quantitative, in a simple exponential attenuation model the surface fraction (SF) is $1 - \exp(-d/l)$, where *d* is the "surface" length and *l* is the mean free path. For conventional ARPES (~50 eV), *d* = 0.5 nm and *l* = 0.5 nm resulting in an SF = 0.63. However, by going to 1000 eV, *l* increases to 2.5 nm, resulting in a SF = 0.18. Therefore, the bulk to surface contrast is increased by 350%. Additionally, by varying the photon energy say between 200 eV and 2000 eV, a comparison of the electronic structure of the bulk to that of the surface is possible.

RSXS is a new diffraction technique in which sensitivity to charge and spin ordering is achieved by tuning the x-ray energy to atomic transitions which weight the valence band. Intermediate energy x-rays (0.2 < E < 2.0 keV) are particularly powerful in this regard as they access the K edges of N through S, the L edges of the transition metals, and the M edges of the rare earths, which probe the valence bands of most known strongly correlated electron systems. RSXS is therefore the natural probe of any kind of translational symmetry breaking in these systems and has recently been used, for example, to study orbital ordering in manganites [Wilkins, 2003; Thomas, 2004; Dhesi, 2004], Wigner crystallization in spin ladders [Abbamonte, 2004], and spectral weight modulations in stripe phases [Abbamonte, 2005]. Unfortunately, facilities in the United States for RSXS are currently inadequate. The proposed facility will provide full angular flexibility, access to liquid helium temperatures (< 10 K), polarization selectivity, fluorescence rejection optics for reducing the background known to plague this technique, and a factor of 50 more intensity than the most powerful source currently available.

2. Role of the APS

The combination of a high-energy 7 GeV storage ring and an appropriately chosen insertion device, such as an APPLE II type elliptically polarized undulator, places APS in the unique position to develop an intermediate energy x-ray facility. As seen in Fig.1, the on-axis brilliance of the proposed 2.4 m long APPLE II device with a 7.8 cm period will deliver the world-highest brilliance in the 0.2 to 2.0 keV range. High brilliance is very important because it allows an improved performance from a grating monochromator.

This broad tuning curve can be only achieved by a 7 GeV stored particle beam. The calculation was based on the current 5 m available straight sections assuming that a maximum of two of these devices can be placed in a straight section. However, in the APS Renewal plan, standard straight sections will be extended to 8 m, therefore making it possible to install three, even slightly longer devices on line resulting in increase of delivered flux. The flux will be even further increased because the accelerator upgrade will enable APS to run at a beam current to 200 mA, twice compared to existing operation. This is very important because one of the scientific objectives of the IEX-CDT is to reach 10-30 meV resolution between 0.2 to 2.0 keV with variable polarization. To simultaneously provide sufficient flux and high resolution, the proposed extension of the straight section to 8 m length, the increased beam current and an optimized insertion device combined together will provide enormous benefits to IEX scientific programs.

The development of specialized devices such as the APPLE II customized for a specific experimental need is one of improvements proposed in the APS Renewal. The APPLE II source has the advantage of providing radiation with variable linear polarization and circular polarization. In the circularly polarized mode, studies of magnetic structures and excitations can be facilitated, and in addition the source does not produce significant harmonics – an important consideration for all the experiments. Yet another consideration is the fact that a stored 7 GeV electron beam is more stable compared to lower energy (3 GeV) storage rings. This stability invariably transfers to a more stable photon beam, a very desirable condition for all the experiments, particularly for those requiring long data acquisition times.

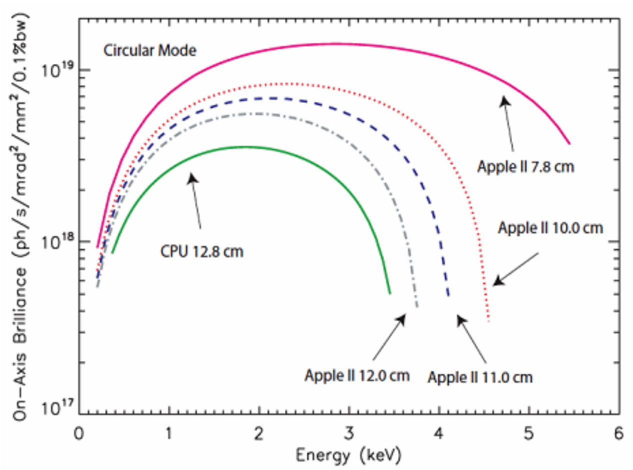

Figure 1: On-axis brilliance tuning curves for circular polarization for the proposed APPLE II device with period ranging from 7.8 to 12.0 cm. Curves are compared to the existing circularly polarized undulator in Sector 4, with a period of 12.8 cm. Calculations are done for 2.4 m long devices of 100 mA beam current.

Finally, the size of the experimental hall at the APS will make it possible to build a long beamline of the order of 80 m, a pre-requisite for high-resolution performance of reflective optics used in this energy range.

In summary, the APS already has a potential to provide x-rays in the 0.2 to 2.0 keV energy range with unprecedented brilliance, tunability, polarization characteristics and stability. With the proposed APS Renewal, the brilliance will be even higher, providing conditions to further propel IEX programs to world-class level.