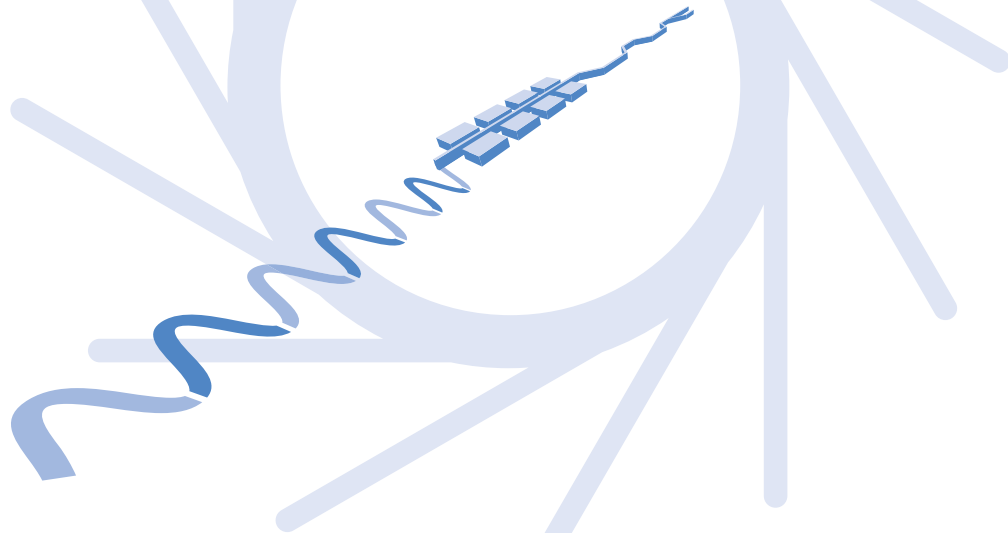# Scientific Needs for Future X-Ray Sources in the U.S.

A White Paper

Based on a study group co-chaired by Roger Falcone and Joachim Stöhr, and members: Uwe Bergmann, John Corlett, John Galayda, Jerry Hastings, Robert Hettel, Zahid Hussain, Janos Kirz, Bill McCurdy, Tor Raubenheimer, Fernando Sannibale, John Seeman, Z.-X. Shen, Robert Schoenlein, and Alexander Zholents.

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Contents

Contents

Preface

This document is the result of collaboration among scientists associated with the Lawrence Berkeley National Laboratory (LBNL) and the SLAC National Accelerator Laboratory (SLAC), including faculty of the University of California, Berkeley, and Stanford University. The collaboration consisted of scientists from a broad range of scientific disciplines and included experts in x-ray and accelerator science, two core competencies of the Laboratories. The collaboration benefited from the broad combined knowledge base at the two Laboratories, including expertise in x-ray science dating back to pioneering x-ray experiments in the 1970s at SSRL and in ultrafast science in the 1980s at LBNL, existing forefront x-ray programs on the third-generation storage rings ALS and SPEAR-3, considerable current expertise in ultrafast laser and x-ray science at both Laboratories, and forefront knowledge about next-generation sources based on the construction of and the scientific user program at LCLS, the first x-ray laser, to be commissioned in 2009. Information was also provided and coordinated with external experts and colleagues from around the world.

ONE

Introduction: Science and X-Rays

Many of the important challenges facing humanity, including developing alternative sources of energy and improving heath, are being addressed by advances that demand the improved understanding and control of matter. While the visualization, exploration, and manipulation of macroscopic matter have long been technological goals, scientific developments in the twentieth century have focused attention on understanding matter on the atomic scale through the underlying framework of quantum mechanics. Of special interest is matter that consists of natural or artificial nanoscale building blocks defined either by atomic structural arrangements or by electron or spin formations created by collective correlation effects (Figure 1.1).
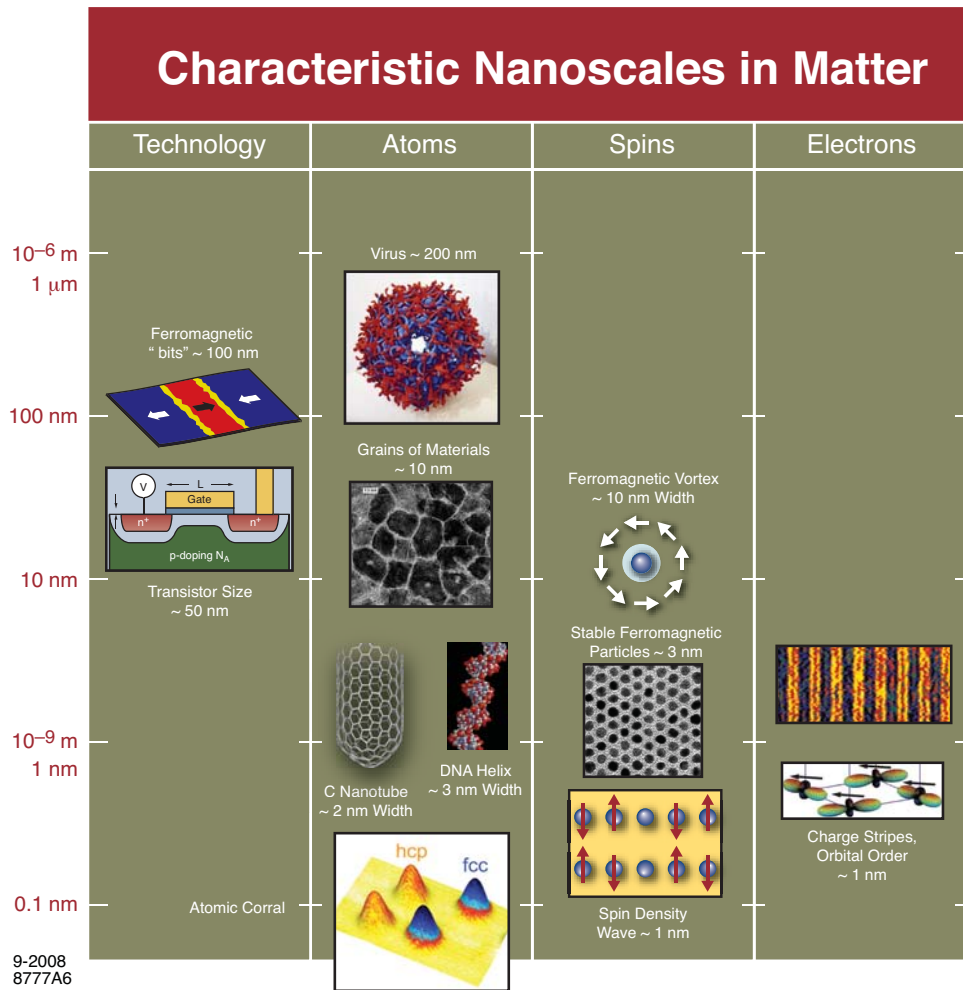
Figure1.1. Illustration of characteristic features on the level of atoms, electrons, and spins on nanometer length scales in comparison with typical lateral length scales in present advanced technological devices.

The essence of the challenge to the scientific community has been expressed in five grand challenges for directing matter and energy recently formulated by the Basic Energy Sciences Advisory Committee [1]. These challenges focus on increasing our understanding of, and ultimately control of, matter at the level of atoms, electrons. and spins, as illustrated in Figure 1.1. Meeting these challenges will require new tools that extend our reach into regions of higher spatial, temporal, and energy resolution.

Since the fundamental interaction that holds matter together is of electromagnetic origin, it is intuitively clear that electromagnetic radiation is the critical tool in the study of material properties. On the level of atoms, electrons and spins, x rays have proved especially valuable.

A. Brief History of X-Ray Science

The understanding of matter was revolutionized by Röntgen's discovery of x rays in 1895, and since that innovation, four invaluable and well-known uses of x rays have emerged: (1) medical imaging, first demonstrated by Röntgen himself; (2) x-ray diffraction revealing the atomic structure of matter, pioneered by von Laue and the Braggs; (3) x-ray absorption and emission spectroscopy by Barkla, uncovering the complete electronic shell structure of atoms; (4) photoelectron spectroscopy revealing the signature of bonding in molecules and solids, pioneered by Siegbahn, Spicer, and Turner, followed by demonstration of its spin dependence by Siegmann.

Before the advent of dedicated synchrotron radiation sources in the early 1970s, these applications of x rays had not only revolutionized our knowledge of matter on the fundamental level of atoms, electrons, and spins but also redefined entire fields of science, such as physics, chemistry, biology, and medicine. The 19 Nobel Prizes that have been awarded for related work, as summarized in Figure 1.2, prove the impact of x rays.
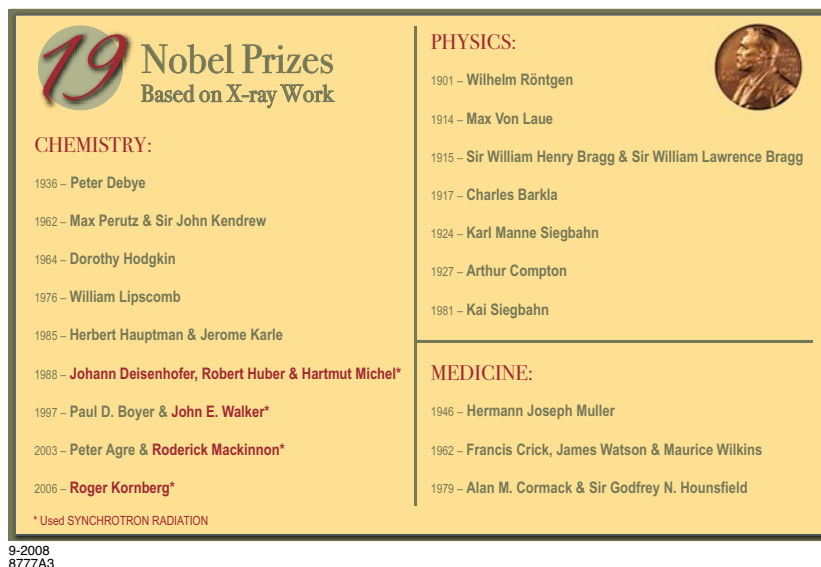
Figure 1.2. The impact of x rays is proven by 19 Nobel Prizes that have been awarded for related work.

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[1] *Directing Matter and Energy: Five Challenges for Science and the Imagination.* Basic Energy Sciences Advisory Committee, U.S. Department of Energy, December 20, 2007. Available online at www.sc.doe.gov/bes/reports/list.html.

While most of these x-ray accomplishments were based on the use of conventional x-ray tubes, as of today four Nobel Prizes can be linked to the use of x-ray synchrotron radiation. Based on the widespread application of synchrotron radiation for the solution of scientific problems it is safe to predict that this picture will change with time.

B. Modern X-Ray Techniques

Key developments in x-ray science over the last thirty years are based on the unique properties of modern synchrotron radiation sources: high brightness and coherence, energy tunability and high energy resolution, polarization control, and pulsed time structure. The key capabilities of x rays in the light of the grand challenges are illustrated in Figure 1.3.

X rays penetrate matter and reveal the "invisible" interior of complex objects, such as living organisms. More recently in the era of nanoscience, x-ray imaging, in the form of both real-space microscopy and inversion of reciprocal-space coherent scattering patterns ("speckle"), has provided high-resolution images on the nanometer length scale of charge and spin distributions in a sample. When combined with tunability and polarization control, x rays can yield element- and chemical-state specific images together with magnetic contrast. Imaging techniques may be carried out both in a surface-sensitive mode with an electron microscope as in photoemission electron microscopy (PEEM) and in a bulk-sensitive mode by use of x-ray transmission. Microscopy is often carried out today in combination with pump–probe studies to reveal nanoscale dynamics on the tens of picosecond time scale (limited by the x-ray pulse length from storage-ring sources). Coherent-scattering studies, carried out in a photon-correlation spectroscopy mode, are used to reveal equilibrium fluctuations on the time scale of seconds. The multi-dimensional combination of nanoscale spatial resolution with temporal resolution down to femtoseconds, even attoseconds, constitutes one of the greatest opportunities of future x-ray studies of matter.

Seeing the invisible

Where are the atoms?

Where are the electrons?

Where are the spins?

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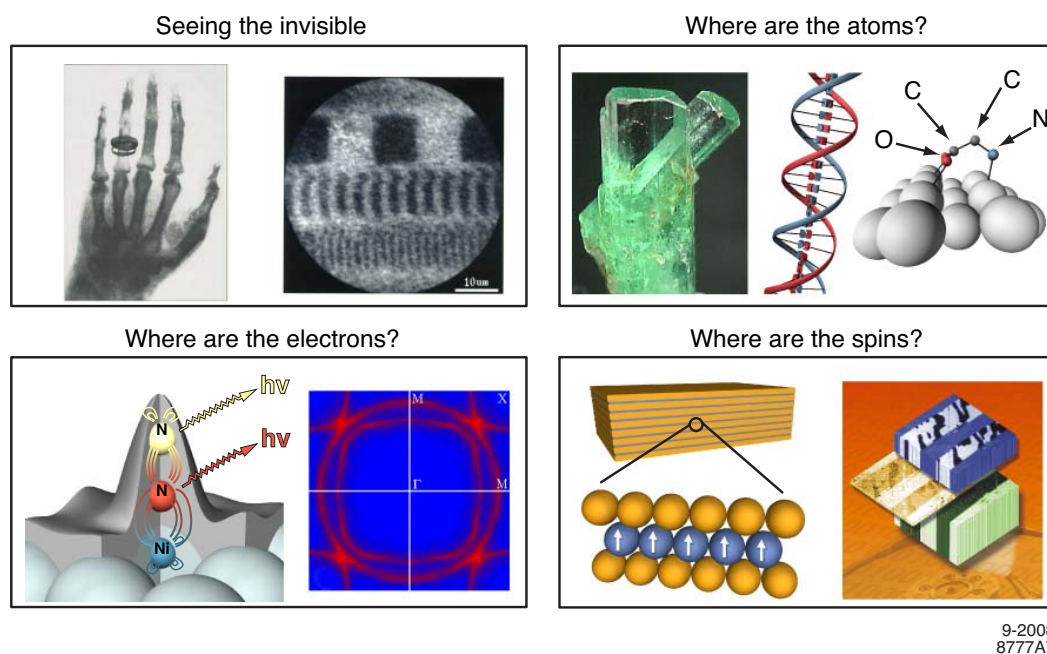
Figure 1.3. The basic capabilities of x rays that provide the basis for the new developments discussed in the text.

X rays are often divided into two major types, reflecting their wavelength, photon energy, and penetrating power, so that the types of experiments to which each is most suited differ. The short wavelength of high-energy, hard x rays (~ 10 keV) enables the determination of the atomic structure of matter, yielding the relative positions of atoms in materials. While the inversion of x-ray diffraction patterns has long been used to give the real-space atomic structure of matter, about 80% of new macromolecular structures determined today by this method use synchrotron radiation. In particular, the technique of multiple-wavelength anomalous diffraction (MAD) has revolutionized macromolecular crystallography. The most difficult and challenging problems, such as large complexes and molecular machines, are almost exclusively solved with synchrotron radiation. By use of coherent x-ray scattering, it is possible today to determine the three-dimensional atomic structure of nanometer-sized objects. The tunability of the photon energy has led to the widespread use of x-ray absorption in the form of the extended x-ray absorption fine structure (EXAFS) for the study of the precise atomic structure around selected atoms. A key component of future studies of matter with hard x rays is the combination of atomic resolution with temporal resolution on the sub-picosecond time scale of atomic motion.

The somewhat longer wavelength of lower-energy, soft x rays (~ 1 keV) enables the investigation of the electronic structure of matter. Soft x-ray spectroscopic studies include near-edge x-ray absorption fine structure (NEXAFS), x-ray emission spectroscopy (XES), and photoemission (PES). These techniques yield information on the electron distribution in occupied and empty orbitals in molecules and the electronic energy bands in solids, providing information on how atoms are bound together in solids, liquids, and molecules. Linear-polarization-dependent NEXAFS and XES studies have provided detailed information of directional bonding and orientational order at surfaces. Photoemission studies have revealed the detailed electronic structure of molecules and the spin-dependent band structure of solids, and angle-resolved photoemission spectroscopy (ARPES) has evolved into one of the most important tools in condensed-matter physics. Key future development areas of soft x-ray spectroscopic techniques are their extension to the multi-dimensional phase space of length scales down to nanometers, time scales down to femtoseconds, or even attoseconds, and energy scales to tens of micro-eV.

Not shown in Figure 1.3 are the recent developments in inelastic x-ray scattering that allow probing elementary excitations of the lattice as well as of the electronic system. This has become possible by coupling to the intrinsic linear momentum of high-brightness hard x-ray beams that are monochromatized to the meV scale. Similar to inelastic neutron scattering, such studies provide details of the excitations of the atomic lattice but are complementary to neutrons in emphasizing charge rather than spin excitations in the electronic system. A particularly important but as yet insufficiently employed variant of inelastic x-ray scattering uses core-electron excitations. Such x-ray Raman spectroscopy uses a penetrating beam of hard x rays and through "x-ray energy loss spectroscopy" provides spectra like x-ray absorption for low-Z atoms in bulk samples that cannot be measured by means of strongly absorbing soft x rays. Examples are vacuum-incompatible samples like liquids and materials under extreme environments like those in diamond-anvil cells.

The angular momentum of circularly polarized x rays has allowed access to the angular momentum of electrons and consequently the ability to quantitatively separate the spin and orbital parts. X-ray magnetic circular dichroism (XMCD) studies preferentially use soft x rays, where the magnetic effects are largest, for either x-ray absorption spectroscopy or resonant scattering. Nanoscale information is obtained via real-space microscopy using zone-plate lenses or reciprocal-space coherent scattering ("lensless imaging"). For modern magnetic materials, which typically come in the form of thin films, multilayers, and

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nanostructures, such studies have replaced neutron scattering as the technique of choice because of their elemental specificity and sensitivity due to large cross-sections (see Figure B.1 in Appendix B).

While the above discussion amply demonstrates the power of x rays for the study of matter, the question arises whether the capabilities of x-ray sources that exist today or are under construction are sufficient to address the scientific challenges before us. Specifically, we note that control of matter will ultimately flow from the greatest understanding of the underlying structure and properties of the atoms and bonds that underlie matter and that yield an incredibly rich set of properties that range from superconductivity, to conversion of sunlight to electricity, to catalytic conversion of greenhouse gases, to miracle drugs.

We have concluded that the nation's scientific needs will not be met by current facilities and those under construction, and in the next sections of this document we demonstrate that the scientific needs for new sources are clear and that the technological readiness to construct suitable sources is within our reach.

TWO

Overview of Scientific Drivers and Required Enhanced X-Ray Capabilities

In this section we give a brief overview of the scientific challenges associated with the understanding and control of matter at the level of atoms, electrons, and spins in the domains of space, time, and energy, and we outline four key x-ray capabilities that are required to address the scientific challenges. A more detailed discussion of the science drivers will be given in Section 3, and the basic types of x-ray sources that can provide the required capabilities are discussed in Section 4.

A. Scientific Drivers

While there is compelling historical evidence that the construction of new x-ray sources by itself has served as scientific enablers leading to originally unimagined applications, here we take the approach of using identified scientific challenges to demonstrate a need for future x-ray capabilities.

The innovation expected from the construction and operation of enhanced x-ray sources lies in combining and extending existing knowledge of the nanoworld, illustrated in Figure 1.1, with extreme time and energy resolution. Properties of anticipated new x-ray sources include the ability to reach to the frontier of ultrafast timescales of electron motion around an atom, the spatial scale of the atomic bond, and the energy scale of the bond that holds electrons in correlated motion with near neighbors. In addition, these novel sources have an intensity and brightness needed to observe the subtlest of nature's secrets at these frontier space, time, and energy scales.

While our understanding of the time- and energy-dependent behavior of matter in its ground or natural state is still severely limited, we know even less about excited states. Such states are of key scientific and technological importance, for they typically determine how matter functions during chemical reactions and during physical and biological processes. Excited states of interest span a vast range, being close to the natural (ground or equilibrium, as appropriate) state (as in electronic transport), relatively far from it (as in photo-chemical reactions), and very far from it (as in extreme conditions that can be imposed by pressure, radiation, or electric and magnetic fields).

To briefly illustrate the need for advanced x-ray capabilities, we below give two examples of what we need to learn. They are related to "chemical reactivity" and "complex materials," two of the five important scientific areas that are discussed in more detail in Section 3.

- Knowing the importance of energy in guiding chemical reactions, we envision driving chemical transformations by controlled optical or infrared pulses and understanding the atomic and electronic transformation by means of advanced x-ray spectroscopies. More specifically, we plan to capture, with snapshots on the femtosecond timescale, the making and breaking of chemical bonds and the crucial transition-state intermediates in chemical reactions.

- We foresee development of an understanding of the origins of nanoscale charge and spin order and their dynamics in correlated materials through high-resolution energy- and time-dependent x-ray spectroscopies. We plan to visualize through ultrafast x-ray motion pictures the performance limits of materials, e.g., the speed limit of reliable switching of a spintronics device.

Embedded in these challenges is the exploration of the atomic or nanoscale on the "natural" time scale of atoms, electrons, and spins and on the "operational" time scale that determines function and is the key in technological applications. As illustrated in Figure 2.1, there is presently a striking discrepancy between the natural time scales of atomic motion (about 100 fs), spin motion (down to about 1 fs), and electronic motion (down to attoseconds) and the fastest operational timescales (approaching 100 ps). A critical scientific challenge is to pave the way for technology to expand into the ultrafast, with the opportunity of five orders of magnitude improvement down to the intrinsic time scale of charge and spin motions of valence electrons.
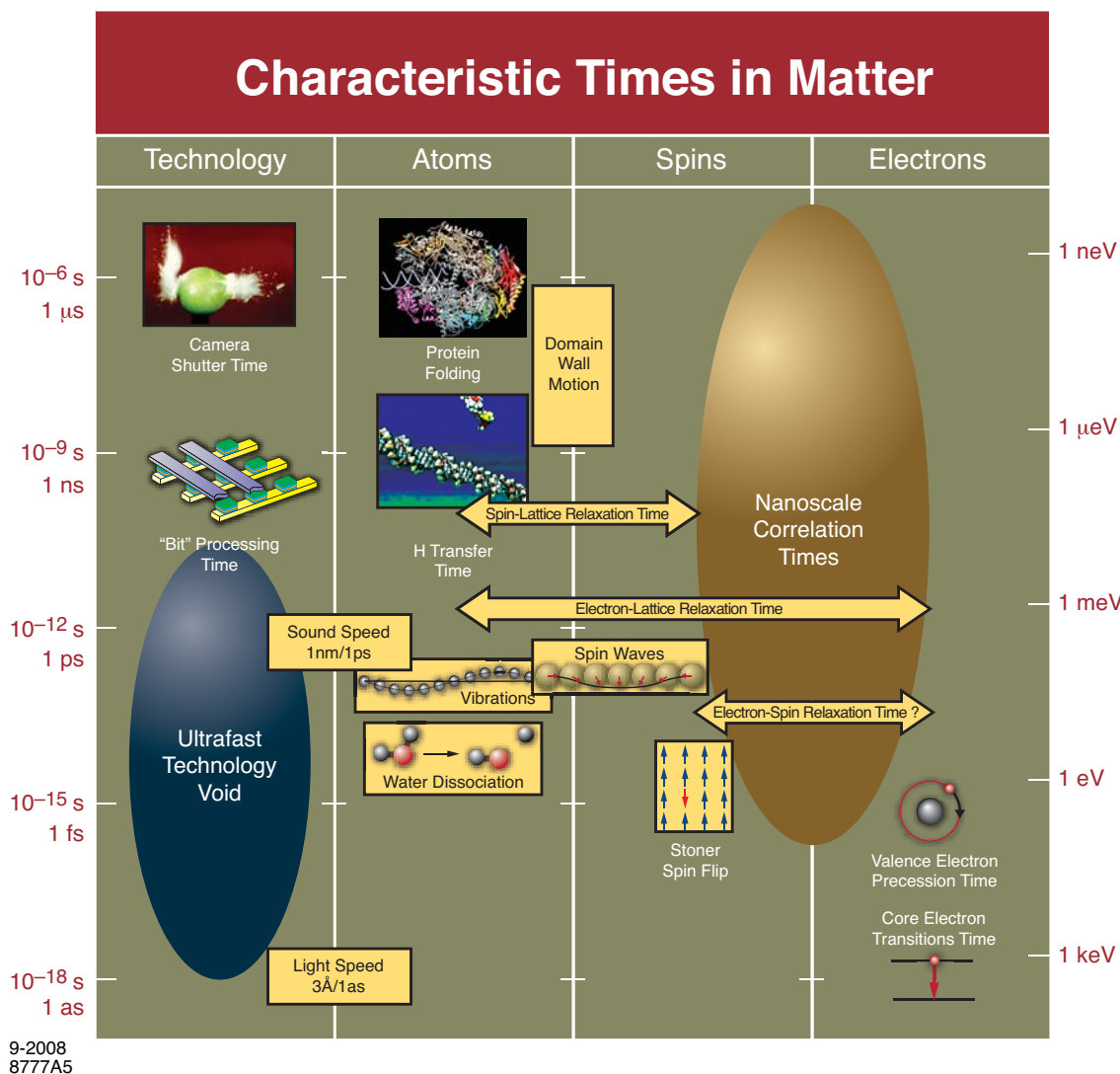
Figure 2.1. Illustration of our present knowledge about typical times involved in the interactions of atoms, electrons, and spins. On the right, we have indicated the corresponding quantum-mechanical interaction energies as estimated from the energy–time correlation $\Delta E \cdot \Delta t = h \sim 4$ fs·eV.

The unique strength of future x-ray sources lies in their ability to combine coverage of the atomic and nanoworld with the entire time domain from seconds to attoseconds. In Section 3, we provide more specific examples of scientific challenges in different areas of science that call for enhanced x-ray capabilities.

B. Essential New X-Ray Capabilities

Here we briefly describe four key performance areas where significant advances in x-ray capabilities are both required by the science, and technically feasible in the near-term. Not only are these individual requirements beyond the capabilities of present light sources, the paramount scientific challenges often require a source providing several of these features simultaneously.

1. X-Ray Time Structure—Complete Control of Longitudinal Phase Space

- Pulse duration and temporal resolution extending to the attosecond regime.

- Fourier-transform-limited pulse structure extending from few-picosecond pulses with meV bandwidths to sub-femtosecond pulses with 10-eV bandwidths.

- Control of longitudinal pulse shape, amplitude and phase.

- Synchronization and full integration with conventional pulsed laser sources.

Access to fundamental time scales (and energy scales) combined with atomic resolution, element specificity, valence-electron sensitivity, etc. is a requirement that emerges throughout the scientific grand challenges. Initial work at third-generation synchrotrons has launched this research area, and first-generation free electron lasers (FELs) will provide an important advance in this emerging field. A key missing capability is longitudinal coherence (and pulse control) that is achievable using advanced seeding techniques and is essential to maximize the scientific impact of future short-pulse x-ray sources. Concurrently "conventional" laser sources must be synchronized and fully integrated with future short-pulse x-ray sources in order to provide for tailored ultrafast sample excitation spanning the UV, visible, infrared, and THz regimes.

2. Full Transverse Coherence

- Requirements set by real-space imaging, diffractive imaging, and photon-correlation spectroscopy.

- Optical systems to preserve and exploit transverse coherence.

Present ring-based sources provide diffraction-limited radiation in the vertical plane up to several keV and in the horizontal plane up to ~100 eV (see Figure 4.3 below). Extending these capabilities to higher photon energies (at high average brightness) is essential for advanced x-ray imaging of nanostructures, domains, and emergent phenomena that are manifest through phase separation, domain fluctuations, etc. At the same time, enhanced coherent flux will allow time-domain measurements to be extended from seconds to microseconds. X-ray FEL sources now in construction provide transverse coherence, but the usable flux may be limited by sample damage. Therefore higher repetition rates and lower peak brightness may be required in addition to tunability and extended energy range.

TWO
Overview of Scientific Drivers and Required Enhanced X-Ray Capabilities

3. High Average Flux and Brightness

- Quasi-CW (up to 1 GHz) sources with average flux and/or brightness substantially beyond existing sources in the hard and/or soft x-ray range.

- Short-pulse sources with high repetition rates (up to 1 MHz) providing average flux and/or brightness substantially beyond existing sources in the hard and/or soft x-ray range.

The tremendous scientific promise of x-ray Raman, inelastic x-ray scattering and related x-ray tools is significantly limited by the flux (photons/bandwidth) available from present x-ray sources in both the hard and soft x-ray ranges. The scientific impact of present short-pulse x-ray sources is similarly limited by the available peak and average flux and brightness. Furthermore, time-resolved experiments and nonlinear x-ray science at short-pulse sources require the ability to trade-off peak and average brightness for specific applications, including correlation spectroscopy.

4. Tunability, Polarization Control, and Extended Photon Energies

- Soft x-ray tunability (throughout the transition-metal L-edges) and polarization control and modulation.

- Hard x-ray tunability and energy range extending to 20 keV.

While tunability and polarization control are well developed at third-generation synchrotron sources, high-sensitivity measurements with future rings require the ability to modulate the polarization at high frequencies (kHz), a capability presently not available. Polarization control will also be an important capability at future short-pulse x-ray sources, especially in the soft x-ray range, to exploit powerful spectroscopy techniques (e.g., NEXAFS, XMCD, x-ray magnetic linear dichroism or XMLD, etc.) for probing valence charge, spin, bonding dynamics, and emergent phenomena on fundamental time scales. Tunability is essential across the entire spectral range for elucidating the dynamics of local atomic structure (e.g., via EXAFS), providing element specificity, penetration capability, interface sensitivity etc.

THREE

Examples of Scientific Drivers for Future X-Ray Sources

This section presents examples of scientific challenges in five areas where advanced x-ray capabilities are envisioned to contribute to breakthroughs in our knowledge. The chosen areas are (1) atomic and molecular science—studying fundamental processes involving atoms, electrons and spins (2) chemical reactivity—discovering the key processes related to energy production, climate change, and environmental processes, (3) complex materials—the core subject of condensed-matter physics and engineering sciences, which often drive technological advances, (4) materials under extreme conditions—revealing new synthesis pathways of tailored matter and discovering the limits of materials stability, performance, and function, and (5) life sciences and soft matter—studying the structure and dynamics of organic building blocks and promising the improvement of health.

While the above topics were chosen to cover key societal issues, our discussion is necessarily incomplete. It could be easily extended to other scientific areas. Furthermore, we believe that the proposed x-ray facilities will enable research on yet unidentified scientific and technological topics that will emerge over the next two decades. The present section should be read with this understanding.

A. Understanding and Controlling Electronic, Atomic, and Molecular Dynamics on Their Natural Timescales

The first of the challenges in the BES grand challenges report is "Quantum control of electrons in atoms, molecules, and materials." This issue is now a central one for the scientific community because we are at a point of transition in our study of the most basic questions in chemistry and materials science. For a century we have focused on how electrons, which are the "glue" of chemistry, determine the properties of materials and direct chemical and physical changes. We are now at the dawn of the new science of quantum control where we will go beyond probing what is there in molecules and materials to controlling processes through the direct manipulation of the electrons. Our primary tools for this task are short pulses of electromagnetic radiation that were made possible in recent decades by the advent of ultrafast laser pulses in the infrared (IR), visible, and ultraviolet (UV). Powerful, ultrafast pulses in the extreme ultraviolet (EUV) and x-ray regions form the other half of the tool kit that will open the doors to a complete and successful experimental attack on this most fundamental of challenges.

At stake are the solutions to some of the world's most important technological problems where quantum physics and chemistry play a central role. How can we produce enough energy? How can we make computers smaller, faster, and cooler, and what are the true limits of computation? How can we detect harmful pathogens early when their concentrations are almost vanishingly small? Most fundamentally, how can we make quantum-scale systems work for us? To answer these questions we will have to go beyond the simple pictures of electron orbitals and the separation of the timescale of electron motion from that of the nuclei in a molecule, on which, until now, our physical and chemical intuition have been based.

1. Attosecond Manipulation and Control of the Correlated Motion of Electrons

In the valence shells of atoms and molecules, where bonding forces are created, the natural timescale of the motion of electrons responsible for generating the most important properties of molecules is of the order of a few hundred attoseconds. Recently, sub-femtosecond pulses in the soft x-ray regime have opened up the possibility for direct study and manipulation of electronic motion on that timescale. The experiments that will do so in the future are dramatically more challenging than those being undertaken today, in terms of both detection and combinations of multiple radiation pulses, and will rely on the robust detection of signals from effects that are almost undetectable today. To be successful, they will require flexible and intense soft x-ray sources that provide those signals and that will allow multicolor x-ray experiments.

The essential idea of these experiments is to catch electron dynamics in the act during transitions between electronic states caused by any perturbation. One class of experiments will first pump an atom or molecule into an excited state, for example an autoionizing state with a finite lifetime, and then probe it with a sub-femtosecond pulse during that lifetime to ionize or doubly ionize the target. These experiments will make use of momentum-imaging detectors of the electrons and ions produced in coincidence to give a complete momentum map of the final state, a map in which the signatures of correlation in the initially pumped metastable state and its decay are written. Other experiments will probe transitions between electronic states caused by the motion of nuclei in molecules when those electronic states are close in energy. Experiments such as these will push the limits of FEL sources, since they require: tunable and intense sub-femtosecond pulses, synchronization with other sources (lasers, high-harmonic generation or another FEL pulse), and high repetition rates.

2. Understanding Chemical Dynamics When the Timescales of Nuclear and Electronic Motion Can Coincide

While the natural timescale of electron dynamics is of the order of a few hundred attoseconds, that of the motion of the atoms themselves during a reaction or as they vibrate in a molecule is of the order of tens to hundreds of femtoseconds (see Figure 2.1). However, when the nuclei in a molecule are near the molecular geometries where two electronic states have the same energy, the collective motion of the electrons as they make a transition from one state to the other can match the slower timescale of the motion of the nuclei. This phenomenon, shown schematically in Figure 3.1, is commonly called the "breakdown of the Born-Oppenheimer approximation." Up to now, this approximation has provided the conceptual basis for most of our understanding of chemical reactions. The grand challenges report identifies this phenomenon as a central challenge to our understanding of photosynthesis and the photochemistry of the first step in vision, for example. The surprise is that in large molecules involved in photochemical processes like photosynthesis, there are generally many such intersections, and nature has made use of them to control the flow of energy and chemical change.

Soft x-ray FELs will open new doors to understanding and exploiting these phenomena. In conventional femtosecond photochemistry, a vibrational wave packet is created within a single electronic excited-state and the subsequent molecular dynamics are determined by the Born-Oppenheimer potential-energy surfaces on which the wave packet evolves. The advent of extreme ultraviolet (EUV)/soft x-ray attosecond pulses affords access to a new excitation regime that cannot be described within the Born-Oppenheimer approximation. For example, a 300-as pulse has a bandwidth of 6 eV, sufficient to create a novel type of electronic wave packet, i.e., a coherent superposition of multiple electronic states. The possibility of determining the character of that wave packet opens an entirely new dimension for molecular control. For

example, the coherent interaction of electronic wave packets at conical intersections has the potential to control the branching between electronic states. Phase and amplitude shaping of the attosecond excitation pulse enables complete manipulation of the initial electronic (and vibrational) wave packet and its evolution. Attosecond soft x-ray probe pulses will be essential for following the electronic and molecular dynamics via time-resolved photoelectron spectroscopy and/or x-ray absorption spectroscopy techniques.
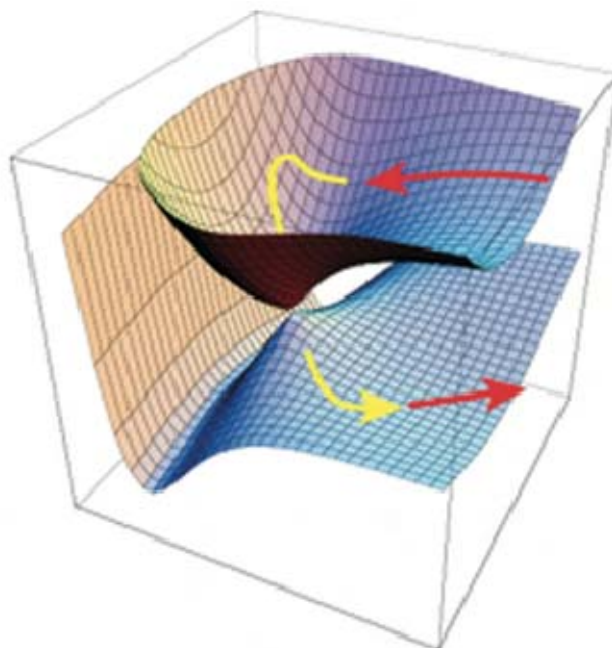
Figure 3.1. Example of a conical intersection, the point of contact between the potential-energy surfaces of two different electronic states in a molecule. [Source: BESAC grand challenges report]

3.   Detecting the Correlated Motion of Electrons with a New Kind of X-Ray Spectroscopy

In the past decade, dramatic advances have been made in optical correlation spectroscopy, a descendant of transient four-wave mixing, in which carefully timed sequences of femtosecond pulses are used to probe the vibrational and electronic structure of matter. This approach is analogous to multidimensional nuclear magnetic resonance (NMR) spectroscopy, which probes the atomic structure of small molecules and for which the 1991 Nobel Prize in Chemistry was awarded. Optical correlation spectroscopy provides a powerful tool for studying how electronic and vibrational states interact, but it cannot access the temporal evolution of the valence change density directly.

The prospect of correlation spectroscopy in the x-ray region suggests the possibility of a revolution in our understanding valence charge structure, correlation, and its evolution, particularly in complex molecular systems and correlated materials. Major advantages are element specificity, sub-femtosecond resolution, and unrestricted access to the valence states. X-ray correlation spectroscopy exploits core-level resonances (rather than homo-lumo transitions) with coherent attosecond pulses, and directly probes the dynamic correlation between valence charges associated with different atomic sites. These electron correlation effects are ubiquitous in chemistry; they are comparable in magnitude to chemical-bonding energies and

are thus crucial for predicting molecular geometries, reaction barriers, and reaction rates with chemical accuracy.

Figure 3.2 illustrates the concept of x-ray correlation spectroscopy applied to a simple molecule, aminophenol. The goal is to detect the correlation of electronic excitations associated with the oxygen atom with those associated with the nitrogen and to follow this coupling on the natural time scale of electron motion. Attosecond pulses at 400 eV and at 535 eV initiate $1s$ core excitations in N and O atoms, respectively, in a sequence of three x-ray pulses separated by times t_1 and t_2. This sequence generates a coherent four-wave mixing signal $S(t_3,t_2,t_1)$ that is measured in amplitude and phase via heterodyne detection. The two-dimensional Fourier transform of this signal with respect to t_1 and t_3 yields a two-dimensional electronic spectrum in frequency space. Off-diagonal features in this two-dimensional spectrum are present only when there is correlation between the two excited electrons on the N and O atoms. Calculations show that the extent of this correlation depends not only on molecular structure but also on the nature of the molecular orbitals excited within the energy envelopes (~10 eV) of the incident pulses.

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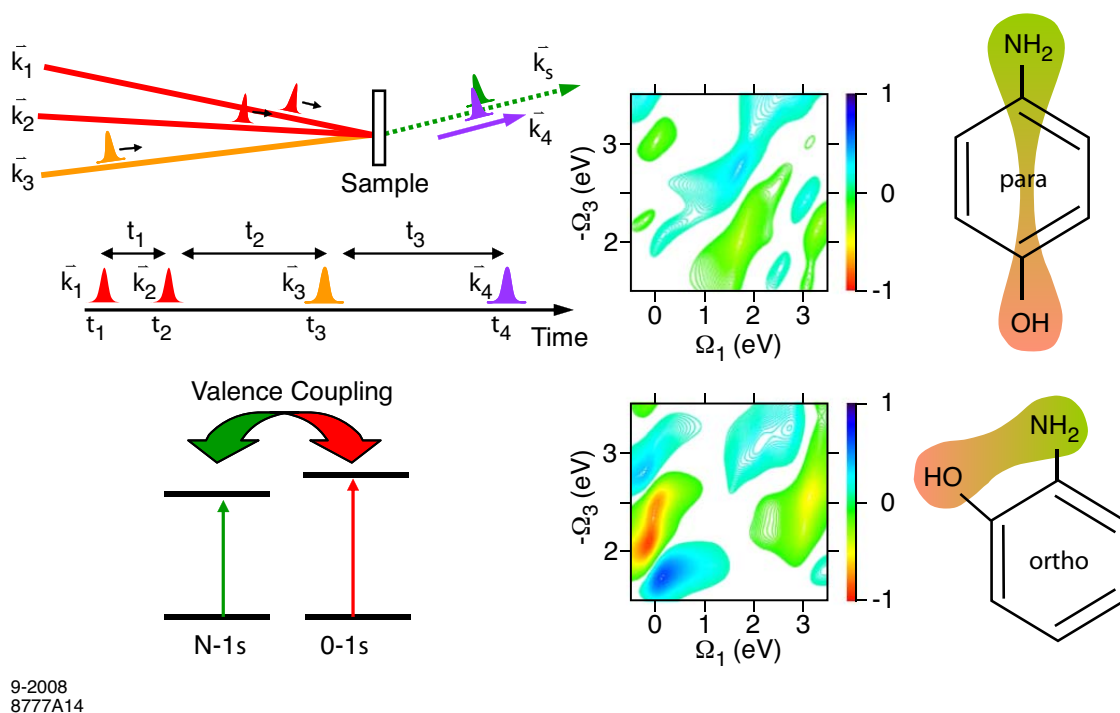
Figure 3.2. Left: Proposed four-wave mixing of ultrashort x-ray pulses resonant with the O-1s and N-1s levels; Middle: theoretically predicted two-dimensional spectra the lower of which exhibits the coupling of excitations on the oxygen with those of the nitrogen in para and ortho-aminophenol molecules at right. [Source: S. Mukamel]

4.   What Is Needed?

The experiments of the next two decades will make use of a variety of pump–probe configurations in which an ultrafast soft x-ray probe will interrogate a system by chemically selective ionization following excitation by a short pulse in the visible, UV, or XUV. New FEL facilities in the soft x-ray will have to provide exquisite synchronization with ultrafast laser sources providing short pulses from the XUV to the far IR region. Other experiments will require two-color x-ray pump–x-ray probe capabilities. Thus the synchronization of two different FEL pulses will also be an essential characteristic of these new sources.

Experiments in which electrons or ions are detected will demand high repetition rates, while experiments exploiting nonlinear x-ray optics will demand longitudinal coherence and control.

B. Chemical Reactivity: From Nature's Catalysts to Controlled Reactions

Understanding and controlling chemical reactivity at the scale of atoms and electrons will have a profound impact on many areas of human endeavor. Examples include sustainable energy production and storage, new materials, environmental and atmospheric science and remediation, and human health. Arguably the most important chemical reactions and those most difficult to understand and control occur in liquids or solids and, especially, at surfaces or interfaces between solids and liquids. These molecular reactions underlie essential processes in chemical or biological catalysis, solar energy harvesting by natural and artificial systems, energy storage and transport, and biochemical processes. Moreover, life itself is based on the chemistry in heterogeneous aqueous environments.

The main challenges for chemical reactivity in such systems are: (1) to understand the interplay between electronic structure (valence charge distributions, energy levels, bonding, spin) and atomic arrangement (bond distances and angles, coordination), and (2) to control the course of the reactions. Molecular transition states, which are intermediate between reactant and product species, are of particular significance in catalytic reactions. In this transient regime, subtle conformational changes and chemical-bond formation and dissolution ultimately determine the reactive pathways. Understanding and identifying the various reaction pathways and the energetics of the intermediates involved in the reactions is critical for improving catalysts. In order to arrive at this deeper understanding, we must be able to observe the reactions in real time and identify the transient chemical species on timescales ranging from milliseconds to femtoseconds. Both the kinetics and dynamics of a reaction step within the sequence are critical, and new x-ray sources are needed to achieve this goal.

1. Understanding Light-Induced Catalysis—Learning from Nature

Nature uses remarkably varied systems and mechanisms to perform catalytic reactions with extraordinary efficiency, speed, and complexity. At the active site of many enzymes is a metal center with just a few atoms that is responsible for the rearrangement of electrons and atoms needed to carry out the catalytic reaction. For example, the light-induced oxidation of water to dioxygen during photosynthesis by green plants and cyanobacteria is a process that occurs on a large scale in the biosphere and is essential for life on Earth. Using energy from sunlight, manganese centers in cyanobacteria and plants can split water and evolve O_2 at a rate of almost 1000 molecules/sec, three orders of magnitude faster than we can viably accomplish in the laboratory today. This sunlight-driven reaction couples the one-electron primary charge separation (photo-oxidation) with the four-electron water-oxidation chemistry. The electron extracted from water can be used for the reduction of H^+ to H_2 or for CO_2 reduction (see Figure 3.3). This natural catalytic scheme motivates design and synthesis of artificial photosynthetic systems based-on mixed-valence transition-metals bridged by oxygen and driven by visible light to produce carbon-neutral fuels from CO_2 and water. However, the development of economically feasible photo-catalysts requires a much better understanding of how these complex molecular systems function.

Much of the current progress in understanding the geometric and electronic structure of these systems on an atomic level is based on x-ray crystallography and spectroscopy studies (EXAFS, NEXAFS, XES) of

reactant-states or intermediate-states trapped at low temperature following photo-excitation. However, our understanding of both natural and artificial photosynthetic systems is significantly limited by our inability to probe the molecular structure (with atomic resolution) and to follow the valence charge distribution and oxidation states under ambient conditions and on their natural times scales, whose range covers femtoseconds (characteristic of energy transfer and charge separation), ~100 fs (characteristic of molecular vibrations, bond formation/dissolution), picosecond timescales (characteristic of molecular conformational changes and diffusive processes), and micro and milliseconds (reactions in complex biocatalysts). In addition to the need for time-resolved crystallography and x-ray spectroscopy studies, proposed multi-dimensional x-ray spectroscopy techniques may have a tremendous scientific impact by enabling us to follow both the coherent and incoherent flow of valence charge between different active metal sites in catalytic complexes. All of these studies are beyond the capability of present x-ray sources, and yet they promise to have profound consequences for understanding complex molecular systems.
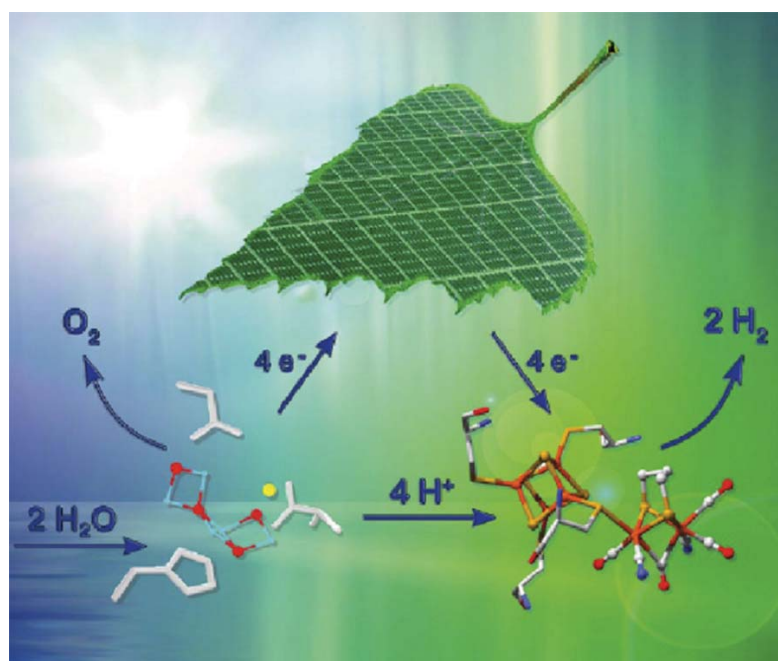
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Figure 3.3. Photosynthetic water splitting and a hydrogenase powered by a light-harvesting antenna complex. [Source: W. Lubitz, E.J. Reijerse and J. Messinger, *Energy Environ. Sci.* **1**, 15 (2008)]

2.   Real-Time Catalysis and Surface Chemistry

Heterogeneous catalysis provides a compelling example of both the scientific challenges and the impact of understanding and controlling chemical reactivity. A vast number of important processes rely on catalysis on surfaces, and their significance is evidenced by the 2007 Nobel Prize in Chemistry. Nevertheless, much needs to be learned about the intermediate steps of catalytic reactions at surfaces. In particular, we currently have very little knowledge of the dynamics of elementary processes on ultrafast timescales. Figure 3.4 illustrates the various steps of a typical surface reaction including adsorption, formation of different intermediates, and desorption. During each reaction step, there are rapid charge and energy transfers between the different adsorbates and the catalytic substrates that determine the dynamics and thereby the overall efficiency of the sequence. Examples of important catalytic reactions that involve

dissociation of the strong internal N_2, O_2, or CO bonds as a rate-limiting step include ammonia synthesis for fertilizer production, Fischer–Tropsch synthesis of fuels and oxygen reduction in fuel cells. The dissociation steps are strongly exothermic, and the dynamics of the energy dissipation between the final products and the substrate, where potentially large non-adiabatic processes can occur, is not understood.
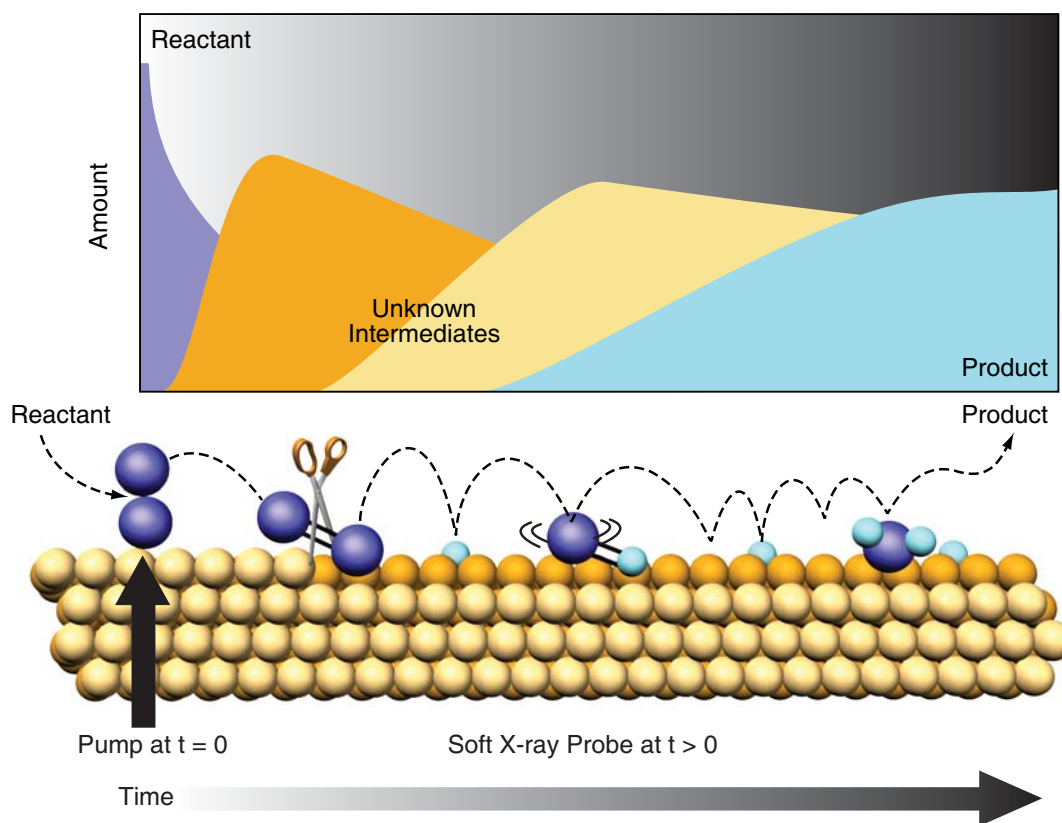
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Figure 3.4. Elementary steps in a catalytic reaction involving molecular oxygen and hydrogen. The reaction can be stimulated by a laser of THz pump pulse and probed with an FEL soft x-ray probe pulse.

In order to optimize catalysts it is essential to develop predictive experimental capabilities of the chemical dynamics. With new x-ray sources we have the potential to experimentally resolve the dynamics of elementary steps and the energy transfer with the substrate during a surface reaction. This can be achieved by coupling a controlled far-infrared or THz radiation pump with a time resolved x-ray spectroscopy probe (e.g., XES and XPS) that provides element specific information about the local electronic and atomic structure.

3.   Heterogeneous Chemistry of Aerosols—Geoengineering and Health

A deeper understanding of surface catalysis and condensed-phase reactivity will drive a significant advance in the field of aerosol chemistry where chemical reactions occur on the surface of nano-sized particles, or within liquid droplets dispersed in a gas. Such processes are responsible for cloud formation and rain and are a major factor in determining the solar irradiation of the earth, the integrity of the ozone layer, as well as health of humans, plants, and rivers. In order to understand the processes involved, it is

mandatory to chemically analyze the surface of these particles in their atmospheric carrier (lower atmosphere to stratosphere) while subjected to solar radiation and ozone. The most suitable tools for these objectives are x-ray-based spectroscopies (see Figure 3.5).
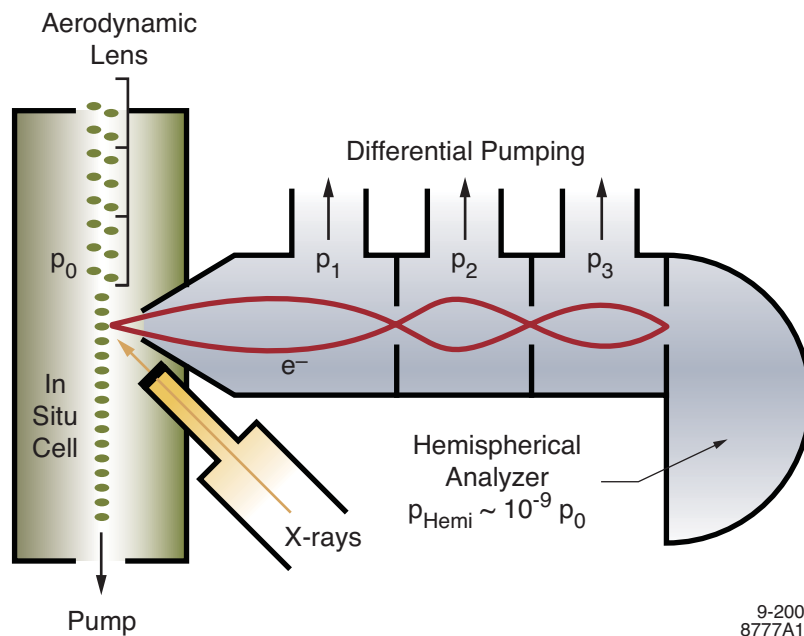
Figure 3.5. Schematics of x-ray spectroscopy setup for in situ aerosol studies. Carrier gas is kept at the appropriate pressure p_0 while the electron spectrometer is in ultrahigh vacuum.

However, with current x-ray sources such in situ studies are not possible with the relevant particles of ~100 nm diameter. X-ray sources with a much-increased average brightness are needed for chemical analysis of aerosol surfaces under realistic conditions of aerosol size and concentration (10^6 - 10^7 particles/cm^3). In addition, new soft x-ray sources with ultrashort pulses would make possible time-resolved studies in the femtosecond to picosecond range that could reveal the reactivity of particles in optically excited states.

The impact of such studies would be dramatic. We could use the knowledge for modeling the influence of anthropogenic aerosols on climate and health and for effective rain making and hail prevention through cloud seeding. Furthermore, the risks connected with the controlled injection of nano-sized aerosols into the stratosphere to counteract global warming could be minimized. Such injections are cost effective, but their implementation is presently too risky because of possible side effects, including the depletion of the ozone layer as well as reduced rainfall, soil moisture, and river flow.

4.   What Is Needed?

The experiments discussed in this section will make use of various pump–probe configurations. Work on photosynthesis requires visible light excitation and very bright x-ray probes ranging from femto- to millisecond time resolution. For spectroscopy experiments that require scanning the x-ray beam energy (e.g., NEXAFS and EXAFS) in microsecond studies a very stable high-average-brightness source is required. For spectroscopy studies based on wavelength-dispersive x-ray optics, either sources with

ultrashort x-ray pulses or high average brightness might be best suited, depending on the timescale of interest, sample and sensitivity to radiation damage. Multi-dimensional x-ray spectroscopy to follow both the coherent and incoherent flow of valence charge between different active metal sites will require two-color x-ray pump–x-ray probe capabilities in the soft x-ray regime.

In the surface catalysis studies, an ultrafast soft x-ray probe will follow the excitation by a short pulse in the THz, visible, or UV. The repetition rate will vary depending on whether systems are reversible or not and at which speed non-reversible systems can be renewed.

Work on aerosols requires very a high average brightness source for chemical analysis and pump pulses in the optical and near UV range combined with ultrashort soft x-ray probe pulses for reactivity studies.

C. Understanding Complex Materials

Central to all the DOE-BES grand challenges is the ability to determine and control electron and spin properties of matter with high precision in the energy, momentum, space, and time domains. "Materials," are a key form of matter that are the subject of core scientific and engineering disciplines and often drive progress in technology. More recently, materials in the form of nanostructures have become of increasing importance. In this section we discuss challenges in understanding the properties of "complex materials," which exhibit properties that cannot be explained by assuming independent electrons but arise from their correlations. Such materials come in various forms ranging from the pure elemental ferromagnets with strong exchange interaction between the electronic spins, to oxide-based "correlated materials" with coupling of charges and spins into nanoscale regions, to multiferroics with unique coupling of electric and magnetic order parameters, to glassy materials such as colloidal gels and glasses.

Today, a key challenge of condensed-matter physics lies in the understanding and control of materials that exhibit strong electronic correlations on the nanoscale. The intrinsic complexity of such "quantum matter" is illustrated in Figure 3.6. While bulk thermodynamic and transport measurements provide much information about these materials, truly deep insight on the level of atoms, electrons, and spins usually comes from scattering, spectroscopy, and imaging experiments. With the power of future light sources, we can for the first time perform combined scattering-spectroscopy-imaging experiments with sufficient resolution and sensitivity to study important emerging properties.
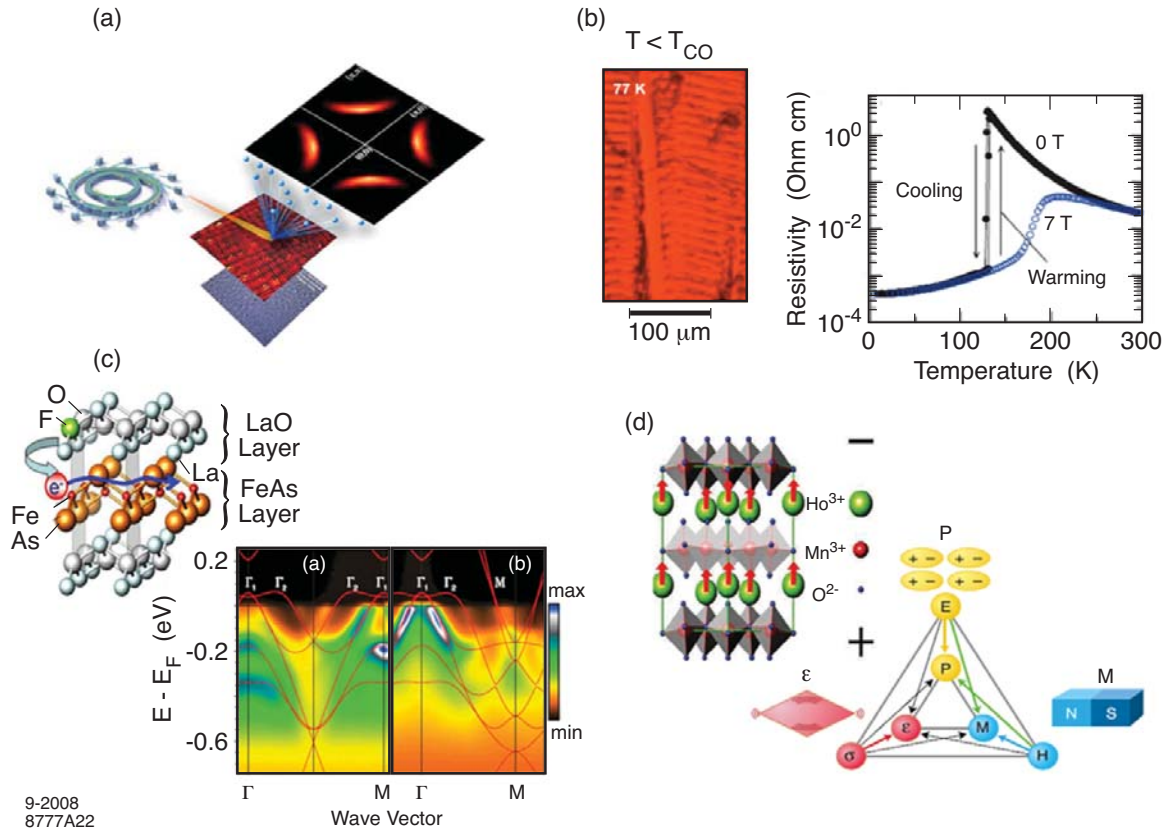
Figure 3.6. Examples of complex quantum materials: (a) High-T_c cuprates. (b) Manganites. [Source: Y. Tokura and N. Nagaosa, *Science* **288**, 462 (2000)] (c) New Fe-based superconductors. (d) Multiferroics. [Source: N. A. Spaldin and M. Fiebig, *Science* **309**, 391(2005)]

1.   Electronic Structure on the Nanoscale

The need for more complete information for complex materials may be readily illustrated for the case of complex materials in the form of transition metal oxides. Angle-resolved photoemission spectroscopy (ARPES) has emerged as the mainstream experimental probe for the electronic structure and low-lying excitations. Modern high-resolution ARPES provides unprecedented insight into the electronic structure of solids by measuring the single-particle spectral function in energy and momentum, $A(\mathbf{k}, \omega)$. The technique has had major impacts on a number of important topical fields, such as high temperature superconductivity, colossal magnetoresistive matter, and most recently graphene and topological insulators. However, these materials tend to exhibit pronounced spatial inhomogeneities on the nanoscale and ARPES today can only provide spatially averaged signals, resulting in the loss of critical information. ARPES with nanometer spatial resolution (nano-ARPES) and meV energy resolution overcomes this problem, providing information from individual ordered regions, thus combining information in space, momentum, and energy. Pioneering instrumentation is being developed at the ALS to measure the electronic properties down to a length scale of 50 nm, limited by the available photon flux density, to understand the electronic states within individual domains as illustrated in Figure 3.7.

(a)

Sample Electrons

Electron
Spectrometer

Zone Plate

Aperture

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Synchrotron
Photons

(b)

2 μm

Energy (eV)

Angle (degree)
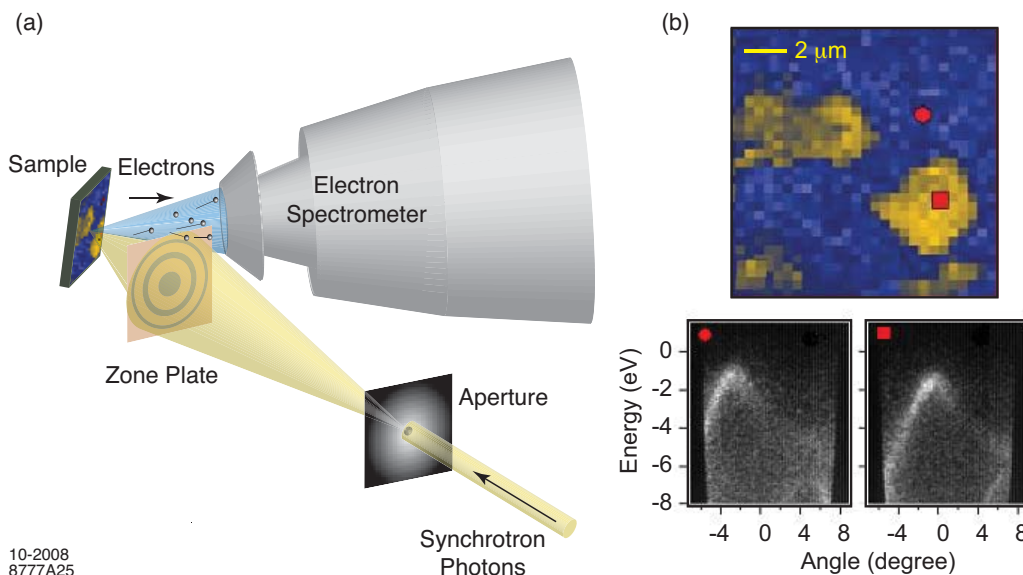
Figure 3.7. (a) Schematic of nano-ARPES set up. (b) Top: nano-ARPES images of randomly oriented crystalline grains of graphite. Bottom: bands acquired from individual crystalline grains. The shift in the bands in the angle direction corresponds to the orientation of each grain. (Conventional spatially integrating ARPES reveals only an average, smeared band).

Nano-ARPES experiments are best carried out on ring-based sources because space-charge issues require the reduction of the number of photons per pulse, and in rings this reduction may be compensated by high repetition rates. Another breakthrough that will be enabled by future rings is spin-resolved ARPES, which in the past has been impeded by the low efficiency (about 10^{-4}) of spin detectors. This technique is envisioned to become widely applicable with our ability on future sources to go to space charge limited photon flux per unit area. The last area where the new sources will make an impact would be time-resolved ARPES, which enables one to directly observe the time evolution of electronic states. Pioneering experiments using lasers indicate exciting promise, but new light sources are needed to fully exploit the potential of time-resolved ARPES for study of complex materials.

2.   Study of Emergent Phenomena in Correlated Materials with Soft X-Ray q-Resolved Inelastic Scattering

Often the complexity that lies at the heart of materials near several competing instabilities can be detected by studying a suite of spectroscopies measuring charge, spin, orbital, and lattice degrees of freedom. Critical information concerning the collective properties of solids has been obtained from inelastic neutron scattering, which directly probes the spin dynamics. Comparably direct information about the charge dynamics is only just beginning to be accessible through soft x-ray inelastic scattering (IXS) (see Figure 3.8a). Such information will clearly be essential to fully understand the interplay between charge order and superconductivity that occurs in the cuprates, and it will likely give key information concerning the role of various collective excitations in a wide array of complex and correlated materials. Additional features that distinguish IXS from presently available probes of electronic structure include: bulk sensitivity; suitability for probing insulating samples (organics, bio-materials etc.); compatibility with external fields (magnetic, electric), pressure, and optical excitation; and sensitivity to symmetry-forbidden optical transitions.

However, IXS experiments are very photon hungry, since the cross-section for IXS is many orders of magnitude smaller than it is, for example, for ARPES.
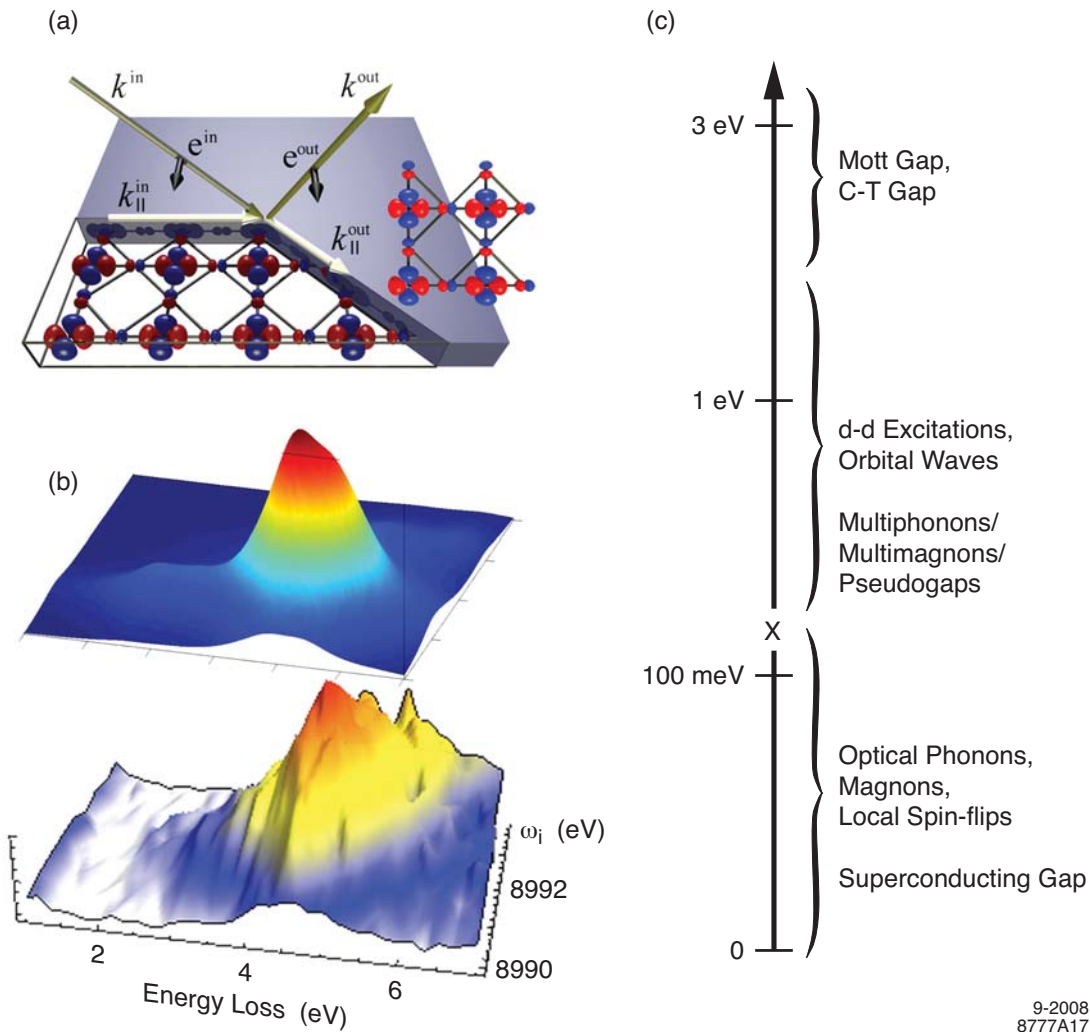
Figure 3.8. (a) Schematic of RIXS measurement with respect to underlying CuO_2 plaquettes. (b) Comparison of calculated (top) and measured (bottom) RIXS for q=(π,π) in La_2CuO_4. (c) Important energy range for collective charge excitations in correlated materials.

Although comparisons between theory and experiment for resonant IXS (RIXS) have been rather successful on the energy scale of a few eV (see Figure 3.8b), low throughput (low average photon flux) and low energy resolution of current light sources have precluded the observation of excitations at smaller energy scales relevant to the complex orders in oxides. For IXS studies of correlated materials, energy scales of important excitations include ~10 meV for a typical superconducting gap, ~20 meV for optical phonons or magnons, ~100 meV for pseudogaps, and 1 eV or greater for *d-d* excitations, charge-transfer gaps, or Mott gaps Figure 3.8c). The existence of novel charged excited states as well as collective modes in the range of 1–100 meV in high-temperature superconductors has been suggested by numerous theories. Such collective excitations are not directly observable in single-particle-excitation spectroscopies like ARPES. Inelastic scattering or time-resolved x-ray studies are thus vital for their detection. Next-generation light sources will provide many orders of magnitude more flux with higher resolving power that will help study of low-energy excitations with a high resolution that is not possible today.

IXS, as discussed above, is spontaneous Raman scattering (SRS) in the x-ray regime. A powerful extension of SRS, *coherent* Raman scattering or stimulated Raman scattering, which is formally similar to RIXS, is a four-wave mixing process whereby three incident fields $E_n(k_n, \omega_n)|_{n=1,2,3}$, generate a stimulated signal, e.g., $E_{sig}((-\omega_1 + \omega_2 + \omega_3)$, in the momentum-matched direction, $k_{sig} = -k_1 + k_2 + k_3$ (see for example, Section 3.1.c). This kind of spectroscopy allows one to detect the correlation between two processes in a material system—a new dimension to understand the correlated materials. However, the estimated cross-sections of four-wave mixing experiments with x rays are estimated to be quite low, so that these classes of experiments may be highly challenging even with next-generation of FEL. Therefore, we expect to start with a relatively simpler version by pumping a system of correlated complex materials with terahertz or far-IR radiation for selective excitation of some modes (phonon excitation, magnetic excitation or breaking pairing of electrons in superconductors) and probing the system with IXS as a function of delay time. This will enable us to dissect different aspects of correlation one after the other in a very controlled and selective way and thus will allow understanding and eventually controlling emergent phenomena in complex materials, which is one of the BES grand challenges.

The intertwining of charge, spin, orbital, and lattice degrees of freedom in complex materials leads to non-trivial spatial and temporal evolution of charge dynamics that govern the way in which the individual electrons may untangle and respond when driven by external fields. This lack of knowledge of the quantum many-body state is one of the major unanswered issues in all of physics, having an impact on both fundamental and applied questions of physics. Local information concerning the projected electron excited-state wave functions (i.e., element- and chemical-specific information) can ultimately be obtained from the dependence of the scattering cross-section on the energy, momentum, and polarization of the incoming and outgoing photons. By manipulating incident and scattered light polarization, energy, and momentum, charge dynamics from different orbitals and different regions of the Brillouin zone may be individually selected and probed. Recent theoretical development shows that even with limited resolution, RIXS may "peel the skin off the onion" to reveal the correlated nature of quantum many-body wave functions. The advance came from the realization that, while the normal scattering cross-section is fully determined by the momentum transfer **q** and is independent of initial ($\mathbf{q}_i$) and final ($\mathbf{q}_f$) states, the resonance cross-section is modulated by the overlap between initial and final states with the complete set of intermediate states. By exploiting this modulation, one gains information of the wave functions.

3. Nanoscale Dynamics: Natural or Equilibrium Times

The study of the dynamical properties of nanoscale order in complex materials also provides a severe challenge. The natural time scales of the physical processes in such systems span an enormous range of about fifteen orders of magnitude, depending on the atomic, electronic, and spin structure and parameters such as temperature (see Figure 2.1). Observation of materials with nanoscale resolution over this broad time scale can only be effectively covered by a combination of ring-based sources, covering the range from seconds to tens of picoseconds, and linac-based FELs for shorter times.

The natural time scale associated with nanoscale order, which is associated with non-repetitive processes such as equilibrium fluctuations, can be directly studied by x-ray scattering techniques, either in the energy domain by IXS or the time domain by hard x-ray small-angle scattering. In both cases the dynamic information represents statistical averages as a function of momentum transfer or length scales in the sample. More recently, with sufficiently bright x-ray sources, conventional time dependent small-angle scattering has been extended to the use of soft x rays that are coherent (coherent power scales as wavelength squared) across the entire sample region (typically several microns) of interest. Any temporal

change within the sample region is directly reflected by an intensity modification within the coherent scattering (speckle) pattern. Often it is not even necessary to invert the speckle pattern into real space but simply record its time dependence as illustrated in Figure 3.9.
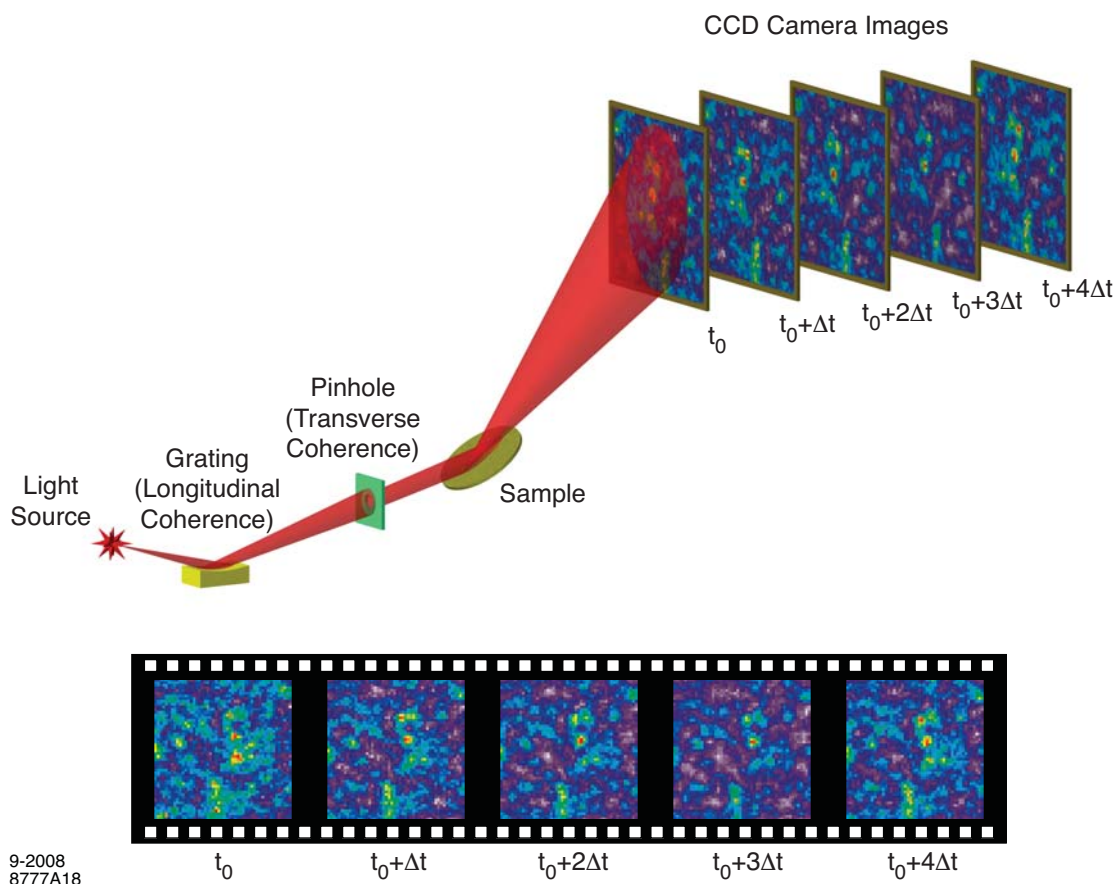
Figure 3.9. Movie of the "speckle" pattern (frames were taken 240 seconds apart and each frame averaged 96 seconds) from the orbitally ordered domains in the cubic manganite $Pr_{0.5}Ca_{0.5}MnO_3$, recorded at the ALS. The slow dynamics of the image indicates that the domain walls are pinned by disorder, but the nature of the pinning is unknown.

The results suggest that physics on the large "mesoscopic" length scale is intimately related to atomic and/or electronic disorder at the Å scale. Naturally, one would like to extend such experiments, presently limited to time scales of many seconds, to much shorter time-scales. The signal to noise ratio of such experiments scales as $\tau^{1/2}B\sigma$ with the sampling time τ, the source brightness B, and the scattering cross-section per volume σ and thus greatly benefits from the increase in average brightness available on future rings.

As IXS in the energy domain yields information on local collective charge excitations, studies in the time domain offer complementary insight into how interactions on the local level manifest into the formation and dynamics of nanoscale domains. This is one of the promising pathways for using x rays to open windows into the phenomenon of emergence, linking cooperative charge dynamics from both local and nanoscale perspectives.

Faster dynamics require the use of x-ray FELs (X-FELs) and fast 2D detectors with time correlators, so that the speckle patterns recorded with individual ultrafast pulses may be compared. Such comparison

depends on the readout speed of position-sensitive detectors, which is presently inadequate and needs to be greatly improved. In the near future, this limitation can be overcome by comparing images recorded with one shot with those recorded with multiple shots, so that temporal changes are reflected by increased blurring with the number of shots. With suitable delay lines it will be possible to study nanoscale dynamics down to the femtosecond time scale.

In principle, real-time motion pictures of nanoscale equilibrium dynamics are also possible by x-ray microscopy, e.g., PEEM, but they are presently also limited to timescales of seconds by the available average x-ray flux density on storage rings. Ultrafast x-ray movies of real-space fluctuations could be produced with adequately spaced X-FEL pulses, each sufficient to record a picture. This, however, requires the development of ultrafast position sensitive detectors.

4.   Nanoscale Dynamics: Operational Times

In Section 2 we have discussed the technological bottleneck that exists around 100 ps. An example is the speed limit of writing and reading electronic "bits" in data storage. Since technology depends on repeatable processes their exploration naturally lends itself to pump–probe techniques. Since optical pulses can be ultrafast, one of the forefront questions in magnetism research today concerns the possibility of manipulating the magnetization optically. Figure 3.10a shows that this is indeed possible. The laser pump–probe experiment shown reveals the ultrafast demagnetization of a ferromagnet by an intense (> 1 mJ/cm^2) laser "pump" pulse. Figure 3.10b illustrates that the use of circularly polarized light may indeed supply the angular momentum to enable "all-optical switching" on the femtosecond time scale.

While we have now observed such "effects," the underlying processes remain the subject of great controversy. It is clear that the sample evolves through a laser-excited electronic state where the modifications of the magnetization take place. Simply replacing the femtosecond laser probe pulse with ultrashort probe pulses supplied by a soft x-ray FEL would allow the understanding of this transition state. Time-dependent XMCD measurements would reveal the separate evolution of the spin and orbital components of the magnetization, providing key information on the transfer of angular momentum out of the spin system that has to accompany any magnetization change. Time and spin resolved photoemission spectra would directly measure changes in the spin resolved band structure in the laser-excited state. As illustrated in Figure 3.10c, we do not know at present whether and how the electronic bands, the exchange splitting, and the spin-orbit coupling change in the laser-excited state.

5.   What Is Needed?

From the above discussion, it is apparent that complementary access to the nanoscale is provided by both soft and hard x rays, and therefore bright x rays over the spectral range from tens of eV to tens of keV are needed.

In some cases, we clearly need the ultrafast capabilities of FEL sources. Often the availability of ultrashort pulses at high repetition rates on the order 100 kHz is necessary to obtain sufficient signal. We envision that for certain experiments we may need to reduce the number of photons per pulse to avoid sample "damage" or space-charge effects. An example is the ultrafast spin polarized photoemission experiment discussed in conjunction with Figure 3.10.
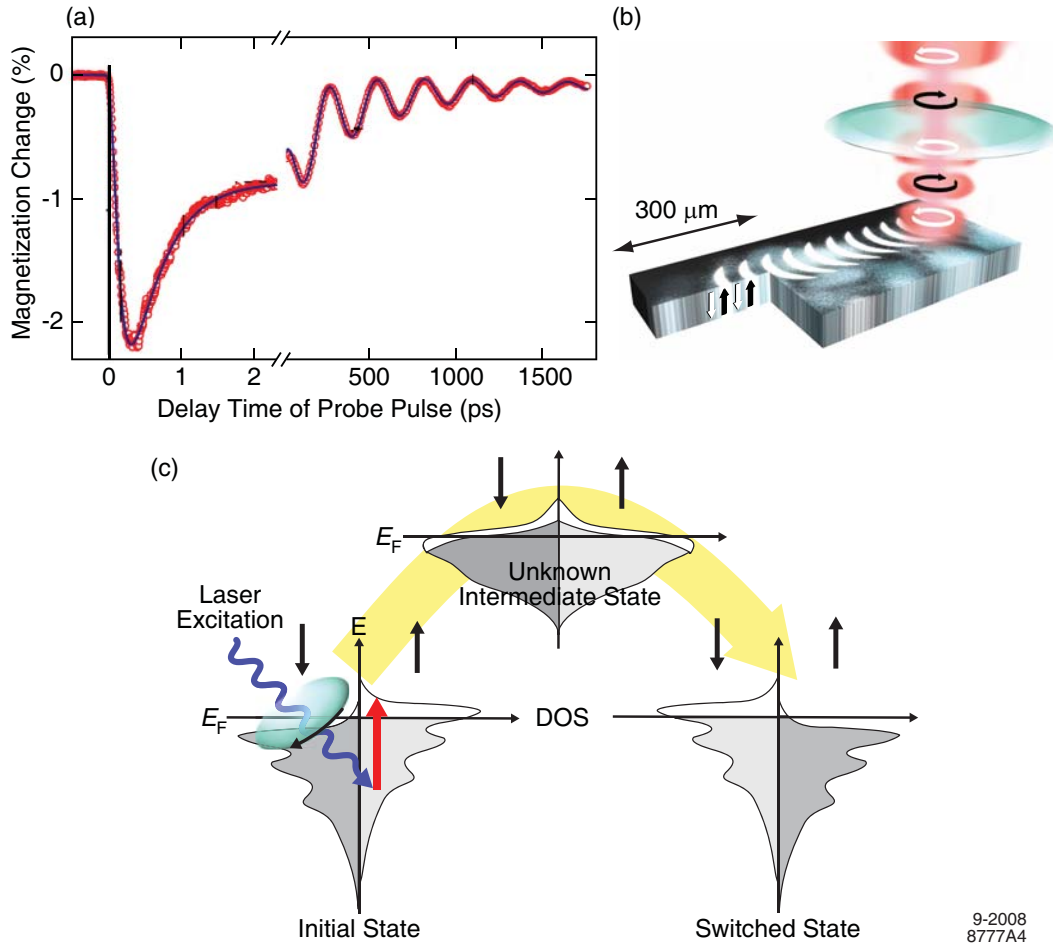
Figure 3.10. (a) Reduction of the perpendicular component of the magnetization *M* after laser heating a Ni thin film at *t*=0. The sub-picoseconds quenching of *M* is followed by a recovery via electron–phonon relaxation, and at *t* > 100 ps by a damped precession of *M* about the applied field. [Source: B. Koopmans, J.J.M. Ruigrok, F.D. Longa, and W.J.M. de Jonge, *Phys. Rev. Lett.* **95,** 267207 (2005)] (b) Demonstration of compact all-optical recording of magnetic bits in $Gd_{22}Fe_{74.6}Co_{3.4}$ by scanning a circularly polarized laser beam across the sample and modulating the polarization of the beam between left and right circular. [Source: C.D. Stanciu *et al. Phys. Rev. Lett.* **99,** 047601 (2007)] (c) The evolution of the spin-dependent electronic structure through excited states after laser excitation remains an unsolved science problem.

We envision that even in the future the largest number of x-ray users studying "materials" will benefit from high average brightness provided by CW-like storage rings that also offer superb beam stability in position and time, which is required for optimum signal to noise and background ratios. In practice, the key parameter is often photon flux (number of photons/time/area), not brightness, since techniques such as x-ray absorption, x-ray emission and photoemission are insensitive to momentum spread (angular divergence) of the incident photons and also do not require lateral coherence. In some cases, such as nano-ARPES, high repetition rates are needed to avoid space charge effects while still achieving the desired resolution and signal to noise ratio.

The complicated problem of the interaction of increasingly intense x-ray pulses with the sample, for both FELs and future rings, will be addressed in more detail in Appendix B.

D. Novel Materials Design and Characterization under Extreme Conditions

Synthesizing novel materials in extreme environments (pressure, temperature, electromagnetic fields, magnetic flux compression, radiation) may seem like an exotic approach, but nature already provides us with an excellent example, diamond, whose stability field is at high pressure and temperature (*P-T*), but whose metastability field extends back to ambient conditions. Understanding the high-pressure phase of carbon led to the development of synthesis techniques that mimic its creation (high *P-T* methods) and that now drive the billion-dollar diamond-abrasives industry. In addition, knowledge of the sp^3 carbon bonding of diamond has led to a new synthesis pathway that uses sp^3 bonded carbon in a methane plasma as a starting material for chemical-vapor-deposition growth of diamond in a near-vacuum environment; high pressure is no longer required. This synthesis technique shows promise for growing large single-crystal diamonds for high-tech applications.

The general objective is to interrogate, direct, and manipulate bulk and nanoscale material properties at the level of atoms or electrons in order to discover new synthesis pathways, create new forms of tailored matter, and visualize the fundamental processes underlying materials at their limits. Only a few tools exist to carry out this task. Intense ultrafast electromagnetic fields (from terahertz to x rays) can be used to induce exotic transient phases of matter that may subsequently be quenched into technologically useful metastable states. Magnetic-flux compression creates magnetic fields as large as 1000 T for a duration of milliseconds, matching naturally occurring exchange fields. Diamond-anvil cells create huge static pressures on materials up to several hundred GPa, as present, for example, in the center of the earth.

Today, x rays are the most powerful tools to characterize materials in extreme conditions, but new sources are needed to push this field to maturity. For ultrashort intense excitations, powerful ultrashort probe pulses from both soft and hard x rays are required. For millisecond excitations, both hard and soft x rays with high average brightness are needed. Studies of samples inside diamond anvil cells require the penetration of the probe into the pressure cell combined with its sensitivity to the local structure and dynamics. X-ray diffraction and inelastic hard x-ray scattering from electronic and collective excitations are such probes.

1. Static High Pressure

Extreme pressures and temperatures are a still largely unexplored in the search for phases with new properties and the study of the interiors of large planets. They can also point the way to the development of unique new materials that can be synthesized and used at practical conditions. Of critical importance for tapping this potential is the ability to characterize the samples and understand their behavior at extreme conditions. X-ray Raman spectroscopy (XRS, see Figure 3.11) is a powerful probe for studying bonding changes in elements and compounds.
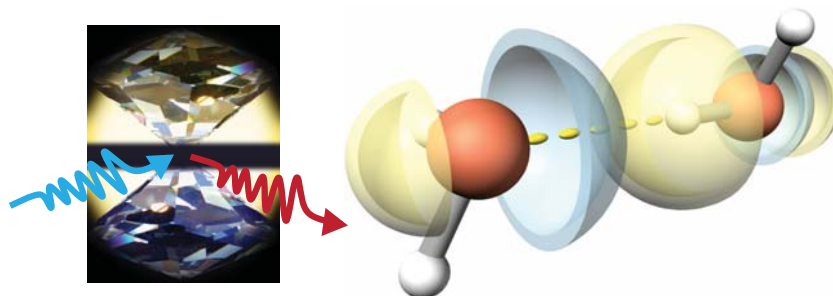
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Figure 3.11. In x-ray Raman spectroscopy, hard x rays are inelastically scattered from a sample inside diamond cell (left). The energy loss excites a core electron to probe empty valence orbitals (right) that are sensitive to the local structure.

This technique has been successfully applied at moderate pressures, but these studies are severely limited with existing x-ray sources, and systematic studies at ultrahigh pressures (> 100 GPa) are currently not feasible. A much brighter x-ray source would overcome the present trade-offs and limitations of sample size, data-acquisition time and accuracy, as well as spatial resolution. It makes possible accurate studies at ultrahigh pressures, simultaneous high temperatures, and temporal resolution that are crucial for next-generation explorations. For example, we would like to study the novel photochemistry and behavior in systems relevant for production and storage of energy. Very recently it has been found that, at moderate pressure, H_2O decomposes in the presence of an intense x-ray beam to form a new molecular H_2–O_2 compound (Figure 3.12).
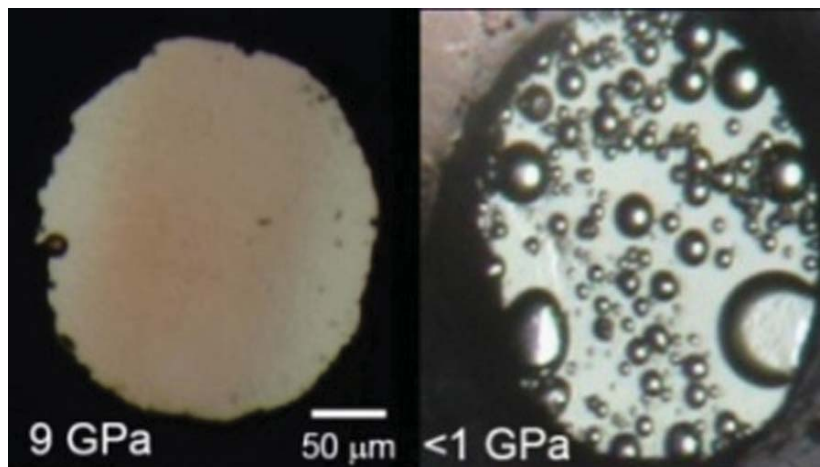
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Figure 3.12. Compressed H_2O sample after x-ray exposure. Left: New H_2–O_2 compound has formed (see brown region). Right: after decompression, H_2 and O_2 gases are released.

This reaction has implications for understanding how to efficiently split H_2O and store H_2 in a crystalline material. There are many basic questions that need to be addressed in order to understand the reaction process and the new materials that are created. What intermediate species are formed during the reaction? What energy levels are being excited? What metastable states can be created? Are similar phenomena at play in other basic hydrogen-rich molecules like NH_3 and CH_4? Currently, such studies are impractical for

following the reaction progress because of the lack of brightness from existing sources. With a much more intense source, we will be led to new synthesis pathways for generating and storage of high-energy-density materials and for understanding fundamental physics and chemistry in simple hydrogen-rich systems.

We would be able to address the synthesis of metallic hydrogen by directly probing the band gap and the plasmon when H_2 metallizes. We could test theoretical predictions for two coexisting superconducting superfluids (electronic and protonic) at the minimum melting point and liquid ground state of hydrogen. Moreover, we could unravel the high *P-T* behavior of H_2O including the superionic phase, symmetrization of H-bonding, and liquid–liquid transitions through XRS at the oxygen K edge. Using the superior spatial resolution, we would be able to carry out XRS studies of chemical bonding of carbon at pressures exceeding 100 GPa. Results may reveal the fundamental changes of carbon at high pressures and novel discoveries in inorganic carbon (e.g., CO_2), organic carbon, and elemental carbon (e.g., fullerene, graphene, and amorphous carbon).

2.   Polycrystalline Materials Science

While static high pressure is commonly achieved in pressure cells, transient high pressures can be produced by a variety of tools, e.g., laser, pulsed power, and particle beams. The lattice-level response of polycrystalline matter to high pressures is considerably more complicated than that of mono-crystalline matter, yet of eminent practical concern. It is well known that uniaxial strain can be made to occur at the macroscopic level in many experiments, but it is not clear what actually occurs on the length-scale of a single grain. Indeed, even under identical elastic compression, or mild plastic compression (where few dislocations are generated and/or moved) one might expect differing responses from individual grains depending on their orientation. Our understanding of the pathway and time-scales in which individual grains react in such circumstances is in its infancy. Ultimately, one wishes to understand how individual grains respond elastically and plastically to compression as a function of their orientation with respect to a given overall uniaxial pressure wave, and the degree to which stress and/or strain is continuous across grain boundaries, i.e., in the terms of material science, the degree to which the Voigt (where strain is uniform) or Reuss (where stress is uniform) conditions hold. The interrogation of crystals via in situ x-ray diffraction under shock and/or static pressure at the granular level will lead to an understanding of the stability of materials and improve hydrodynamics simulation codes to model large-scale phenomena.

Clearly an x-ray FEL with high peak brightness and multiple beams that can be either delayed with respect to each other and/or used to synchronously probe different orientations can address many fundamental questions. The narrow divergence and bandwidth allow for diffraction from polycrystalline foils that have been simultaneously shocked with a synchronized optical laser, with high peak brightness sufficient to ensure that the response of individual grains of varying orientation with respect to the shock propagation direction can be interrogated. The micro-radian scale divergence of the FEL beam will be far less than the rocking curve width, i.e., acceptance angle, of an individual grain. Ignoring defects, the typical acceptance angle of a grain is on the order of the number of unit cells across the grain.

3.   Intense Electromagnetic Fields

Electromagnetic excitations may be broadly classified by their field strengths or intensity (field strength squared) and time duration and/or frequency. For materials, a useful reference is provided by the bonding "field" experienced by valence electrons, which is of the order of 1 V/atom, corresponding to a field strength of $\sim 10^{10}$ V/m. Characteristic time scales in solids are set by nuclear motions (> 100 fs) and

valence electron motions (~ 1 fs). With our present and future capabilities to create electric fields that exceed the bonding fields and span a large frequency range from terahertz or picosecond fields to x-ray or attosecond fields we are entering new regimes of experiment and theory that go well beyond our present knowledge. This will require the development of new theoretical approaches that include dynamic field effects.

Femtosecond optical excitation of materials coupled with new sources that provide atomic-scale resolution probes opens up new possibilities for elucidating the fundamental structural and dynamical properties underlying nanoscale materials at extreme conditions, as illustrated in Figure 3.13.

Atomic-scale rearrangements and synthesis of new structural forms may be enabled by the ability to characterize and capture snapshots of the transient states induced by high temperature and/or high pressure. It is largely unknown how phase diagrams at the nanoscale are modified. Techniques are required for traversing and accessing new regions of the phase diagram that are impossible to reach with present steady-state methods. This may be achieved through direct optical excitation of materials as well as through the generation of shock waves that rapidly traverse the material to be studied, both requiring transient techniques to characterize the structural evolution. Such studies are also critical for discovering new pathways for synthesis of materials of enhanced stability and strength.
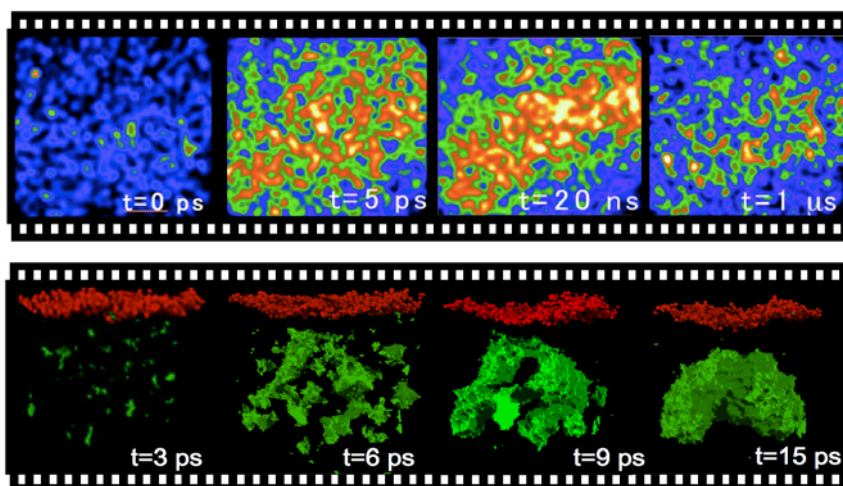
Figure 3.13. Top: Raw x-ray diffuse scattering measurements capturing the nucleation of nanoscale voids in optically excited semiconductors in real time. Bottom: molecular dynamics (MD) simulations showing the formation of nanoscale voids beneath the surface, supported by the experimental data.

The interaction of with materials with electromagnetic fields in the optical frequency range is accompanied by heating, melting or even plasma formation caused by photoexcited interband transitions. On the other hand, far IR or terahertz radiation of meV energy leads only to intra-band excitations. We are only beginning to explore the effects of high power THz fields on matter, as it has only recently become possible to produce ultrastrong half-cycle pulses of THz radiation with electric-field strengths rivaling bonding fields in materials (~ V/atom) and magnetic fields exceeding typical laboratory fields (> 10 Tesla). First experiments conducted by exposing ferromagnetic thin films to such fields have revealed remarkable new physics that is presently ill understood. The ultrastrong magnetic fields are found to lead to a nonlinear magnetic response that fractures the magnetization and imposes a speed limit on the

technologically important magnetic-switching process. The electric fields are capable of distorting the atomic valence charge on the level of ligand field-like anisotropies and lead to a transient novel magneto-electronic anisotropy, possibly opening the door to manipulating the magnetization by electric fields alone. Most surprising is the absence of Joule heating in the sample. This indicates that extreme electric fields can temporarily transform a transition metal into a new electronic state, indicating physics beyond present models. These experiments are just the first experimental evidence for unusual behavior of matter in extreme THz fields. An entire field is open for exploration and, in particular, the combination of THz pump and x-ray probe experiments will open the exciting opportunity to separately study the atomic, electronic and magnetic response of materials to extreme electromagnetic fields.

4.   What Is Needed?

Inelastic x-ray scattering (both, from electronic and collective excitations) and diffraction on systems under extreme static pressures will require x rays with the highest possible average brightness at photon energies ranging from 7–20 keV. Work on systems in intense electromagnetic fields will require synchronized ultrashort x-ray pulses in the soft and hard x-ray regime.

E. Life Science and Soft Condensed Matter

Biological imaging covers many orders of magnitude in scale, from atoms, molecules, macromolecular assemblies, cells, to entire organisms. Different physical methods are used to cover these different sizes: optical microscopy, x-ray microscopy and tomography, cryo-electron microscopy, and x-ray diffraction. Among these techniques, x-ray diffraction plays a pivotal role at the understanding of biological systems at a molecular level by revealing structures of proteins, nucleic acids, and their complexes, ranging from small enzymes to very large assemblies such as the ribosome. Increasingly though, as structural biologists are attempting to study ever more challenging and complex systems, x-ray crystallography is hampered by obtaining crystals suitable for conventional diffraction data collection at third-generation light sources, and new approaches will be needed. A way forward is to extend conventional crystallography to allow studies on very small crystals.

1.   Atomic-Resolution Crystallography

It has long been suggested that the use of very bright x-ray micro-beams (10–1 μm or smaller in diameter) would offer potential advantages for discovery science in the study of very small crystalline materials. For example, such micron-sized beams allow selection of small regions in the crystal, thus enabling one to select a small sub-volume of the crystal that is relatively well ordered and at the same time to increase the signal-to-noise ratio of the observed intensities by strongly reducing background scattering. Critics of this approach have argued that the radiation damage that occurs in a small crystal volume would prevent the collection of a full diffraction data set. Theoretical estimates suggested that the required crystal volume has to be at least 8000 μm^3 for unit cell dimensions around 100 Å. However, recent experiments at the ESRF and at the Swiss Light Source have demonstrated that crystals with a volume of 20 μm^3 are entirely sufficient to obtain a full diffraction data set of high quality to high resolution using a beam with 1-μm diameter. Thus, it appears that the limit of the smallest possible crystal volume usable for diffraction studies has not yet been reached. Furthermore, even if severe radiation damage were to occur, it is possible to collect complete datasets by irradiating different regions of the crystal or to collect data from a large set of different crystals of the same material, as has long been the practice in virus crystallography.

At the same time, techniques to obtain micron and submicron crystals are undergoing a dramatic development. Microfluidic devices are undergoing rapid development to perform crystal screens with much reduced sample requirements. It is often possible to obtain conditions that produce small (submicron) crystalline "entities" too small to be studied with current techniques but accessible to the bright micron or submicron x-ray beams of a new generation source. Another approach is the use of biological or chemical scaffolds that promote the formation of micro- to nano-sized crystals consisting of engineered lattices. A particularly interesting example is the use of a virus system that forms micron-sized protein crystals inside infected cells. These crystals form a highly ordered molecular scaffold for a protein that is to be studied. Crystal sizes range from a few microns to as small as about 200 nm and can be obtained as homogeneous populations and in large quantities. Thus, the need arises for development of x-ray sources that produce extremely bright, coherent beams with diameters in the submicron range with a brightness comparable to or exceeding today's most powerful storage-ring-based light sources.

Clearly, conventional methods of crystal mounting cannot be used for submicron crystals. One approach is to scan the entire sample and then to piece together a complete diffraction data set from crystal "hits" within the sample. Ultra-fast data collection is now possible with pixel array detectors (PADs). Crystal "mounting" may also take novel forms, such as using laser tweezers, simply passing the object through the beam using a capillary injector, or mounting samples on micro-fabricated grids that are then translated in the beam. Data collection is most efficient if the object's position in the beam can be timed to coincide with the x-ray pulse. For reproducible, mass-selected samples this arrangement may take the form of "serial crystallography," proposed by Spence and colleagues. "Orienting" may be by use of lasers or other external fields, or by detection of the directionality of molecular fragments. Phasing could be accomplished by iterative algorithms developed for diffraction microscopy (see below).

2.   Nanoscale Structure and Dynamics

Development and successful deployment of new paradigms for routine "submicron" crystallography would have a wide-ranging and broad impact. Achieving an understanding of the structure of large macromolecules and their complexes including membrane-bound proteins, how the interactions between macromolecules in these larger assemblies confer and control function (i.e., the workings of "molecular machines"), and ultimately how these assemblies are organized and networked at the cell and tissue level will lead to a knowledge base that will provide for a predictive understanding of complex biological systems from the sub-nanometer scale up through the cellular level. Such information would transform medicine, renewable energy research, environmental remediation, and other fields that are informed by or harness biological systems.

Conformational changes in macromolecules and their complexes are of enormous interest, as scientists try to understand protein folding, the functioning of enzymes, molecular machines, transcription, etc. Small- and wide-angle x-ray scattering have provided much information in this area but have been limited in time resolution to the millisecond and slower range in most cases. By using nanofluidic mixers, fast area detectors, as well as pump–probe techniques, high-brightness x-ray nanobeams will extend the time resolution well into the critical microsecond range and perhaps below. At this time scale, molecular dynamics calculations begin to overlap the observations, leading to a much more accurate description of the processes involved. This technique promises to revolutionize the study of the dynamics in soft matter in general, in ways that should have both fundamental (phase transitions and self-assemblies) and technologically important (mixing) applications. Given a highly parallel micron-size beam of hard x rays,

the steps in macromolecular folding can be followed on a microsecond time scale at nanometer resolution using a nanofluidic mixer and temperature-jump- or photo-activation, as illustrated in Figure 3.14.
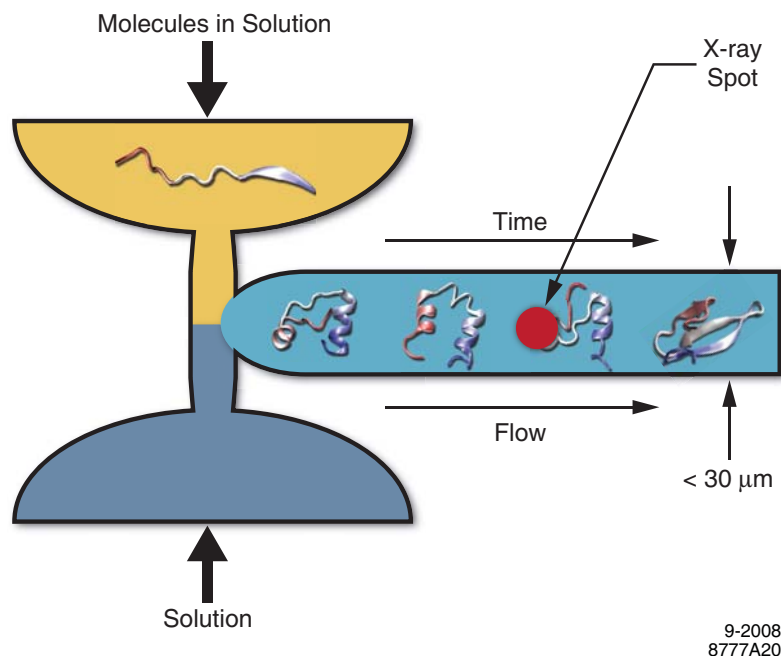
Figure 3.14. Schematics of a time-resolved x-ray scattering experiment using a nano-fluidic continuous-flow mixer [Matsumoto *et al., JACS* **129**, 3840 (2007)]. An unfolded-protein solution (top) is mixed with a second solution (bottom) that promotes folding. The x-ray beam interacts with the solution jet at different distances from the mixing point, corresponding to different time points in the protein folding process [The folded protein models adopted from an MD simulation by Freddolino *et al.,. Biophys. J.* **94**, L75 (2008)].

There is growing evidence that conformational fluctuations in proteins and macromolecular complexes play a fundamental role in target recognition and functional switching. Dynamical structural techniques to supplement "static" atomic resolution structures are thus crucial in all aspects of structural molecular biology and in gaining an understanding of the relationship between structure and function. X-ray photon correlation spectroscopy is one such technique that has been highly effective in studying slow non-equilibrium phenomena in soft condensed matter. It requires coherent beams, and is therefore limited in terms of the parameter space it can reach today and in the mid-term future. It will clearly benefit from the coherent beams available at future enhanced x-ray sources, on the microsecond time scale from storage rings and at faster time scales from FELs, for the studies of biologically relevant dynamics as well as soft-condensed-matter dynamics in the microsecond to millisecond time scale.

How are macromolecular complexes organized into organelles and organelles organized within the cell? Much of what we know in this area comes from electron microscopy. While it provides superb resolution, it is limited to very thin samples. In contrast, visible-light microscopes are limited in resolution. X-ray microscopy and, especially, cryo-x-ray tomography has been developing rapidly with the goal to fill the gap between electron and visible-light microscopies. This very powerful new technique is providing exciting results at the 30–50 nm resolution level in intact cells, limited by available x-ray optics. To go beyond this limitation, diffraction microscopy and ptychography (a technique for solving the phase problem in diffraction patterns by interfering adjacent Bragg reflections coherently and thus determining

their relative phase) are being developed at several laboratories. These lensless forms of 3D x-ray microscopy require intense, coherent x-ray beams.

Coherent beams from FELs will revolutionize these approaches. Illumination with an ultrashort and extremely bright x-ray pulse can outrun key damage processes over a very short period of time. This can be exploited to extend the diffraction signal to the highest possible resolution in flash diffraction experiments where we expect to achieve resolutions well beyond those limited by conventional damage barriers (Figure 3.15). A detailed analysis of imaging live cells with x-ray lasers suggests that nanometer to sub-nanometer resolutions could be achieved with intense, ultrashort x-ray pulses at wavelengths of about 1.5–0.5 nm. This is substantially higher than resolutions possible in any other type of biological imaging experiments on non-reproducible objects. A high-resolution 3D data set from a non-reproducible object would require simultaneous flash illumination by many pulses from many directions. This could, in principle, assure that significant damage does not develop during data collection. Orthogonal illumination could be realized with split-beam geometry with soft x-ray pulses for which efficient multilayer mirrors exist.
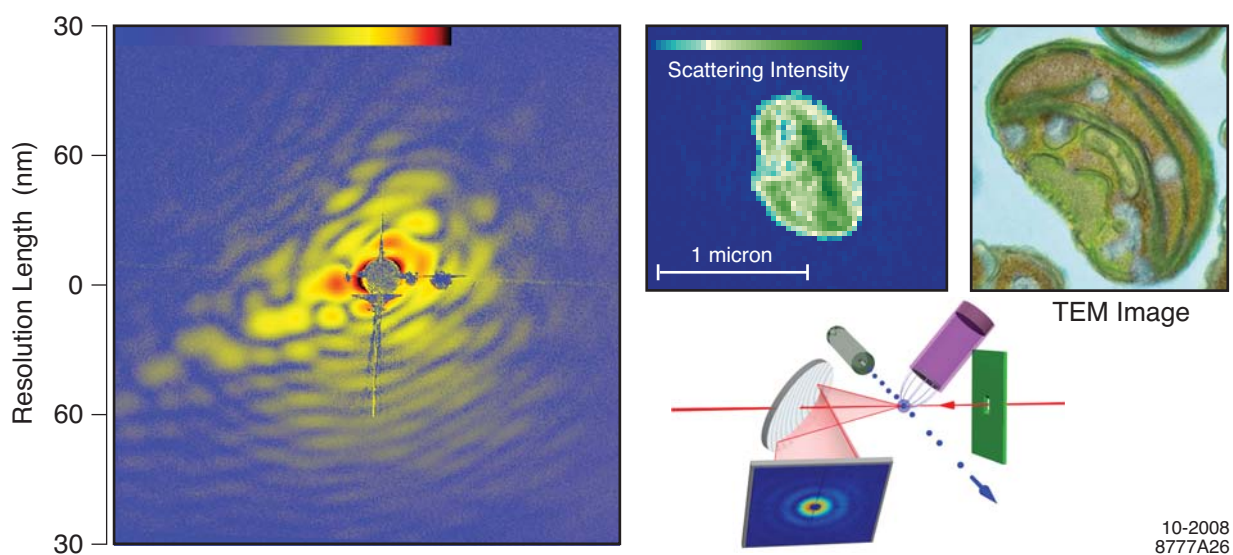
Figure 3.15. Left: Femtosecond single-shot x-ray scattering pattern. Top middle: The reconstructed image of a live picoplankton that was injected at 200 m/s into the beam of the soft x-ray laser FLASH operating at 7-nm wavelength, 15-fs pulse duration, and ~10^{14} W/cm^2 power density. Image reconstructed using Shrinkwrap (Sebastien Boutet). [Source: J. Hajdu et al., private communication] Top right: transmission electron microscopy (TEM) image. [Source: W. Eikrem and J. Throndsen, Univ. of Oslo]. Bottom right: Schematic setup of diffraction experiment. The soft x-ray image can be extended to molecular resolutions with an x-ray laser operating in the wavelength range from 1.5–0.5 nm.

3. Trace Element Mapping

X-ray fluorescence coupled with x-ray absorption spectroscopy techniques offers superb trace-element sensitivity and the ability to determine chemical speciation and local electronic and geometric structure of metal constituents in biological systems. To understand the biological significance of trace elements (especially metals) in health, disease, and pharmacological treatment, it is important to understand their distribution, concentration, and chemical state in cells and tissues as well as their many roles as key components of proteins and macromolecular complexes. For example, typically 25–30% of a given genome codes for proteins that contain or involve metals. Important first steps toward the high-resolution mapping of trace elements have been taken at third-generation light sources, but restrictions in source

brightness have made the process too slow to make a major impact and have limited resolution in many cases. Much higher average-brightness x-ray sources are required to bring this technique into the forefront of metal toxicology, pharmacology, and physiology. Higher brightness will also enable the study of metal-containing systems at in-vivo concentrations and under conditions where they are actually undergoing reactions and changes in the sub-millisecond time domain. In addition, use of genetically encoded metal-binding tags could open up studies of sub-cellular localization of any protein of interest.

4.   Damage Considerations

In all areas of life science and soft matter research with x rays, radiation damage is a critical limitation. As one moves to higher resolution, smaller samples, and higher sensitivity, one needs to be even more careful in the design of the experiment to make sure that the information collected is not compromised. There is considerable effort going into understanding these issues and finding mitigation strategies. In some cases, making the exposure at cryogenic temperatures is sufficient. In other cases a useful approach is to continually refresh samples, as with nanofluidics, a stream of molecules, droplets, or nanocrystals. Or, with sources of sufficient peak brightness and short duration, the information may be collected before the damage manifests itself. In all cases tested so far, there is evidence from imaging experiments at soft x-ray free electron lasers that this technique can work.

5.   What Is Needed?

To attain the goals in atomic-resolution crystallography and SAXS, a stable, ultrabright, hard x-ray source based on a ring is needed to provide highly parallel, micron, or submicron-sized beams. For fluorescence mapping, a similar source is needed to create an intense hard x-ray nanoprobe. Flash imaging and photon correlation spectroscopy require an ultrashort-pulse soft x-ray FEL with repetition rate adjustable from a few Hz to 100 KHz.

FOUR

X-Ray Source Types, Capabilities, and Their Trade-Offs

A. Types of X-Ray Sources with Enhanced Capabilities

In this section, we briefly discuss the major options for new sources in the 10–20 year time frame, based on the essential new x-ray capabilities that were described in Section 2.B, and what we believe is realistic. Figures 4.1 through 4.4 below summarize the approximate performance of present and future sources, including storage rings, energy-recovery linacs (ERLs) and linac-based FELs, based on projections from the present state of the art. While undulators are the primary source of photons for all sources described here, their performance characteristics are strongly linked to the accelerator technology used, and we group our discussion in terms of accelerator type accordingly. A combination of ring-based sources, including storage rings and ERLs, and linac-based FELs is identified as meeting the needs described in Section 2.B. A broader discussion of source options, including novel approaches that require considerable R&D before becoming practical, is included in Appendix A.

1.   Storage Rings

Storage rings provide highly stable average flux and brightness with low peak flux and brightness at very high pulse-repetition rates (typically hundreds of MHz) in a tunable photon energy range from the far IR to hard x-ray. Megahertz photon pulse rates, or even sub-megahertz rates for large rings, are possible using fewer bunches, and lower rates for timing or pump–probe experiments can be reached using a beam chopper. Pulse lengths are typically greater than ten picoseconds (rms); shorter pulse lengths can be reached at low current with lattice tuning and at high current with RF deflection systems and laser slicing. "Ultimate" designs offer exceptional brightness by pushing down the electron-beam emittance to ~0.1 nm·rad or less with large-circumference lattices and by supporting relatively high current, although high current operation presents challenges for high power absorber and optics designs. Even present facilities reach the diffraction-limited emittance for 1-Å radiation (8 pm·rad) in the vertical plane by reducing horizontal–vertical coupling and residual vertical dispersion. While ultimate rings would have higher peak brightness than most 3rd generation rings, some existing and in-construction rings reach ultimate ring peak brightness levels using long undulators and by storing a large current in a small number of bunches. With sufficiently bright beams partial x-ray lasing may be achieved, resulting in an amplification of emission by one or two orders of magnitude through the self-amplified spontaneous emission (SASE) process. Storage rings support a large user community with several tens of beam lines at each facility. Storage-ring technology and implementation methods are presently the most mature and reliable for multi-user facilities.

2.   Energy-Recovery Linacs (ERLs)

ERLs, now the target of R&D for future light source use, promise to provide very high average brightness by circulating a very low-emittance (~10 pm·rad), low-energy-spread (order 10^{-4}) round beam from a full-

energy superconducting linac for one turn around a ring-like transport line containing insertion devices. The beam is then returned to the linac, where the beam energy is recovered in the superconducting structure. Because the beam is not stored in the ring, the very low beam emittance from the linac, which actually decreases with increasing beam energy, is approximately preserved in the single turn around the ring, and performance limitations associated with ultimate storage rings, such as injection accumulation, lifetime, and intrabeam scattering, are avoided. Very high brightness with low current is reached by virtue of the very low emittance, reducing the total synchrotron radiation power load but also the photon flux, as compared with storage rings of similar brightness and higher current. While the bunch length is on the order of 1 ps in the high-current (~100 mA), high-brightness/coherence mode, a short-bunch mode is envisioned that would provide bunch lengths of tens of femtoseconds at the expense of much lower current and higher emittance. Bunch repetition rate may be adjusted from the MHz or less level for short-bunch, high-charge operation up to the linac frequency (potentially GHz) for high-current, high-brightness operation. The low energy spread enables an effective gain in brightness from high-order undulator harmonics and from long insertion devices (~25 m), but the energy spread must remain small to sustain the energy recovery in the linac. Emittance and energy spread increase as the beam travels from the first to last insertion device, especially in a short-bunch mode, reducing source brightness from the downstream devices accordingly. Low-energy ERLs are being used to drive infrared FELs, while high-energy ERLs would provide sufficiently small emittances and peak currents to drive SASE and possibly cavity oscillator FELs (XFELOs—see Section 4.A.3).

3.   Linac-Based FELs

Linac-based FELs will provide short x-ray pulses of unprecedented peak brightness and power, up to tens of GW per pulse, and photon energies from the far IR to hard x-ray. The first x-ray FELs will utilize SASE, producing pulses with very high spatial coherence and some degree of temporal coherence to provide line widths of ~10^{-4}. An FEL can also be used as an amplifier, as an oscillator, and as a harmonic generator. Employing seeding and other optical manipulations with conventional lasers such as high-gain harmonic generation (HGHG), self-seeding, electron-bunch slicing and/or length and charge optimization, and x-ray FEL oscillator (XFELO, or regenerative amplifier FEL—RAFEL) configurations, exquisite control of the x-ray pulse properties can be achieved in principle. A single linac can drive multiple FELs, each with independent tunability, and each configured for specific capabilities. X-ray pulses of 10–100 femtoseconds, and even sub-femtosecond pulses could be generated, as well as long (picosecond) pulses with near transform-limit temporal coherence (meV bandwidth).

The electron source for FELs can be a pulsed room temperature or superconducting linac, or a CW superconducting linac. The FELs under construction today achieve the high-energy electron beam required to radiate in the hard x-ray range using pulsed linacs with repetition rates for the macro pulses of ~10-120 Hz. Pulsed linacs operating in bunch train mode can provide high bunch repetition rates over the duration of the linac pulse length. Future CW superconducting linac-based FELs promise to provide high pulse repetition rate with even spacing, from tens of kHz to MHz or greater as electron gun technology is developed, and offer a route to higher average brightness with a time-structured beam that is well suited to many experimental needs, particularly pump–probe. The development of CW SCRF technology, also used in ERLs, offers improved stability over pulsed machines, allowing the use of broadband feedback loops to control beam position and energy.

4. Graphical Representation of Source Capabilities

Brightness: X-ray source brightness is defined as photon density in 6-dimensional phase space, with phase space dimensions defined by horizontal and vertical beam size and divergence, time and fractional bandwidth of energy (or frequency). The usual units for brightness are photons/s/mm^2/mrad^2/0.1% bandwidth. Average and peak brightness of various source types are compared in Figure 4.1.
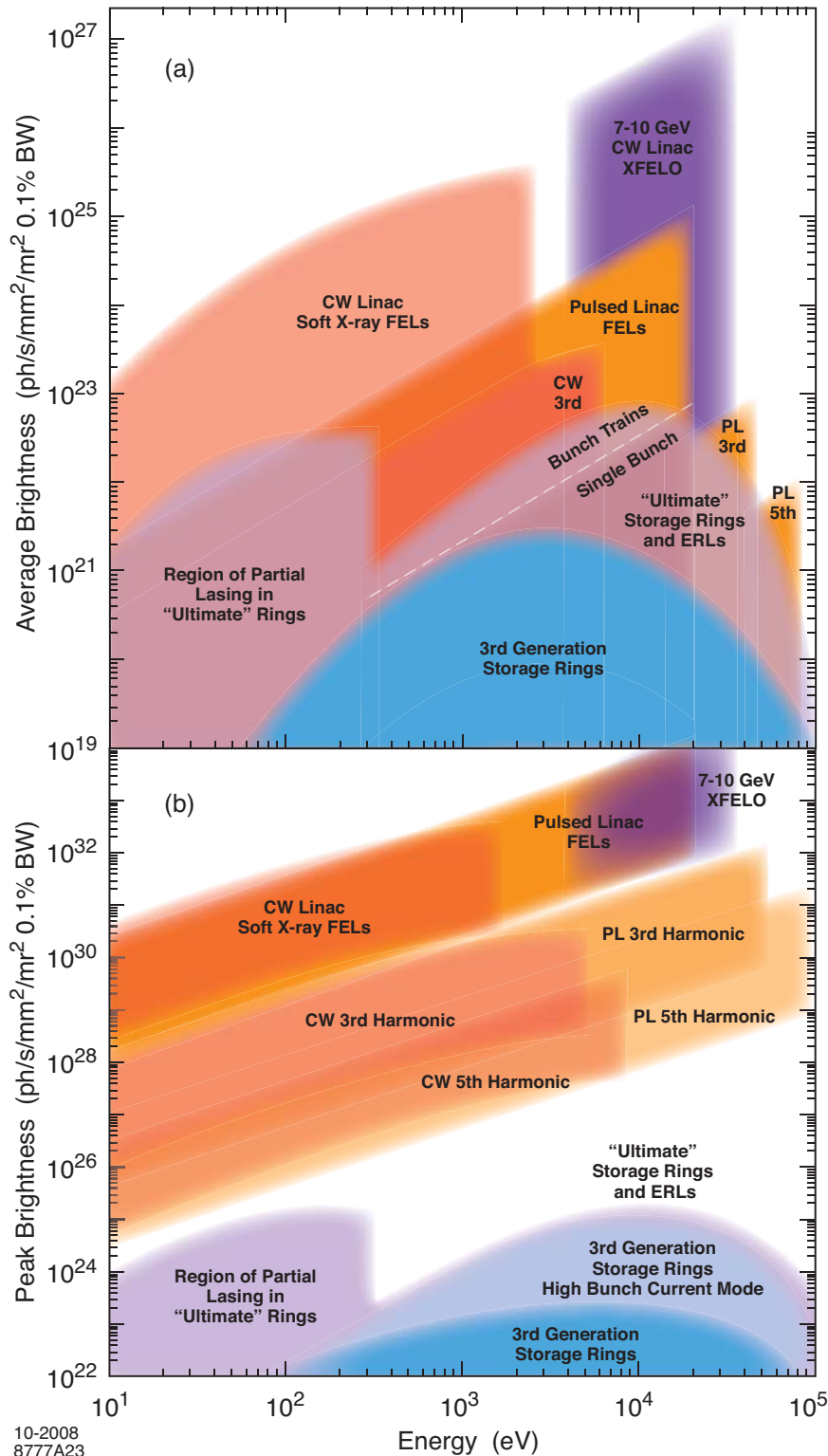
Figure 4.1. Composite envelopes indicative of the approximate range of average (a) and peak (b) spectral brightness for present and potential future technology source types. The envelopes are not inclusive of all sources that exist or are envisioned. The high average brightness from the linac- and ERL-based FELs, and speculative preliminary estimates for x-ray FEL oscillators (XFELOs), all of which have low repetition rate compared to ring- and ERL-based spontaneous sources (see Fig. 4.2), is not usable by a large class of experiments due to the assoc-iated very high peak brightness. Bright-ness of the seeded FEL and FEL oscillator sources is increased by their intrinsically narrow bandwidth, which can approach the transform limit, and can be several orders smaller than incoherent sources (see Figure 4.4). ERL-based, single-pass FELs may achieve peak brightness comparable to the CW and pulsed linac FELs, and average brightness comparable to the CW linac FELs indicated. XFELOs utilize crystal reflecting mirrors and schemes allowing wide tuning range are being investigated. See text below for discussion of assumptions made in figure.

In Figure 4.1, future-technology ERL performance assumes a 25-m undulator in high-brightness mode; 3.5–8-m undulators are assumed for large-circumference "ultimate" ring performance. Peak brightness from advanced 3rd generation ring sources having long undulators (~20 m) and operating with a large current per bunch (>~3 mA/bunch) in a small number of bunches approaches that shown for "ultimate" rings having shorter undulators and much less current per bunch. The peak brightness from long undulators on "ultimate" rings would be greater than shown if they too were operated with greater current per bunch. Pulsed FELs are presently in construction. CW-linac FELs and ERLs require future technology; notional designs indicated are for a 100-kHz bunch rate CW soft x-ray FEL and a 1.3-GHz bunch rate hard x-ray ERL. The range shown for FEL performance reflects SASE and seeded modes of operation and ability to tune performance over a wide range. The peak brightness for FELs is several orders of magnitude greater than incoherent sources, resulting in high average brightness even at low repetition rate. The average brightness from pulsed linac FELs can be increased by operating with a large number of closely-spaced bunches within the RF pulse of the linac, resulting in a "burst-mode" time structure—in this notional design 4000 bunches spaced by 200 ns repeating at 10 Hz. For FELs and sub-10 pm·rad emittance ERLs, the flux is spatially coherent; some FELs may have a high degree of temporal coherence. The potential brightness range of an x-ray FEL cavity oscillator (XFELO) is based on a 1-MHz pulse repetition rate with low-charge (< ~50 pC) picosecond electron bunches provided by a CW linac, multipass recirculating linac, or an ERL, and an x-ray optical cavity tuning system using four crystals movable in position and angle. The depicted upper energy limits for linac-based FELs are not absolute; they may be extended with higher-energy linacs. The lower energy bound for all sources extends to the sub-eV (THz) range.

While linac-based FEL sources can achieve high average brightness with bunch repetition rates much lower than those of rings and ERLs as a result of their coherent radiation, this fact has to be interpreted with care. The potential drawbacks of operating linac FEL sources in high average brightness mode are discussed in Section 4.B and in Appendix B.

Pulse Structure: The temporal characteristics of different sources are illustrated in Figure 4.2. The indicated notional designs indicated are for a 10- kHz bunch rate CW soft x-ray FEL and a 1.3-GHz bunch rate hard x-ray ERL. The number of photons per pulse for FELs is dependent on wavelength and electron-beam energy. The range in FEL performance reflects SASE and seeded modes of operation, and ability to tune performance over a wide range. For pulsed FEL bunch trains, the average repetition rate is calculated assuming 4000 bunches spaced by 200 ns, repeating at 10 Hz. The XFELO is based on a 1-MHz pulse repetition rate with low-charge (< ~50 pC) picosecond electron bunches provided by a 7-10 GeV CW linac.
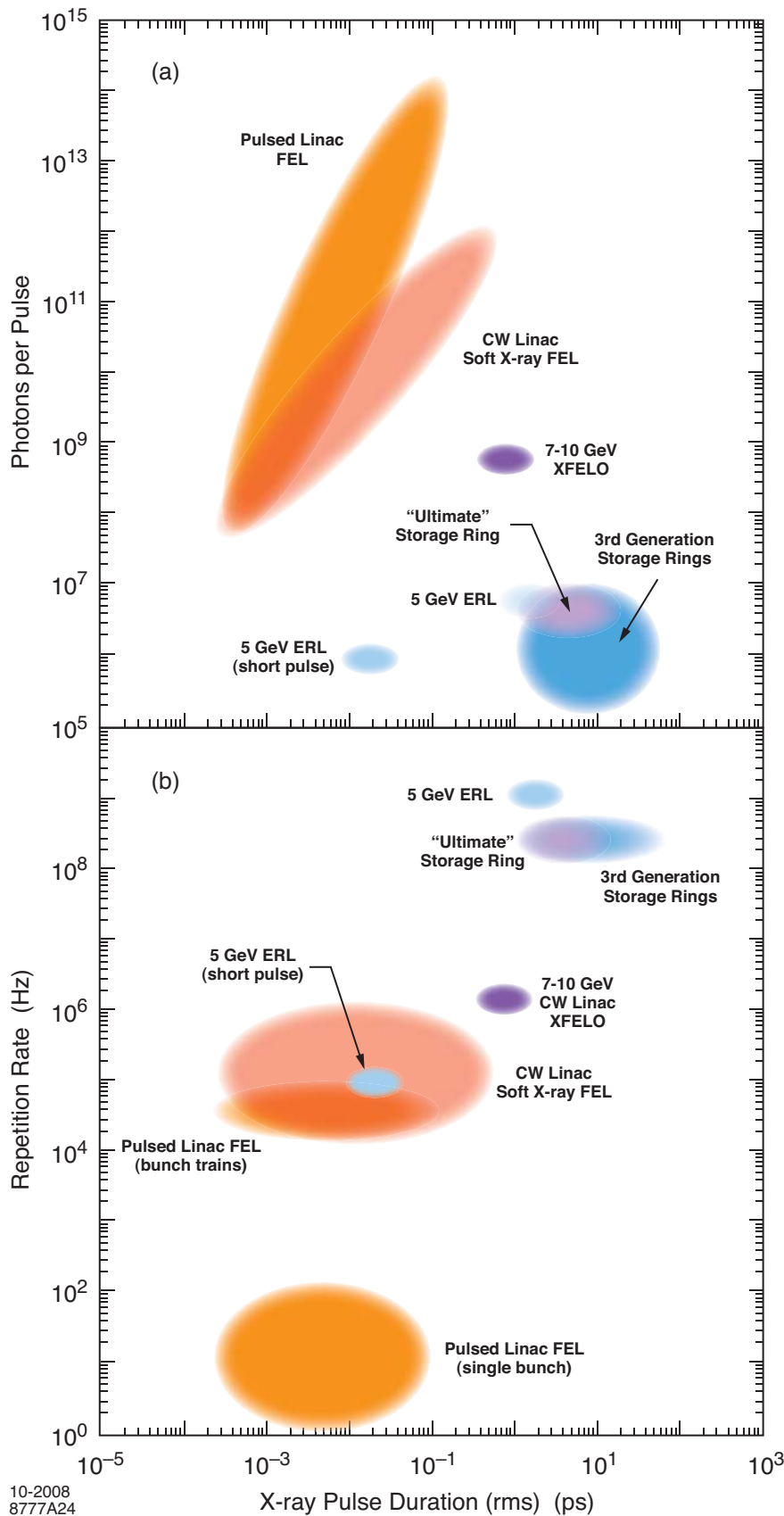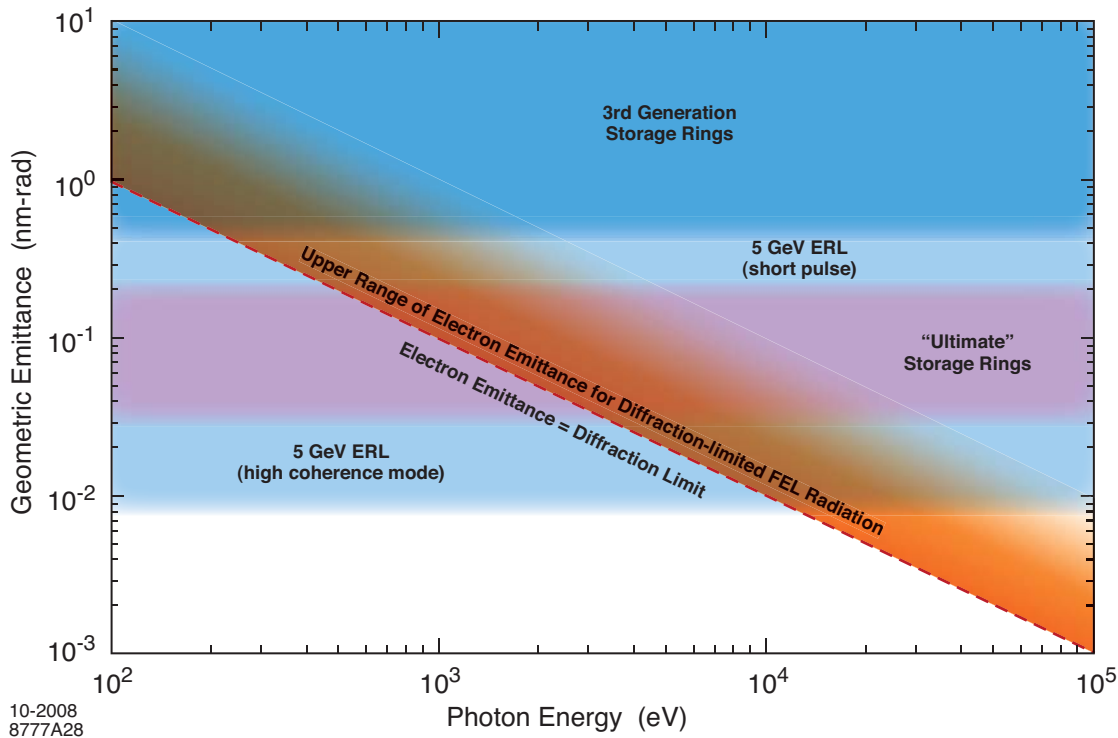
Figure 4.2. Envelopes indicative of the approximate performance ranges of present and future technology source types illustrating the trade-offs between peak and average performance parameters as discussed in the text. The envelopes are not inclusive of all sources that exist or are envisioned. See text below for discussion of assumptions made in figure.

Transform Limits: The quality of a photon source may be characterized by two limits, characterizing a perfectly coherent source. The "diffraction-limited" emittance ε_{ph} of a photon beam expresses the fundamental coupling of transverse source size and angular divergence in the space-momentum domain and defines complete spatial coherence. For a Gaussian beam, $\varepsilon_{ph} = \lambda/4\pi$. For spontaneous radiation generated from magnets in an electron accelerator, the photon beam emittance for a given wavelength is a function of source magnet properties (i.e. undulator or dipole magnet parameters) and of the electron beam emittance. In effect, the radiated photon emittance at a given wavelength is a convolution of the diffraction limited photon emittance and the electron emittance. As the electron emittance in one transverse plane approaches the diffraction limit for a given wavelength, the degree of spatial coherence, or coherence fraction, of the radiated photons in that plane increases, reaching the order of 50% from undulators when the electron emittance reaches the diffraction limit, subject to the matching of the electron beam emittance ellipse to the diffraction-limited photon beam ellipse. The coherent fraction increases towards 100% as the electron emittance is reduced below the diffraction limit. In contrast, the amplified radiation from FEL sources is 100% transversely coherent and diffraction-limited in both planes when the emittance of the round electron driving beam is close to or, for sufficiently long undulators, even greater than he diffraction limit, by virtue of "gain guiding", where only a single diffraction-limited radiation mode is amplified to saturation in the FEL process.

In Fig. 4.3 we have plotted the electron beam emittance versus photon energy for the different undulator-based sources of Figures 4.1 and 4.2.

Figure 4.3. Envelopes indicative of the approximate emittance ranges of present and future technology source types. The transversely coherent fraction at a given photon energy for spontaneous radiation sources on storage rings and ERLs approaches 100% as electron emittance is reduced below the diffraction limit. Radiation from single-pass FELs having sufficiently long undulators is 100% transversely coherent even if the driving electron beam emittance is somewhat larger than the diffraction limit.

Just like the "diffraction limit" characterizes complete spatial coherence, the "transform limit" (also called the "Fourier transform limit" or "bandwidth limit") defines complete temporal coherence. It expresses the fundamental coupling between energy resolution and time resolution, and is described by the product of time duration and energy width of the x-ray pulse, and for a Gaussian pulse intensity is given by $\Delta t \bullet \Delta E = h/4\pi = 0.33 \times 10^{-3}$ [ps (rms) eV (rms)].

In Figure 4.4, the bandwidth attainable with coherent and incoherent undulator-based sources is indicated for the notional designs discussed in Figures 4.1 and 4.2 and accompanying text.
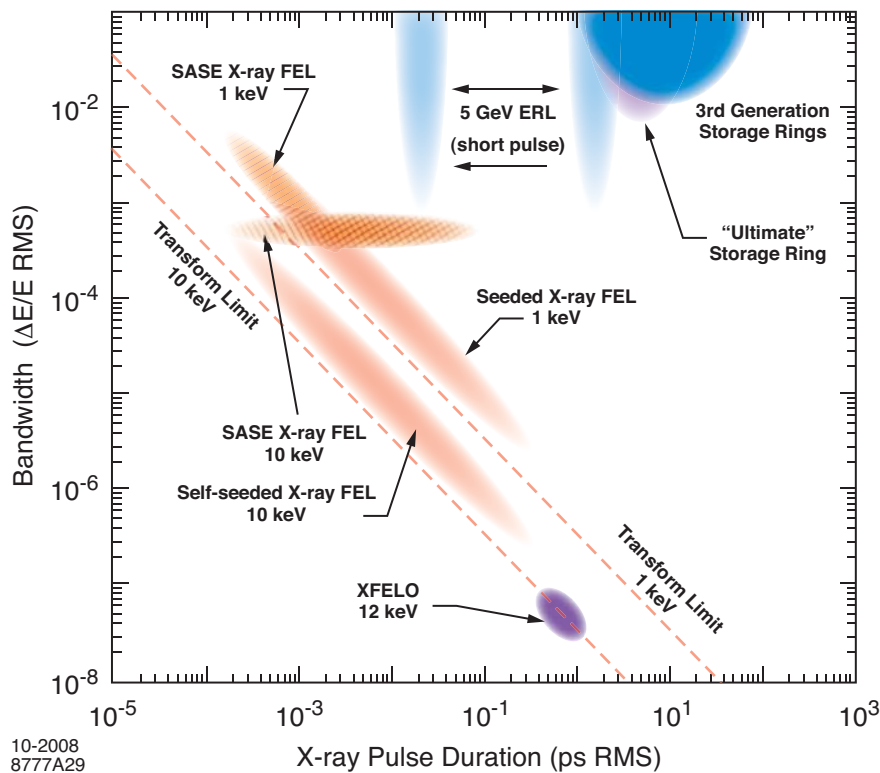
Figure 4.4. Envelopes illustrating fractional bandwidth capabilities of undulator-based sources in present and future x-ray facilities. Bandwidth is given for direct output from the source with no additional wavelength filtering. Dashed lines indicate the transform limit for 1 keV and 10 keV photon energies. Seeded FELs and oscillator FELs can have bandwidths approaching the transform limit, as can SASE FELs with very short bunches. Photon energies are indicated for specific examples used to illustrate coherent sources. Narrow bandwidth contributes to increased brightness. See text below for discussion of assumptions made in figure.

In practice, a transform-limited pulse has the shortest possible duration for a given energy bandwidth or resolution. Transform-limited pulses yield the highest brightness for a given total pulse energy, and are important for extremely brightness-hungry experiments such as those involving coherent multi-photon (nonlinear) processes. They are also required for phase-controlled pulse shaping techniques to be used.

Seeded FELs and FEL oscillators with long bunches can approach the transform limit with very narrow bandwidths. For these sources, deviations in the electron phase-space distribution along a bunch are expected to limit the bandwidth to a factor of a few above the transform limit. The seeded FEL envelopes in Figure 4.4 represent external laser seed and self-seeding capabilities, for FELs with 1 keV and 10 keV photon energies. The XFELO example in Figure 4.4 is for a 12 keV photon energy FEL oscillator. SASE

FEL envelopes are shown for photon energies of 1 keV and 10 keV. For SASE, the bandwidth is determined by the cooperation length (the distance over which the radiation field moves forward from the electrons in one gain length), not the bunch length. For ultra-short bunches the cooperation length can approach the bunch length, and the (large) bandwidth can be close to the transform limit. For ring and ERL–based sources, undulators radiate the smallest intrinsic fractional bandwidth (given by $\Delta E/E \sim 1/N_u$ where N_u is the number of undulator periods), over a very wide range of photon energies (from a few eV to tens of keV), with bandwidth down to a lower limit imposed by the coherent energy spread in the electron beam (typically $>10^{-3}$ for rings and $>10^{-4}$ for ERLs). This characteristic of ring-based sources, and the much broader spectrum offered by dipole magnet radiation (where $N_u \sim 1$), can have advantages in experiments where a large bandwidth is required. For large bandwidth sources, the bandwidth of radiation for an experiment is typically determined by that of the beam line's energy selection and filtering device, such as a crystal monochromator, and is therefore an adjustable parameter, which can be advantageous for some experiments. In contrast, this bandwidth selection is not possible for many FEL sources, although their very narrow bandwidths can eliminate the need for monochromators.

B. Discussion of Relative Capabilities and Trade-Offs

In the discussion of x-ray source performance capabilities in begun Section 4.A, and continued in Appendix A, brightness (both peak and average), coherence, pulse structure and transform limits have been used as metrics. While the relative advantages and disadvantages of source types might be assessed using these metrics, the choice of a preferred source for any given experiment is ultimately based on maximizing the number of usable photons delivered in the phase-space acceptance of the experiment within the data acquisition period of its detector system, and is thus experiment specific. Some experiments demand the high peak brightness and coherence of an FEL source; others demand high flux in a relatively large acceptance phase space and are therefore not entirely brightness limited. Such experiments would benefit from a higher current ring-based source. As another example, experiments demanding very high energy resolution might not benefit from ultra-short pulses, while those demanding high time resolution would benefit. The breadth of beam property requirements for present and future x-ray science ensures that, given our present understanding, a combination of linac-based FEL and ring-based sources will be required to accomplish the x-ray science of the future. The relative capabilities and trade-offs of these new sources are discussed below.

The spectral brightness characteristics of the sources discussed in Section 4.A are compared in Figure 4.1, which reveals that FEL sources are not only superior in peak brightness but also have larger average brightness than storage ring sources. The FEL advantage in peak brightness arises because the FEL amplification process produces a very large number of photons in each pulse (see Figure 4.2), and packs these photons into a small (diffraction-limited) phase space. The high average brightness arises because the FEL peak brightness is so great that it more than compensates for the low repetition rate. FELs have another advantage over ring-based sources, due to the fact that their electron pulses are prepared by a linac and then used only once to create x rays. This allows, in principle, precise control over each individual x-ray pulse. For example, pulses with a useful number of photons within durations of less than about 10 fs can only be achieved with FEL sources (see Figure 4.2). The large number of photons per pulse can be an advantage for any single-pulse experiment, where the experimental conditions are not repeatable (see Section 3.D above). At a more profound level, the greater coherence of FEL x rays compared to spontaneous x rays could enable new research techniques involving nonlinear x-ray interactions. FELs therefore offer a range of x-ray capabilities that is unique and cannot be covered by storage ring or ERL sources.

One may be led to conclude that all future sources should be FELs. The situation is however considerably more complex, as already briefly mentioned in Section 3.C.5, and further discussed in Appendix B. Many experiments will be unable to exploit the high average brightness of an FEL, because they will be overwhelmed by the huge peak brightness. Limitations may arise from practical matters such as instantaneous detector count rates, or from sample "damage" (significant modification of the sample properties by the FEL pulse during the measurement, see Appendix B). FEL bunch trains may exacerbate these difficulties, if the spacing of the bunch train does not allow synchronization with pump lasers, sample relaxation or recovery, or replenishment or translation. Also, not all experiments are improved by added brightness; for many the figure of merit is simply the flux into the (fixed) sample volume as discussed above.

Ring-based sources (storage rings and ERLs) provide high average flux and brightness with low peak flux and brightness. They support tunable devices that provide photon pulses at very high repetition rates, and may be considered "CW" sources for many applications. In addition, storage-ring-based sources now incorporate more highly developed technologies for stabilizing source position and angle than do linac-driven sources, relying on the static nature and periodicity of storage rings. (Nonetheless, for a linac the position and angle can be maintained to a fraction of the radiation pulse radius and angular divergence.) Storage ring-based sources are proven technology, have well-established user communities, and are essential to a broad range of science; the next-generation storage rings use mature technology, and are ready to be built.

Only the combined capabilities of ring-based and FEL sources provide the complete range of x-ray characteristics required to address the grand challenges and to meet the broad set of science requirements with the wide range of technologies users need. A combination of FELs of different kinds (SASE, seeded, and oscillator configurations have unique and complementary characteristics) and storage-ring or ERL sources will provide the "total information" needed to understand complex and emerging phenomena in quantum matter and detailed dynamics of chemical reactions and biological processes.

Existing storage-ring spontaneous x-ray sources can produce a maximum photon density that is equal to the value of the fine-structure constant, or about 0.01 photons per "coherence volume", a unit of phase space whose 6 dimensions satisfy transform limits (sometimes referred to as the degeneracy parameter). Note that this is simply another way of normalizing the spectral brightness, or of describing how far the pulse is from the transform limit in which all photons are in the same mode. Because ring-based sources produce low-degeneracy x rays, they are not well suited for exploiting nonlinear x-ray effects. ERLs produce photons from a smaller phase-space volume, and may have degeneracy of ~100; FEL sources, through the coherent amplification process, produce highly degenerate x-ray pulses (10^7 or more photons per coherence volume, with further orders of magnitude increased degeneracy obtainable from seeded and oscillator FELs). The high degeneracy available from FELs is relevant for any non-linear effect requiring the simultaneous presence of multiple coherent photons within a short pulse. In principle, this should enable the extension into the x-ray range of nonlinear techniques (frequency doubling, 4-wave mixing, etc.) that are used routinely in the optical frequency range. For these techniques to be generally useful, it would also be necessary to identify non-linear x-ray processes with reasonable cross-section. This research, and the exploitation of nonlinear x-ray effects, falls within the range of science described in Section 3.A.

FIVE

Other Considerations

A. U.S. X-Ray Sources in the International Environment

In the last five years, there has been a great effort worldwide to construct and develop x-ray light sources culminating in the successful completion of a number of new third-generation sources, in the upgrade and improvement of several others, and in the construction and development of the first fourth-generation sources. The relevance of such activity is clearly evident in Figure 5.1 that shows the proposed and funded x-ray light sources and R&D facilities around the world.

Several of the new third-generation light sources have been successfully commissioned (DIAMOND in the UK, SOLEIL in France, ASP in Australia, and SPEAR III at SLAC), and a number of them are under construction (SSRF in China, TPS in Taiwan, ALBA in Spain,, and NSLS II at Brookhaven) . Groups at some of the older light sources are now proposing significant facility upgrades and modifications that will enhance their source performance (APS at Argonne, ALS at Berkeley, ESRF in France, and Spring-8 in Japan), and at former collider facilities conversion to high-brightness light sources is under way (PETRA III at DESY in Germany) or in conceptual planning (PEP-X at SLAC). Significant effort worldwide is aimed at bringing in the next decade the ring-based sources from the present brightness of ~ 10^{19} B.U. (B.U. = brightness unit = photons/s/mm^2/mrad^2/0.1%BW) above 10^{22} B.U., towards their ultimate limit near 10^{23} B.U.
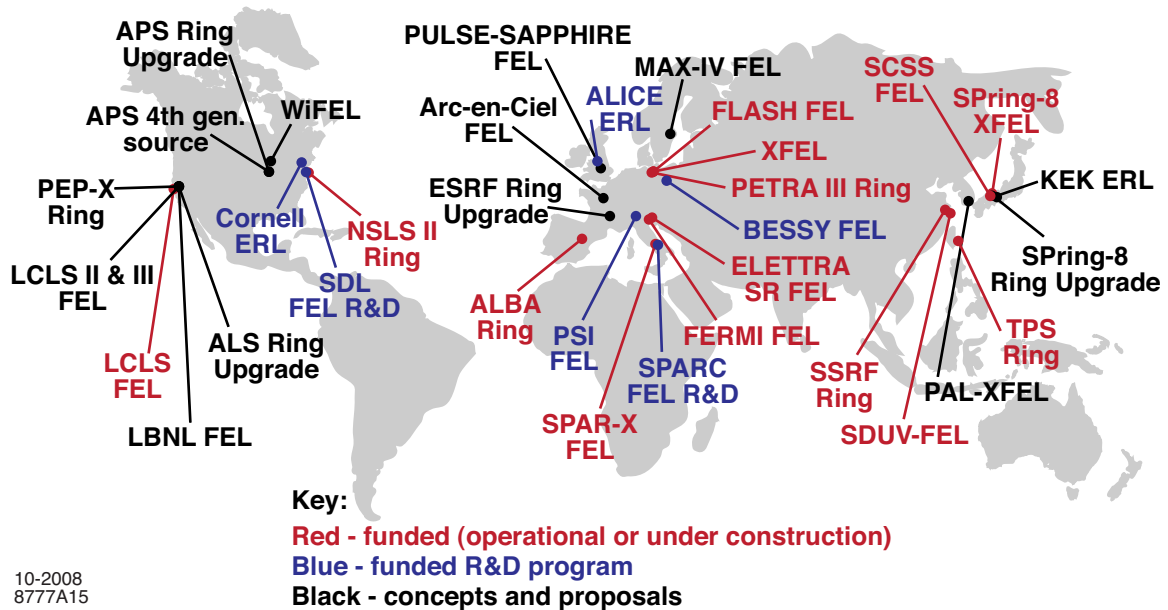

Figure 5.1. Proposed and funded x-ray light sources and R&D facilities around the world.

In parallel, interest in a "fourth generation" of light sources has shown a sharp increase in the international communities of users and accelerator physicists. FELs offer revolutionary performance, including peak

brightness of more than 10^{33} B.U. and sub-ps pulse lengths, open the door to new types of experiments, and nicely complement the outstanding science results in the consolidated fields investigated by the large and steadily growing user communities of third-generation light sources. ERLs with their very high average brightness exceeding 10^{22} B.U. and short bunch capabilities, together with "ultimate" storage rings, represent a natural extension of third-generation light sources beyond their present performance limits. In fact, some of the ERL concepts under development include an existing storage ring in their layout.

The first VUV-soft x-ray fourth-generation light source user facility, FLASH in Hamburg Germany, operating between 50 and 6.5 nm, has demonstrated the ability of an FEL-based source to perform as a reliable facility and is already generating significant science results. Recently the SCSS Test Accelerator in Harima, Japan, has opened a user program operating in the 50 to 60-nm range and will be developed further to reach the 3 to 4-nm range. In the hard x-ray portion of the spectrum, LCLS at SLAC (1.5 Å) is presently under commissioning, the European XFEL (1.0Å) has been launched with construction to be in Hamburg Germany, and the SPring-8 Joint Project for XFEL is building a 1.0-Å facility in Harima, Japan.

The broad international activity in light source construction, development, and planning highlights today's strong user support, as well as competition, for the development of next-generation light sources. In the construction and upgrade of third-generation light sources, Europe and Asia have invested and are continuing to invest large resources. A few years ago, the U.S. upgraded SPEAR III at SLAC and recently funded NSLS II at Brookhaven, thereby taking very important steps towards maintaining the U.S. competitiveness in the field. To pursue the same objective in the future, the effort should continue by funding the upgrades of the veteran U.S. facilities and by supporting the construction of new ones.

Activities in the development of VUV and soft x-ray FEL facilities are dramatically growing as well. In addition to the already operating FLASH, a large number of laboratories around the world are proposing or developing concepts for FEL facilities in this wavelength range (LBNL FEL at Berkeley, BESSY-FEL in Germany, WiFEL in Wisconsin, SAPHIRE and PULSE in the U.K., Arc-En-Ciel in France, MAX IV FEL in Sweden, and PAL in South Korea). The SPARX project in Italy and the SDUV in China has been funded, and FERMI@Elettra is presently under construction. An interesting characteristic of soft x-ray FELs is that their lower photon energy makes the use of seeding techniques appealing and more realistic, and in fact the majority of the projects include seeding schemes for improving the temporal and spectral characteristics of the photon pulses.

Preserving the U.S. competitiveness in the case of soft x-ray FEL sources requires effective short-term action in supporting R&D which will allow implementation of advanced x-ray facility capabilities within a decade. In Europe and Japan, several FEL R&D facilities have been funded and some are already in construction. In the U.S. the activity is in an initial proposal phase where innovative ideas and creative schemes using state-of-the-art techniques and beyond are ready to be exploited if the projects are funded. Support for initiatives within the U.S. can allow us to compete with and perhaps lead the world in such an important field.

Concerning hard x-ray FELs, the U.S. is presently at the forefront of the field with the LCLS scheduled to begin operations in summer 2009. In order to maintain this leadership when the XFEL in Europe and the SPring-8 XFEL in Japan start operating, development and upgrades of the LCLS as well as new initiatives in the field should be seriously considered. Argonne is considering a fourth-generation source based on a hard x-ray FEL oscillator. R&D on a high-brightness electron injector and 250 MeV accelerator at PSI in

Switzerland is leading toward design of a hard x-ray facility reaching 1.0Å. Argonne is considering a fourth-generation source based on a hard x-ray FEL oscillator.

Development of x-ray ERL facility concepts is also underway. After the successful operation of smaller-scale and longer-wavelength ERL-based facilities (JLAB FEL in Virginia, JAERI in Japan, and the ERL at BINP in Russia), Arc-En-Ciel has a 2-GeV ERL option, and the studies at Cornell and at KEK and JAEA in Japan for the construction of 5-GeV hard x-ray facilities with average brightness exceeding 10^{22} B.U. and below 100-fs bunch length capabilities are in an advanced stage of study with R&D efforts in progress. The Cornell layout includes the existing CESR ring, and the APS group at Argonne have discussed a similar ERL machine as a possible long-term upgrade for their facility. R&D on ERL technologies (also relevant to FELs) is in progress at the ALICE facility in the U.K. The U.S. effort is led by the funded R&D program at Cornell. The main international competitor is the ERL being studied at KEK and JAEA in Japan and Arc-En-Ciel in France, but currently the US has a lead in design and prototyping for an ERL, which should not be lost.

In competition with developmental ERL-based x-ray sources, and sharing the user interest and support for developing high average brightness, low peak brightness, high repetition rate sources, are "ultimate" storage rings. While these rings have longer bunches with larger energy spread than ERLs, they have brightnesses similar to ERLs, achieved with more conventional technology by virtue of their large circumferences, enabling very low emittance lattice implementations, and, in some cases, also due to their higher operating current (Section 4.A.1 and Appendix A.1). Ultimate rings are being investigated by SPring-8, the APS and most recently by SLAC, which would use the 2.2-km tunnel, injector and much of the accelerator technology developed for the recently closed PEP-II B-Factory.

The broad interest in next-generation light sources is also evident from the number of R&D facilities proposed and operating around the world to investigate critical subsystems and techniques used in FEL or ERL schemes. These include among others, high-brightness electron guns, emittance compensation and manipulation, seeding techniques, laser synchronization and ultra-stable timing distribution systems, laser and cathode development, and electron and photo beam diagnostics. R&D facilities such as SPARC in Italy and the SCSS test accelerator in Japan are already in operation; the MAX-Lab seeding facility in Sweden and ALICE in U.K. are being commissioned; the PSI FEL R&D in Switzerland has been already funded; and others such as STARS in Germany and APEX at Berkeley, have been proposed. Cornell is commissioning a prototype ERL gun and injector, KEK is preparing for a test ERL loop, and JAEA is developing an ERL high current and ulta-low emittance gun. Appendix A.7 highlights x-ray source R&D requirements.

B. Growth in the U.S. User Community

One of the great strengths of national x-ray facilities is that they support a broad user program, bringing together researchers from academic institutions, government laboratories, and industry not only nationally but worldwide. Because access is free and solely based on the quality of the proposed research, such facilities foster research by small groups that often are not from research universities but undergraduate institutions who bring their students. X-ray facilities thus play a major role in student education.

The historical growth of the synchrotron community through 2007 is illustrated in Figure 5.2. The figure illustrates only the number of on-site users that actively participated in experiments. The total number of scientists involved in synchrotron radiation research is considerably higher. History has shown that the

growth of the community generally increases with the offered opportunities, e.g., the availability of experimental stations, and we expect this trend to continue.
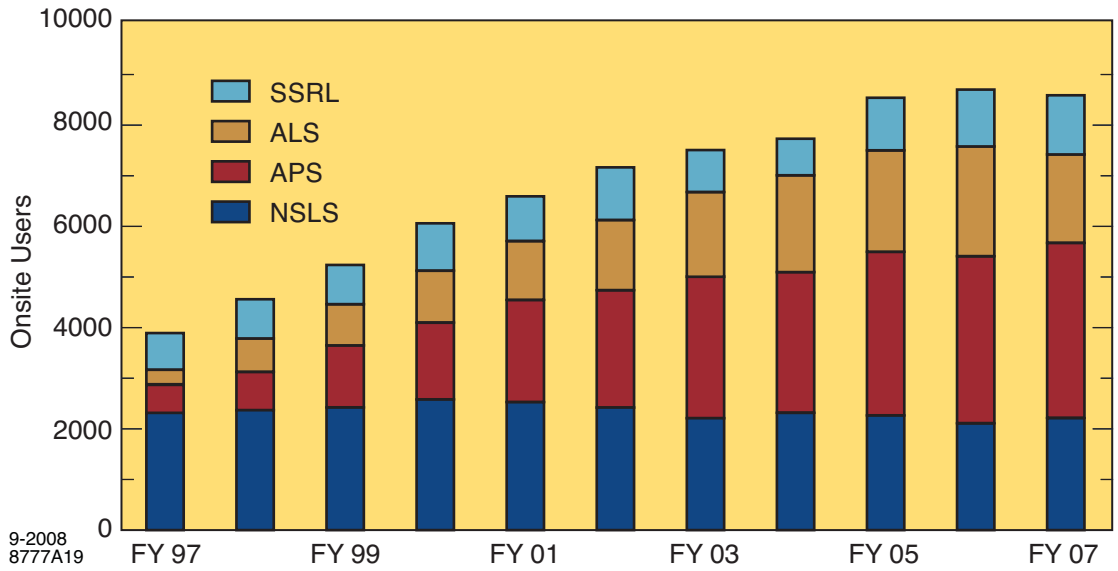
Figure 5.2. Growth of the number of on-site users versus fiscal year of the four BES operated light sources in the U.S.

APPENDIX A

Basic Types of X-Ray Facilities

A.1 Storage Rings

Storage rings, with electron currents from a few hundred milliamps to the order of 1 amp, provide high average flux and brightness (but low peak flux and brightness) x rays at high pulse repetition rates (up to hundreds of MHz). While the inherent electron bunch energy spread is nominally 10^{-3} (which degrades the brightness of higher undulator harmonics and long undulators), the pulse-to-pulse energy stability is very high ($\sim 10^{-5}$) and meV energy resolution in experiments can be reached. Photon pulse lengths are typically greater than ten picoseconds rms in duration, but shorter pulse lengths can be reached at low current with lattice tuning, and at high current in limited sections of the ring with RF deflection systems and optical manipulations. Laser slicing has been implemented on storage rings to produce ~100 fs x-ray pulses, at repetition rates into the tens of kHz, and with flux per pulse limited by the fractional portion of the bunch charge involved in the process. Storage ring sources provide extremely stable source points and are efficient in operation, have high reliability (>95% up-time for most facilities), provide tunable output from far IR to hard x-ray (up to tens of keV in the harmonics of undulators), and support a large user community with several tens of beam lines at each facility.

While present facilities reach the diffraction limited emittance for 1-Å radiation (8 pm-rad) in the vertical plane by reducing horizontal-vertical coupling and vertical dispersion to very small values, large circumference "ultimate" ring designs (nominally 2-3 km in circumference) begin to approach a diffraction-limited horizontal emittance (in the 10-100 pm-rad range) and offer exceptional average brightness (of order 10^{22} photons/s/mm^2/mrad^2 per 0.1% bandwidth). Because the ring dipole magnet fields are typically very low in these machines, high power wigglers might be used to provide the radiation damping required to reach low emittance. Ultra-low emittance rings tend to have a small dynamic aperture which makes normal multi-pulse refilling of bunch charge during injection more difficult. An on-axis injection scheme that replaces the full charge of a bunch train in one shot might be needed to realize the very lowest emittance rings. Linac current limitations could restrict the attainable current in the storage ring with this on-axis injection mode unless a separate accumulator ring is used to stack bunch charge prior to injection. The beam lifetime in these machines will be very short due to the small bunch volume. The short lifetime can be improved using bunch-lengthening RF cavities, but will nevertheless necessitate frequent top-off or bunch-replacement injection.

Preliminary studies indicate that when the electron emittance is at or below the diffraction limit for a given wavelength in both planes, partial SASE lasing with the stored beam is possible if the peak bunch current is sufficient with only a small impact on electron emittance and energy spread. Seeded lasing in a storage ring, at the repetition rate of the seed laser, has already been demonstrated in the EUV.

Very low emittance ring performance is limited by intrabeam scattering (which determines the minimum attainable emittance for finite bunch current), fast-ion and possibly electron -cloud instabilities, impedances from the vacuum chamber and coherent synchrotron radiation (CSR) that lead to bunch instabilities, synchrotron radiation and RF heating from the relatively high stored beam current, and orbit

and beam line component stability. In addition, having an affordable ring size represents a practical limitation.

A.2. Energy Recovery Linacs (ERLs)

ERLs are a potential source for future generation synchrotron light sources that combine some of the qualities of storage rings with those of linac-based light sources. A CW superconducting RF (SCRF) linac provides high repetition rate beams extending to the RF frequency of the accelerating structure (up to GHz) and, for the highest beam power, the ERL configuration is advantageous in providing an affordable RF power system (although the cryogenic system might be quite costly) by recovering the energy of the electron beam. This emerging technology promises to provide very high average brightness with high spatial coherence (~50% or more) by preserving the very low emittance and low energy spread achievable from a full-energy, high-current superconducting linac (~10 pm-rad and 10^{-4} respectively), for a single turn around a ring-like transport lattice accommodating several insertion devices. The electron beam would then be returned to the linac where it is decelerated to recover the energy in the superconducting RF structure. Growth in the energy spread of the beam, which occurs as the bunches traverse the ring, must be controlled to avoid beam loss in the linac structure during deceleration.

When operating with the design goal current of ~100 mA, ERL average brightness would exceed 10^{22} photons/s/mm^2/mrad^2 per 0.1% bandwidth from a long undulator (~25 m), and the inherently small beam energy spread would enable greater brightness at higher undulator harmonics than can be reached with a storage ring. While the bunch length is on the order of 1 ps in high current (~100 mA), high brightness mode, a short-bunch, low repetition rate (~1 MHz) mode is envisioned that would provide bunch lengths on the order of tens of femtoseconds at the expense of much lower current, and higher emittance. Dozens of high performance beam lines can operate simultaneously on an ERL; the number is ultimately limited by the growth of beam emittance which should not be more than a few tens of percent. In short-bunch mode, coherent synchrotron radiation in dipoles can increase emittance and energy spread as well, although the mitigation of these effects by longitudinal bunch shaping is being explored. When every bucket is filled, the electron charge per bunch for high-current ERL operation is limited to <100pC in today's designs by higher order mode heating in the superconducting linac and by wake-field driven beam break-up. Larger bunch charges need designs with more extensive higher-order-mode absorbers. A total current of up to ~100 mA might be reached by filling every closely spaced bunch (~0.7 ns spacing for the nominal 1.3-GHz linac frequency) in a large ERL ring with 77 pC per bunch. However, ERLs have flexible fill patterns so that more charge per bunch is possible for reduced bunch repetition rate.

Low-energy ERLs are already being used to drive infrared FELs. Future high-energy ERLs might provide sufficiently small emittance, high peak current and low energy spread to drive x-ray FELs. ERL-driven single-pass SASE and seeded FELs providing very high peak brightnesses have been envisioned and x-ray cavity oscillators (XFELOs) are being investigated (see Section A.3). The energy spread induced by such devices might limit the number of simultaneously operating FEL beam lines to only a few. In the case of the exploratory x-ray FEL cavity oscillator, the number of bunches, and thus average current, would have to be drastically reduced (from 100 mA to ~0.025 mA) to limit the cavity power to 20 MW or less. In this case, the XFELO would operate either in a dedicated mode or simultaneously with other beam lines if a separate, higher energy injector for the FEL is used in tandem with the normal ERL injector so that the FEL drive beam can be separated from the high current ERL beam after acceleration to high energy.

ERLs having the properties needed for future x-ray light sources face several technical challenges and are presently under development in the US, Japan and Europe. Performance limiting factors include the length of the linac and its cooling requirements, which limit beam energy, beam break-up and higher-order-mode heating in the linac structure, space charge limitations to low emittance in the gun for high bunch charge, effects on the electron beam from incoherent and coherent synchrotron radiation, nonlinear beam dynamics, ion trapping and orbit instability from ground motion.

A.3   Free Electron Lasers (FELs)

A free-electron laser is a source of coherent radiation that may be implemented on a linac, storage ring, ERL, or other electron source of suitable beam quality. An FEL introduces micro-bunching of the electron beam, with periodic enhancement of the charge density forming micro-bunches, separated by a distance equal to the wavelength of the emitted radiation. These micro-bunches begin to radiate coherently, significantly increasing the peak brightness. FELs can be either single pass, high-gain type, or an oscillator type making use of high reflectivity mirrors. The single pass, high-gain type can further be operated in two modes, SASE and seeded. In SASE FELs, the microbunching arises from the interaction of the electron beam and the synchrotron radiation emitted upstream in the undulator; seeded FELs introduce an external coherent light pulse that produces microbunching in the electron beam. In an oscillator FEL, a light field develops inside an optical cavity as the undulator radiation from previous passes is trapped and filtered by the mirrors and amplified by interacting with new electron bunches in the undulator. The spectral brightness of an FEL can be billions of times higher than that of a typical storage ring insertion device. In addition, light from an FEL has full transverse coherence and, in some FELs, a very high degree of a temporal coherence. Using the same electron beam in FELs and other coherent synchrotron radiation devices, each at a different wavelength, offers capabilities for synchronized multi-color sources.

The electron beam quality for an x-ray FEL must exceed that of a storage ring, and this is typically provided by a linac. The electron beam itself must have laser-like quality, with normalized transverse emittances <1 mm·mrad , relative energy spread $\sim 10^{-4}$, and peak current of hundreds of amperes to kiloamperes. Limitations arise from the thermal emittance of the electron source, and transverse and longitudinal emittance growth from space-charge, CSR, and impedance effects. Bunches may have up to 1 nC charge and achieve these parameters, although production of ultra-short bunches optimized for lasing with only 1-100 pC charge per bunch has the potential to overcome some limitations to emittance and to produce ultra-bright beams and intense hard x-ray pulses with femtosecond or shorter duration.

Existing FEL facilities are limited to ~200 eV photon energy, and the currently achieved normalized emittance of 1 mm·mrad at 1 nC charge requires that the beam energy be greater than 10 GeV for lasing at photon energies extending to several keV. Planned SASE FELs will provide short x-ray pulses (tens to hundreds of fs), of unprecedented peak power (10 GW), and peak brightness is expected to be of the order of 10^{33} photons/s/mm^2/mrad^2 per 0.1% bandwidth, many orders of magnitude greater than storage ring or ERL incoherent sources could provide. Implementation of optical manipulations could allow exquisite control of the electron beam, and development of techniques such as high-gain harmonic generation (HGHG), enhanced–SASE (ESASE), and self-seeding, are expected to enhance performance of planned and future FELs, by controlling x-ray pulse duration (<1–100s fs), imparting a very high degree of temporal coherence, providing synchronization to another laser for pump–probe experiments, and increasing peak current, thus reducing the length of the radiator undulator required. Seeded FELs may be able to operate with high stability at pulse energies below saturation, reducing the peak output flux and

brightness in a controlled way. Seeded long (~ps) pulses of close to transform–limit quality could provide meV resolving power, as could FEL oscillators. Seeding by conventional lasers and harmonic generation in an FEL has been demonstrated at VUV wavelengths. Feedback control of the seed laser could shape the amplitude and phase, optimizing x-ray pulse parameters for experiments.

Practical repetition rate for x-ray FELs is currently limited by high-brightness electron gun technology, and the facilities currently under construction are planned to use macropulses at ~10-120 Hz repetition rate. Pulsed linac repetition rates might eventually approach the order of 1 kHz with developments in pulsed high power RF sources and accelerating structures. In bunch-train mode, pulsed linacs can provide high bunch repetition rates (in principle up to the RF frequency of the linac) over the duration of the linac pulse length (approaching ms for superconducting linacs) to achieve average bunch repetition rates that may reach the order of 10^5 bunches/second. In contrast to the high bunch rates achieved in bursts with pulsed linacs, CW superconducting–linac-based FELs promise to provide high pulse repetition rate with even spacing, from tens of kHz to MHz or greater as electron gun technology is developed. CW superconducting–linac-based FELs offer a route to higher average brightness with a time-structured beam that is well suited to many experimental needs, particularly pump–probe. The bunches from the linac can be fanned out to multiple undulators, at the expense of proportionately reduced average brightness for each. The individual FELs can be configured for either SASE, seeded, or oscillator mode of operation dependent on experimental requirements, including use of HGHG, self-seeding, or oscillator configuration to obtain high temporal coherence.

SASE x-ray FELs using pulsed linear accelerators of ~14-20 GeV are being built, and the repetition rate is limited to somewhat less than 1 kHz, by power dissipation in the accelerating structures and other components, for either normal-conducting or superconducting linacs. Average brightness of 10^{25} photons/s/mm^2/mrad^2 per 0.1% bandwidth may be achieved by using trains of bunches within the macropulse. SASE FELs exhibit pulse-to-pulse wavelength jitter of ~10^{-3} and intensity jitter of ~10%-20%. Seeding is required to reduce these values to <10^{-4} and ~1% respectively. A higher energy beam, potentially extending to 50 GeV, would both increase the power output (approximately linear with beam energy) and extend the emission into the several tens of keV range. Reducing the electron beam emittance by about one order of magnitude, made possible by reducing the bunch charge to a few to tens of pC, would also allow the extension of the lasing wavelength to about 0.02 nm, or 60 keV, at about 10–15 GeV beam energy, using a shorter period undulator. Use of tapered undulators, designed to maintain the FEL resonance condition as the electron beam loses energy along the FEL, may give a further increase in output power.

The great attractions of CW SCRF for future FEL facilities are the high repetition-rate achievable with even pulse spacing, and the capability of supporting a large number of beamlines each with high average flux and brightness, when switching beam between different FELs. CW operation of the accelerator, with feedback loops to control beam position and energy, offers improved stability over pulsed machines. A high-brightness source operating in CW mode could drive an array of many FELs, with each beamline operating at high repetition rate (from 10 kHz to MHz and higher) and with even pulse spacing. Higher repetition rates could be supported by the linac, and CW linacs offer the potential for increased average brightness as technology is developed. The x-ray wavelength achievable is limited by electron beam quality, and by linac energy (a cost issue). With a small footprint, 2-3 GeV linac, FELs could operate in the VUV-soft x-ray range, with peak brightness up to 10^{32} photons/s/mm^2/mrad^2 per 0.1% bandwidth, and average brightness 10^{25} photons/s/mm^2/mrad^2 per 0.1% bandwidth, to all beamlines simultaneously, dependent on pulse duration and wavelength.

X-ray FEL oscillators (XFELOs, or regenerative amplifier FELs—RAFELs) with pulse repetition rates on the order of 1-10 MHz may be driven either by ERLs, CW superconducting linacs, bunch trains from a pulsed linac, and in recirculating-linac configuration in which the final beam energy is achieved by multiple passes through a single linac. High bunch repetition rate is required to minimize the length of the optical cavity, in which the photon pulse interacts with the electron beam in an undulator on each pass through the cavity. The XFELO photon characteristics are complementary to high-gain FELs: the number of photons per pulse is smaller by a factor of 1000, the pulse length is larger by a factor of 10, and the bandwidth of the spectrum less by a factor of 10,000. As a result, the peak brightness is similar to the high-gain FEL. However, the pulse repetition rate of an XFELO is 1 MHz or higher. The wavelength of existing FEL oscillators is currently limited to the VUV range by the availability of optics for the mirrors having high-reflectivity at high incidence angle. Extending XFELO implementations to the x-ray region requires using Bragg-reflecting crystals. Methods to achieve tunability for these sources, requiring complex mechanical systems to adjust and hold stable the position and angle of multiple crystal mirrors, are being investigated.

Performance limiting factors for linac-driven FELs and XFELOs include the length of the linac and its cooling requirements, which limit beam energy; space charge limitations to brightness in the gun for high bunch charge; effects on the electron beam phase space, including induced modulations in the 10–100 nm length scale, from space charge and coherent synchrotron radiation; and instability from beam spreader components and ground motion.

A.4   High-Harmonic Generation (HHG)

Wavelengths in the hard ultraviolet or even the soft x-ray spectral region are attainable in the very high harmonics produced when an intense infrared laser pulse is focused into a gas. High-harmonic generation can be produced with temporal and spatial coherence properties similar to the driving laser field, and under special conditions, with sub-femtosecond pulse duration. Conversion efficiency is relatively low, and typically decreases with increasing harmonic order. HHG can provide a source in a single-investigator-sized laboratory, with output pulses synchronized to the driving laser and produced with the same repetition rate. Such sources have been generated using commercial driving lasers at the several-watt level, with repetition rates ranging from 10 Hz to 10 kHz. The cut-off of the harmonic spectrum extends to shorter wavelength as the drive laser intensity is increased, up to a saturation intensity where harmonic generation decreases. By using a gas species with a higher ionization potential, and a high-power longer-wavelength drive laser, together with novel phase-matching techniques in the harmonic-generation medium, the spectral cut-off of HHG may be extended into the hundreds of eV with conversion efficiency of the order of 10^{-6}. With the availability of kW average power lasers, average brightness of 10^{17} photons/s/mm^2/mrad^2 per 0.1% bandwidth may be achieved. HHG sources will be needed to provide the seeding power for FELs, which then act as amplifiers and additional stages of harmonic generation to generate intense soft x rays.

A.5   Advanced Accelerator Concepts

Advanced accelerator R&D is pursuing a variety of approaches to produce accelerating gradients orders of magnitude greater than conventional RF systems, enabling ultra-compact high-energy linear accelerators. Laser-driven plasma and dielectric-based acceleration techniques are being developed to provide electron beams to drive FELs and other light sources. In the same vein, electron beam driven dielectric and plasma-

based accelerators may provide similar high accelerating gradients and, with an RF-photo-injector, could produce drive beams for FELs useful for synchronized soft to very hard x rays. Successful development of these technologies would allow for the development of compact, ultra-bright, ultrafast radiation sources covering the entire spectral area from THz to x rays to gamma ray beams that are intrinsically synchronized to a laser pulse.

A.6   Other Sources

Other incoherent, lower brightness x-ray sources include smaller laboratory, or industrial-scale, devices. Laser-electron beam scattering (inverse Compton/Thomson) sources are under development to provide high-average-flux hard x-ray beams using a laser beam colliding with an electron beam from an electron storage ring or linear accelerator. These sources are envisioned for local applications extending from crystallography to diagnostic imaging. Laser-plasma sources producing x rays by electron acceleration within plasma channels are also under investigation as small laboratory sources.

A.7   X-Ray Source R&D Requirements

The new x-ray sources that will come on line in the next few years – including the PETRA-III and NSLS-II low-emittance storage rings and the LCLS, XFEL, FERMI, SPring-8 XFEL, and SPAR-X FEL facilities – will open a new window into the exploration of matter at the atomic and molecular level. These facilities, however, are only a step along the road of x-ray source and technology development that should provide even greater breakthrough opportunities for science. Future advances in electron sources, normal-conducting and superconducting accelerator and other RF structures, advanced manipulations of electron beams, laser systems, insertion devices, x-ray optics and instrumentation will enhance the performance and operational flexibility of soft and hard x-ray sources, while the development of high gradient RF, laser and plasma accelerators will make them more compact, less expensive and more widely available to scientists world wide. Developments in storage ring accelerator and beam line optics technologies will enable the realization of "ultimate" ring sources having very high brightness, high transverse coherence at angstrom wavelengths and the potential for partial lasing with the stored beam at soft x-ray wavelengths. And, just as atomic and molecular lasers have been developed over the years to reach new levels of power, short pulse length and other characteristics, in the same way short wavelength FELs can be developed to operate at wavelengths shorter than 1 Å, and with increased peak power, full transverse and longitudinal coherence, attosecond pulse lengths, and more.

To go from the conceptual level, to the proof of principle, to the implementation of new concepts, it is essential to have a strong research program and facilities dedicated to the development of advanced x-ray sources. Such facilities should have the manpower and financial support to make possible what is a sophisticated and challenging research task. Supporting such facilities should be a part of a general program of R&D for advanced x-ray sources.

X-ray facilities for research in atomic and molecular science – biology, chemistry, and physics – have already seen a tremendous development during the last decade, and can be further developed to new level of performance, thus allowing the acquisition of new knowledge with its associated economic, security, and societal benefits. The critical condition for this bright future to become reality is a strong R&D program for advanced x-ray sources, supported by the DOE and NSF and, in many cases, in collaboration with international laboratories, providing:

1. Competitive grants for scientists from universities and national laboratories.

2. Resources to attract new students and generate the manpower needed for the operation and development of the present and future generations of light sources.

3. Experimental facilities to test new concepts and develop the electron and x-ray beam instrumentation needed to work in the new regimes of ultra-short, and ultra-bright pulses, at photon energies from ~100 eV to 10 keV or higher.

4. Resources to support existing accelerator research staff at national labs to maintain existing critical knowledge base and technology skills needed for new facilities.

Several areas of x-ray source development are discussed in the following.

Electron guns: Continued R&D in high-brightness electron beam production will benefit future FEL and ERL performance, offering the prospect of higher brightness photon beams with increased coherence and at shorter wavelengths. Developments in high-repetition–rate photocathode, thermionic, and field-emitter electron guns are ongoing and involve normal-conducting and superconducting RF systems, as well as high-voltage DC systems, to produce high-brightness beams at MHz to GHz repetition rate and normalized emittances of 0.1-1 mm·mrad at bunch charges of 10 pC to 1 nC, and with high reliability and long lifetime. Extremely small normalized emittance (~0.1 mm·mrad) may be achieved with low bunch charge (~1-100 pC), and together with development of bunch compression techniques, this offers a route to high-brightness ultra-fast bunches for FEL and ERL applications.

Superconducting linacs: CW superconducting RF systems need further development for high-current applications in ERLs. Cavity HOM damping is required to control multipass beam breakup instabilities. Power lost to HOMs from a high-power ERL beam, and particle loss, must both be minimized to limit the load on the cryogenic system. The energy spread of the decelerated beam in an ERL will experience adiabatic antidamping, so the beam energy spread from impedances must be limited to avoid particle losses prior to reaching the beam dump.

High-gradient linacs: High gradient normal conducting RF systems need further development for application to compact x-ray light sources. Sample accelerator structures have been demonstrated with gradients as high as 150 MV/m, roughly 6 times higher than that in the European XFEL. Cavity HOM damping, required for controlling the multibunch beam breakup instabilities, and single-bunch wakefield control techniques both need to be demonstrated. Development of the high peak-power RF systems is also required. Advances in high ultra-high-power RF techniques and high-gradient RF structures hold the promise of developing compact and cost effective x-ray sources. DOE OHEP has historically supported this R&D, but additional targeted R&D is needed for x-ray source applications

Low-emittance beam transport, control, and manipulations: Understanding of the evolution of the electron beam emittance, from cathode to photon source, and means of its control and manipulation, are essential to achieving the highest brightness beams needed for future light sources. Increasingly sophisticated means of electron beam manipulations are on the near horizon, and promise enhanced capabilities and levels of performance for future light sources. Examples include transverse to longitudinal emittance exchange, bunch compression techniques, and beam conditioning for FEL applications. Not all of these techniques can be tested on existing accelerators, so we must develop many of the concepts, and validate the methodologies, with dedicated development accelerator(s) designed for the purpose.

Simulation and validation of accelerator and FEL design: Multi-physics computer modeling tools need to be developed to allow the full start-to-end simulations required for advanced facilities with high-brightness beams. X-ray FEL performance is extremely sensitive to the details of the electron beam phase-space distribution, and design using state-of-the–art simulation tools is required for the next generation of x-ray FELs. Simulations must be validated in experiments on existing and planned accelerators.

Laser systems: Developments in sophisticated laser systems with high average powers will be required for optimal performance of future light sources, benefiting rings, FELs, and ERLs. Applications are in high-brightness photocathode sources, seeding, slicing and optical manipulations, precision timing and synchronization systems, and end-station lasers. Short-pulse lasers in the micron wavelength range, extending average powers up to the kW level, together with novel phase-matching techniques in the harmonic-generation medium, give promise of future HHG sources extending into the hundreds of eV region with average soft x-ray power in the range of milliwatts. Development of means to control HHG laser system output phase and amplitude is required to enhance their applications in seeding FELs. Such advances in laser technology will have wide-ranging benefit for future light sources operating at high repetition rate.

Seeding and optical manipulation: Seeding by conventional lasers and harmonic generation in an FEL has been demonstrated at VUV wavelengths. However, demonstration of control of electron beams that are seeded at UV and shorter wavelengths, and their performance in harmonic generation at x-ray wavelengths, is needed. Development of control techniques to shape the x-ray pulse via phase and amplitude adjustments to the seed laser may provide significant enhancements to the capabilities of future FELs. Tests of optical manipulation technologies already proposed at a number of FEL facilities are required.

RF technology: High power RF transverse and longitudinal deflecting and accelerating structures can be used to create short or long bunches in storage rings (e.g., transverse deflecting "crab" cavities and high-harmonic longitudinal accelerating cavities) as well as to implement very fast kickers for on-axis bunch replacement injection to ultra-low emittance storage rings. While development of these components, together with efficient sources of very high RF power to drive them and the RF undulators mentioned below, has been supported in the past by the DOE's high-energy physics program, further targeted R&D is needed for x-ray source applications.

Laser- and electron-driven acceleration: Continued advanced accelerator R&D is required for development of high gradient laser-driven and electron beam-driven wakefield technology for light source applications. This technology would enable development of compact, ultra-bright, ultrafast radiation sources covering the entire spectrum from THz to gamma rays. Intrinsic pump–probe synchronization is achieved with laser-based accelerators and electron driven plasma-wakefield accelerators operating with a photocathode gun. The latter technology offers relatively high efficiency in converting wall plug power to beam power. FEL experiments with laser-plasma systems are now being built, and developments are required in further reduction of the electron beam energy spread and emittance, improvements in accelerator tuning and stability, and increasing the average brightness through use of higher-power laser systems. R&D to develop large-aperture dielectric structures (to enhance peak brightness), and experimental demonstration of acceleration gains of tens of MeV are required. Experimental facilities in which to test these and other advanced approaches to particle acceleration and x-ray production are required.

Insertion devices: Development of high performance insertion devices plays a key role in enhancing storage ring, ERL, and FEL brightness and photon energy range. Improvements in in-vacuum and

superconducting short-period undulators, and with control of polarization, are needed. As electron beams with smaller emttances and higher brightness are developed we will need undulators with smaller periods, and large gap–to–period ratio. Development of RF undulator technology to further reduce the period while maintaining a sufficient gap for beam transport, and efficient high power drive components, would enable polarization switching at kilohertz rates, and might push the undulator period into the mm range.

Photon optics and detectors: Existing photon optics, including mirrors, monochromators and lenses, as well as the detectors used for experiments, are insufficient to take full advantage of the high brightness, high power density, high coherence and temporal performance properties of future machines. Optics development will enable preserving and delivering the ultralow emittance and coherence of the photon beams from future sources to the user end stations. Development of x-ray optics analogs to the visible optics that have enabled the use of the unique properties of lasers is required to take full advantage of the next-generation x-ray sources. For example, soft and hard x-ray mirrors capable of reflecting high power beams at high incident angle will enable the configuration of x-ray FEL oscillators on ultra-low emittance accelerators, and mechanisms to precisely control the position and angle of the mirrors to maintain resonance and provide tunability will be needed. Efficient beam splitters will permit the development of delay lines. In some cases, new detector data acquisition and signal processing schemes will need to be developed to reduce sensitivity to residual small instabilities in beam position, angle, intensity, energy and time-of-arrival that are beyond the reach of beam and component stabilizing methods.

High stability mechanical systems: To reach the daunting stability goals for the extremely small photon beams from future x-ray sources, ultra-high stability mechanical designs for critical accelerator and beam line components will be needed in addition to stabilizing feedback and noise-rejecting detector data acquisition methods. These mechanical components include accelerator magnet support structures, beam position monitor assemblies, beam line mirrors, monochromators and other optical components, and experiment sample and detector assemblies. These components need to be stabilized against temperature variations, ground vibration, cooling water flow, electrical and magnetic influences and other "cultural" disturbances.

Instrumentation, control and timing technology: Paramount to the successful realization of any of the future accelerator and photon beam line technologies mentioned above is the concomitant need to develop the advanced instrumentation, control and femtosecond timing technologies they require. Ultra-low emittance storage ring, FEL, and ERL facilities will require very high precision beam position detectors and integrated stabilizing feedback systems in the accelerator and beam lines. High-current storage rings will require sophisticated control algorithms to manage the large transients in the RF cavity systems and the interplay between accelerating and bunch lengthening RF systems. Advanced wideband RF and beam feedback systems will be needed to control impedance-driven instabilities. Short-pulse FEL facilities require highly stable timing and synchronization systems. High-resolution beam diagnostics will be needed to monitor and control micron-sized electron beams, with picocoulomb current. In some situations, close interaction between accelerator and beam line component and detector instrumentation will be required. A continuing R&D program is needed for these and other instrumentation, control and timing systems.

APPENDIX B

Interactions of X-Rays with Matter:
Perturbative Limits and Mitigation Strategies

Today, the use of x rays for studying the electronic and atomic structure of matter is based on the premise that x rays provide information on the sample in the "as-prepared" state, which is either the electronic ground state at the temperature of interest or a state that has been prepared by application of an external stimulus like pressure or fields, or even an electronic excitation by a laser. Although x-ray-induced damage, for example of protein crystals, is sometimes a limiting effect, sample perturbations by the x-ray beam itself either are generally too weak to change the structure of the sample during the measurement or are handled within a linear response approximation, an example being the interpretation of angle-resolved photoemission data in terms of a one-electron spectral function.

With the advent of x-ray sources of unprecedented average and peak brightness, new questions arise as to the cross-over from the weak- to the strong-perturbation regime where the electronic, spin, or atomic state of the sample is no longer in the as-prepared state. In this section, we consider the transition from the weak- to the strong-perturbation regime and discuss methods for mitigating unwanted strong perturbations.

While the strong-perturbation regime can be considerable interest by itself, for example in AMO physics, it is anticipated that, in general, applications of x rays will continue to require weakly perturbing x-ray beams. In these cases the sample remains close to the as-prepared state during the measurement, which may record the results of a single shot or an average over many pulses.

In the following, we consider effects associated with a strong perturbation of the sample, and we shall refer to modifications from the as-prepared state as "damage." It is difficult at present to treat the various aspects of "damage" quantitatively. Furthermore, this complicated problem depends on many different parameters, such as source properties, optics, experimental arrangement, type of the sample, and prepared state of the sample.

It is clear from Figure 2.1 that in general we need to distinguish processes both within and between the thermodynamic reservoirs of atoms, electrons, and spins, and we expect the reservoirs to respond on different time scales. We expect metallic samples to behave differently than insulators and soft matter. Owing to efficient electron transport, metals can be "self-healing" and may exhibit a damage limit that is higher than, for example, soft matter and insulators (although this is not true with, for example, optical radiation). In soft matter, we may create a considerable fraction of broken bonds and radicals, and such radiation damage may prevent the determination of the desired properties of the as-prepared sample. In photoemission experiments, space-charge effects will distort the measured signal. In general, the question of "damage" therefore needs to be considered on a case-by-case basis and depends on many parameters, such as the photon energy, the number of photons per pulse, the pulse duration, the repetition rate, the beam area, the interaction cross-section, the electron mean free paths in the sample, and the thermal diffusion coefficient.

In order to develop a better understanding of the various aspects of "damage", we consider below the example of a ferromagnetic metal where all three thermodynamic reservoirs—the lattice, the electrons and the spins—need to be considered. Also, considerable information has been derived from previous optical-laser experiments.

B.1   Case Study: X-Ray Perturbation of a Ferromagnetic Metal

Under the influence of the x-ray electric field with intensity ~E^2, electrons are excited on the timescale of attoseconds into previously unoccupied electronic states. This photoelectric or x-ray absorption process has by far the largest cross-section, as illustrated in Figure B.1 for Fe metal, and triggers various processes that can modify the sample and lead to "damage." Figure B.1 shows that x-ray absorption becomes significantly enhanced around absorption edges and increases with decreasing photon energy. Thus x-ray "damage" is most severe around soft x-ray absorption edges.

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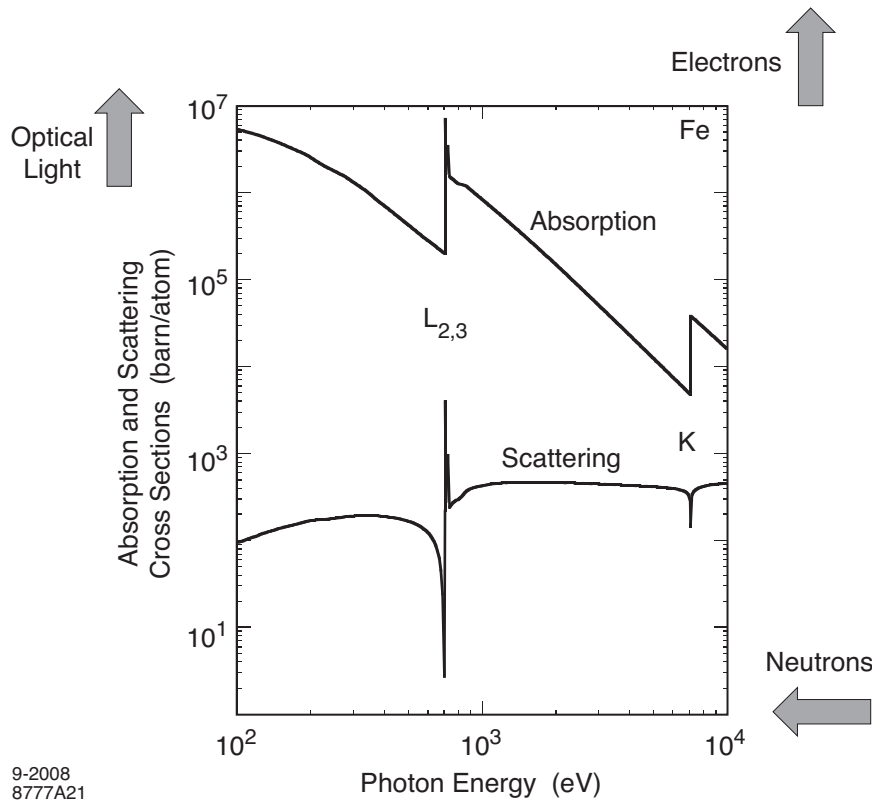
Figure B.1. Energy dependence of the x-ray absorption (photoelectric) and x-ray elastic scattering cross-sections for Fe metal in the 100-eV to 10-keV range. Note the resonant enhancements near absorption edges. For comparison, we indicate next to the graph the approximate range of cross-sections for optical photons and electrons, and the better defined cross section for neutrons.

1.   Response of the Metal on Femtosecond Timescales

For FEL pulses with lengths of tens of femtoseconds, one needs to consider what happens to the electronic system in the presence of the x-ray pulse, since the signal from the end of the pulse may be modified by changes caused at the beginning of the pulse. This question is of particular interest for soft x-ray experiments that are preferentially sensitive to the electronic instead of the crystallographic structure. Furthermore, x-ray cross-sections are large in the soft x-ray region and electronic perturbations will

therefore be most severe. However, other effects may be seen at higher photon energy, where the absorbed energy per absorbed photon is higher.

In the following, we shall consider an experiment at the Fe L-edge at ~710 eV. We neglect linear effects in **E**, such as field distortion and ionization, since they are reduced at x-ray energies because of the high frequency of the fields. We also ignore all effects in the sample arising from the electromagnetic **B** field since they are negligibly small (cross section smaller by a factor of $< 10^{-4}$ of the electric cross-section). Our treatment is restricted to effects following electronic dipole transitions, and within this model we can describe the incoming x rays by four parameters, the number of photons N, the beam cross-section of area A, the photon energy $\hbar\omega$, and the pulse length τ. The quantity $N\hbar\omega/A\tau$ thus defines the incident peak-power density.

Nearly all photons get absorbed in the sample over a length $d = 2/\sigma\rho$, corresponding to two photon absorption lengths, where σ is the atomic absorption cross-section (plotted in Figure B.1) and ρ the atomic density. Thus the x-ray energy $N\hbar\omega$ contained in N photons is absorbed within a time τ through electronic excitations in the volume Ad containing ρAd atoms. The deposited energy per atom per pulse is thus given by

$$U = N\hbar\omega\,\sigma\,/\,2\,A$$

The electronic excitations occur on individual atoms. For resonant L-edge absorption in Fe at ~710 eV, the excitation leads to electrons at energies up to a few eV above the Fermi level and holes in the Fe 2p core shell. Using values for the L-edge excitation of Fe metal, each atom in the absorption volume gets excited (deposition of ~710 eV per atom) for an incident flux of about 3×10^9 photons/pulse/μm^2.

The excited electronic states decay on timescales of attoseconds to femtoseconds through Auger decay, radiative decay and electron–electron scattering and, in general, decay and scattering times become shorter with increasing excited state energy. Electrons with kinetic energies larger than the vacuum level E_V may leave the sample as photoelectrons, Auger electrons, and their secondaries. Because the quantum efficiencies of solid samples are less than a few percent and the fluorescence yields in the soft x-ray region are very small, one can safely assume that only a small fraction (< 10%) of energy leaves the sample. Through repeated inelastic scattering, higher energy electrons produce secondaries, the excitation volume spreads (speeds > nm/fs) beyond the atomic volume, and the secondaries typically end up with a kinetic energy $< E_V$, so that they are trapped by the surface potential barrier (work function). Initially the excited electron gas does not have a well defined temperature which is established only on a timescale of about 100 fs through multiple electron-electron scattering leading to equilibration into a Fermi-Dirac distribution.

The description of the excited electronic state during the pulse is complicated. Optical laser experiments can be used to derive an approximate "damage" threshold on the timescale >100 fs, where the electron gas has equilibrated. In fact, the clearest information comes from the type of experiment illustrated in Figure 3.10c, which has been the subject of extensive work over the last ten years. In such pump–probe experiments on magnetic metals, changes in the electronic state clearly manifest themselves on the 100 fs timescale in changes of the magnetization measured by a delayed weaker probe pulse as a function of the incident power of the pump pulse. For both optical and x-ray excitations, the absorbed photons lead to a "hot" electron distribution extending from the Fermi level to the vacuum level, and one may therefore

infer electronic modification thresholds for x-ray experiments from those observed for optical lasers of comparable pulse lengths by comparison of the deposited energy per volume and time.

As shown in Figure 3.10a, it is known that at room temperature the magnetization changes on the time scale of ~100 fs when a ferromagnet like Ni is excited with optical photons of 2-eV energy, <100-fs pulse length and fluences larger than ~ 1 mJ/cm^2. Since the photon absorption cross-section and absorption length in Fe of 2 eV and 710 eV photons is about the same (~20 nm), we can take this fluence as our electronic modification threshold, and we obtain for Fe L-edge excitation a threshold of about 10^5 photons/pulse/µm^2. This indicates a potential limit in the use of focused beams produced by soft x-ray lasers.

2. Response of the Metal on Picosecond Timescales

On longer timescales, the excited electron gas can equilibrate through heat exchange with the phonon bath, and this process will heat up the lattice. While typical electron-phonon scattering times at room temperature are tens of femtoseconds, the energy transfer from the hot electrons to the lattice takes much longer, owing to the large difference in the specific heats of the electron gas and the phonons. The equilibration between the two reservoirs may take picoseconds to tens of picoseconds and may even be bottlenecked. Because of the different electronic and phonon timescales in solids, it is therefore possible to distinguish the effect of the x rays on the electronic and spin systems from that on the lattice.

A rough estimate of a "damage" limit can be made by simply considering the deposited energy per pulse per atom. In the weak perturbation regime, the deposited energy would have to remain significantly below a typical bond energy of 2 eV/atom. Using the Fe L-edge cross-section 7 Mb = $7x10^{-18}$ cm^2/atom at $\hbar\omega = 710$eV, a deposited energy per pulse of 1% of this bond energy corresponds to about 10^5 photons/pulse/µm^2. This number has the same order of magnitude as that derived earlier for electronic modification.

One may calculate the increase in lattice temperature using the Dulong-Petit value $3k_B$ of the specific heat capacity per atom. In the absence of heat conduction, the increase in lattice temperature ΔT is then related to the number of photons per pulse N according to

$$\Delta T \simeq \frac{N \hbar\omega \sigma}{6 A k_B}$$

For the same parameters used above, the number of incident photons would therefore need to be limited to about $10^3 \Delta T$ photons/pulse/K/µm^2, where ΔT is the tolerable temperature increase. Thus an allowed temperature increase of 100 K would again yield a limiting value of 10^5 photons/pulse/µm^2. Note that this number corresponds to a single pulse and does not take into consideration the repetition rate. On the other hand it assumes no heat conduction out of the volume heated by the x-ray beam. In practice, heat conduction partially compensates for repetition rate effects. The effect of heat conduction is discussed more in Section B.3 below.

These considerations are consistent with the results of early experiments carried out with the FLASH soft x-ray/VUV FEL at considerably higher photon fluences. When a solid sample is subjected to a single unattenuated, nominally 10-fs-long, FEL pulse, a scattering pattern is obtained from the sample, yet after the pulse, the sample is destroyed by heating the surface to plasma temperatures. Although the spatial

resolution of present experiments is limited by the available long wavelength the observations are consistent with the notion that modification of the atomic structure of the sample happens on time scales longer than 10 fs.

B.2   Space-Charge Effects: Photoemission

The above estimates do not take into consideration the detection of the signal. In photon-in/electron-out experiments, space-charge effects determine the maximum fluence. Such effects become increasingly important when the required energy resolution of the emitted electrons increases.

The first core-level photoemission experiments at FLASH at photon energies of ~120 eV, pulse lengths ~ 30 fs, and incident photon energy densities up to 5×10^{10} W/cm^2 have revealed such effects. Significant broadening and shifting of the W 4f photoemission lines was observed to set in around 3×10^7 photons/pulse for a photon spot ~ 300×300 μm^2.

In angle-resolved photoemission experiments on correlated and superconducting materials where very high energy resolution is required, the space-charge effects are even stronger and restrict measurements to considerably lower fluxes. Space-charge effects stem from the Coulomb interaction between electrons created within a pulse (for a low-repetition-rate source) or between secondary electrons from adjacent pulses (for a high-repetition-rate source). In general, a high-repetition-rate source has the advantage over a low-repetition-rate source, since it allows a reduction in the number of photons per pulse while maintaining the same average flux.

B.3   Possible Remedies for Damage

As occurs in protein crystallography and cryo-TEM, we expect that in some cases damage issues may be reduced by special effects or overcome by experimental procedures. In some cases, analysis techniques may be developed to correct for them. Damage may be reduced by simply changing the photon energy, as evident from the strongly changing absorption cross-section in Figure B.1. For example, in resonant coherent imaging (speckle) studies one may tune the photon energy to just below the absorption edge where the absorptive (imaginary part) of the scattering cross-section is minimized but the dispersive (real part) is resonantly enhanced. Such "phase images" offer comparable contrast to "absorption images."

Other experimental remedies consist of spreading the beam over a larger area, e.g., by using a grazing-incidence geometry on the target. Brightness-hungry experiments are often performed under conditions where the beamline optics filters out much of the flux, as when one requires a very high-resolution monochromator or when zone-plate focusing optics are used. In these cases the source-produced intensity is naturally reduced. Other examples involve tenuous samples where absorption is low, as with low-pressure gases and aerosols. In some cases, one may overcome the damage problem by continuously refreshing the sample using micro- or nano-fluidics, jets, droplets, or by scanning a solid sample or surface.

We can envision two other novel effects, as yet unproven, that may provide a way out of "damage". One is operative on ultrashort time scales, the other for nanosized beams.

As discussed in Section B.1, the electronic state of a sample is ill defined on ultrashort excitation time scales (e.g. less than 100 fs for a metal). One could therefore envision that on such ultrashort timescales, i.e. for pulses that are of order of 1 fs, the electronic structure remains frozen in the "as prepared" state during the measurement. Since a new (modified = "damaged") electronic structure becomes uniquely defined only after an electronic equilibration time of about 100 fs, one may be able to avoid "electronic damage" with ultrafast pulses. Only at a delay time of a few hundred femtoseconds would a second pulse then see the modified electronic state and at delay times of picoseconds it would probe the atomically excited sample, which may even have been destroyed.

Another effect may prevent physical "damage" of a sample when the illuminated sample volume becomes very small, e.g. for beams with lateral sizes smaller than a micrometer. In our earlier discussion of "damage", we have considered only the energy deposited in the sample volume illuminated by the beam but not the heat flow out of the volume. In general, this assumption overestimates "damage" effects. Furthermore, we have used the simple metric of constant fluence or energy per unit area to determine damage limits. As the area decreases, this metric leads to a drastic reduction in the tolerable number of incident photons per pulse. In practice, in this way we may vastly overestimate damage effects. We know that electron transport in macroscopic structures like wires is limited to certain current densities because of heating and the ultimate melting of the wire. If the macroscopic rules were applied to nanowires, much of our present technology would not be possible. For example, a macroscopic Cu wire of 1-mm diameter has a current density survival limit of about 10^3 A/cm^2. In metallic Cu nano-pillars of ~100-nm diameter one can achieve current densities 10^8 A/cm^2 without destroying the device. This increase of five orders of magnitude is due to the significantly increased surface-to-volume ratio and the resulting high cooling rate through the surface of the nano-region. We expect similar "cooling" effects when the x-ray beam size is reduced, and hence the physical damage limit would be significantly increased. We note, however, that such thermal equilibration proceeds on the slower pico- to nanosecond time scales.

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