SCIENCE WITH LIGHT AND NEUTRON SOURCES

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Abstract

In this brief review, we shall discuss the evolution of the partnership between particle accelerator physicists and condensed matter physicists, crystallographers, materials scientists, chemists and biologists in developing our current X-ray synchrotron and spallation neutron source facilities for carrying out research in a wide variety of areas of scientific research, which have resulted in several discoveries of importance to society.

INTRODUCTION

The original reasons for the development of particle accelerators, which were to carry out research in nuclear physics and elementary particle physics, have resulted in unexpected benefits to other areas of science. In addition to unlocking the secrets of the atom and attempting to probe the fundamental laws that underpin our physical universe, particle accelerators are now also being used for research as diverse as probing the structure and function of the molecules that constitute all living things, or understanding the behavior of atoms and electrons in crystals as they behave collectively in highly correlated ways to imbue materials with exotic properties such as superconductivity, ferromagnetism, ferroelectricity, etc. As is well-known, the reason for this is the ability of particle accelerators to provide intense beams of neutrons in one case, or beams of x-ray photons (both hard and soft) in the other, which provide some of the most advanced probes for studying condensed matter. The former has led to the existence of dedicated spallation neutron sources [1,2],which exist or are being planned in several technologically advanced countries, and the latter to the existence of an even larger number of synchrotron radiation facilities in many countries around the world. While the fields of neutron scattering and x-ray diffraction and scattering and even x-ray photoemission spectroscopy were all developed without accelerator technology, it has been the harnessing of the power of modern particle accelerators which has enabled both these fields to make quantum jumps in their available fluxes and capabilities. It is now widely recognized that the presence of accelerator-driven x-ray and neutron facilities is an essential requirement for progress in the fields of biology, chemistry, physics, environmental science and energy-related research.

Neutron and synchrotron X-ray sources have provided basic information on the structure of biological macromolecules such as proteins which have led to major advances in the development of drugs to battle infection and illnesses such as HIV and cancer. They have also provided basic understanding of magnetic distributions and switching in magnetic thin films and other nanostructures which can lead to advances in magnetic information storage and retrieval: they are providing information on the structure of polymer coatings of importance to the packaging and medical implant industries, and interfaces in materials of importance to the technology of energy storage, photovoltaics, fuel cells, etc. While examples will be given in my oral presentation, they are too many to describe in detail within the scope of the present article, so I shall restrict myself here to some general remarks and a brief description of some of the most recent developments.

The ultimate dream of scientists has always been to develop the capability of "seeing" atoms and molecules at sub-nanoscopic length and time scales to understand what gives materials their properties. Neutrons and X-rays have been used in various ways to obtain microscopic information about materials over a wide range of length scales and time scales, although usually indirectly by providing information in wave vector and energy space. Scattering experiments can be characterized as measuring the characteristic scattering function $S(Q, \omega)$ of a material (where Q is the wavevector transfer, and $\hbar\omega$ the energy transfer). By the Fluctuation Dissipation Theorem in Statistical Mechanics, this is directly related to the poles of its response function to various external perturbations (magnetic or non-magnetic) and thus yields its natural resonant frequencies, ie. its excitation spectrum. The corresponding energy-integrated function $S(Q)$, or in particular its elastic component yields diffraction and the structure of the material. Both neutrons and x-rays can be scattered from condensed matter over wide ranges of energy and momentum transfers to deduce structure on length scales from nanometers to microns and dynamics from seconds down to femtoseconds with current sources. The intensity of modern sources has also enabled experiments on scattering from surfaces and interfaces of materials using techniques such as reflectivity, and grazing incidence scattering which can be used to obtain structural details about these interfaces, including magnetic distributions in thin films.

In addition X-ray spectroscopies such as photoemission and absorption spectroscopy can be used to study the detailed electronic structures of atoms, molecules and solids. With high brilliance synchrotron sources now providing coherent beams of X-ray photons of significant intensity, it is also possible to do imaging down to the nanometer level.

NEUTRONS

Initially, the existence of nuclear reactors that proliferated after World War II and the development of the theory of thermal neutron scattering led to the development of research reactors, which provided beams of neutrons for basic research purposes. Early triumphs of neutron scattering included the demonstration and investigation of antiferromagnetic spin structures in a variety of solids; the demonstration and measurements of phonon and magnon excitations and their dispersion curves in energy-wave vector space;(the Nobel Prize in Physics in 1994 was awarded jointly to C.G.Shull and B.N. Brockhouse for the above 2 achievements); the existence of rotons in superfluid helium; the proof of the scaling theory of polymers (which, among other things, earned P.G. de Gennes the 1991 Nobel Prize); the measurements of residual stress in engineered materials, and several others. Eventually, the demand for ever higher neutron fluxes, and the political liabilities, security issues and costs associated with constructing super-high-flux reactors using highly enriched U fuel rods, led scientists to look into accelerator-based sources of neutron beams. Spallation of heavy nuclei, such as Bi, Pb W or Hg, by high-energy proton beams, turns out to be a more efficient process energy-wise for producing neutrons than fission. The challenge of producing pulsed (or in some cases, steady state) high-energy (~ 100 MeV to 1 GeV) high-current proton beams, delivering up to 2 MW of power onto adequately cooled heavy metal targets, and suitable moderators for producing tailored pulsed neutron beams was mastered in the U.S.A., Japan, and Europe, all of which now have pulsed spallation neutron sources (the SNS and LANSCE in the U.S., the ISIS and SINQ facilities and the planned European Spallation Source in Europe, and the J-PARC facility in Japan) dedicated to research in materials and other areas of science. Neutron scattering and spectroscopy with pulsed sources relies on the time-of-flight technique and with the advent of sophisticated neutron instrumentation and software, this turns out to be a very efficient and versatile method of collecting data. Thus these sources are regarded as the path to neutron sources for the future.

In spite of their relatively low brilliance (compared to synchrotron X-ray sources), there are certain problems that neutrons are well suited for, such as the investigation of soft matter, such as polymers, complex fluids and biological samples, since they do not damage the sample and the vastly different scattering lengths of H and D allow selective contrast matching, which turns out to be extremely useful for looking at macromolecules in solution. Because of their spin, they also interact strongly with magnetic moments in solids and can thus be useful in studying the magnetic structure and dynamics of solids, and because their energies and wavelengths match those of most elementary excitations in typical solids rather well, they can be used for inelastic scattering studies of such excitations with high resolution in energy and momentum space.

Neutron scattering has played a key role in our attempts to understand the mechanisms underlying superconductivity in the new high-temperature superconductors, such as the cuprates or the iron-based family of superconductors. It has done this by demonstrating the antiferromagnetic structure of the spins on the Cu or Fe atoms in these materials and measuring the spectrum of spin fluctuations which are believed to be strongly correlated with the superconductivity. On more application-oriented levels, neutron diffraction is being used to study the in-situ evolution of the structure of battery materials during operation, and the spatial distribution of residual stresses in materials developed under new engineering processes for producing lightweight materials for the transportation industry.

X-RAYS

The use of X-rays using keV tube sources became rapidly prevalent soon after Roentgen’s original discovery and von Laue and Bragg’s theoretical treatment and demonstration of X-ray diffraction. Synchrotron Radiation from a particle accelerator was first observed and recognized as such in 1947 at an electron accelerator built at GE. Its potential for use in research was recognized quickly, and the development of storage rings on accelerators provided a major advance in the production of usable synchrotron radiation. Initially synchrotron uv and x-ray research was carried out parasitically at machines primarily dedicated to High Energy Physics, but gradually the demand grew for dedicated storage ring synchrotron X-ray facilities, which have evolved into the large modern user facilities of today such as the APS, the ALS, the SSRL, and the currently under construction NSLS-II in the USA and similar sources in Europe and Japan, as well as other countries. Over a dozen Nobel Prizes have been given for work related to X-Rays, including recent prizes in Medicine and Chemistry for solving biological macromolecular structures which Synchrotron X-Rays have enabled.

With current 3rd generation synchrotron sources, it is possible to solve the structure of crystallizable proteins to down to 2 Å, by studying thousands of Bragg reflections and using tricks such as Multiple Anomalous Diffraction (MAD) to obtain phase information. X-Ray diffraction yields the intensities but unfortunately not the phase information about the Fourier Transform of the electron density of a crystal at each reciprocal lattice point. Thus X-Ray crystallographic techniques cannot be used for non-crystallizable proteins. To get around this problem, one can use holographic techniques or so-called phase retrieval methods. Holographic imaging of magnetic domains in thin film samples at the several nm length scale using resonant X-ray magnetic scattering have been demonstrated, and such techniques are being actively developed at third generation synchrotron sources as well as XFEL sources. This method in practice is difficult to apply to get structural information in the sub-nanometer range.

Phase retrieval methods have been developed over the last few years (using algorithms taken over from optical imaging) for measuring the scattering from a non-crystalline sample at enough points in reciprocal space that one obtains an “oversampling” from which both amplitude and phase information can be obtained, using iterative techniques [3,4]. So far, such methods have yielded structures down to tens of Angstroms , but there is no intrinsic limit why they could not be extended down to smaller length scales, provided one has enough intensity in the form of x-ray beams that are coherent across the sample. Unfortunately while such intensities are available from the X-ray Free Electron Laser Sources, they destroy the sample very rapidly by creating a "Coulomb explosion" which blows the molecules apart. The current strategy is to let intense, coherent, femtosecond X-ray pulses from a source such as the Linear Coherent Light Source (LCLS) impinge on a protein sample, so that the scattered X-rays reveal the scattering pattern before the molecule is destroyed. Computer simulations of such a process indicate [5] that this should be feasible with X-ray pulses of widths < 10 fs.

The availability of such intense short pulses in the X-ray regime also allows one to do pump-probe experiments to investigate the behavior of atoms under extremely high electromagnetic fields. Thus recent experiments at LCLS [6] have demonstrated the existence of 6-photon, 10-electron processes, which essentially strip a Neon atom of its inner shell electrons, producing a so-called "hollow atom" with corresponding intensity-induced transparency. Pump-probe experiments are planned which should study the reaction of matter to extreme conditions of pressure and temperature by probing structure and spectroscopy after being exposed to intense short laser-generated pulses.

The equilibrium dynamics of ordinary soft condensed matter is currently being probed via a technique called X-ray Photon Correlation Spectroscopy (XPCS) which looks at the time auto correlation function of the speckle pattern produced by a coherent beam of x-rays impinging on the sample.

Such techniques have been successful for studying the slow dynamics of soft condensed matter on time scales ranging from microseconds to seconds (too slow to study with inelastic scattering). The missing time scales between inelastic scattering and real-time XPCS will soon be capable of being probed at LCLS using delay lines to allow coherent X-ray pulses to impinge on the sample at nanosecond to picosecond time scales.

Thus the last 3 decades have seen a close symbiotic partnership between accelerator physicists, and primarily condensed matter physicists and crystallographers in developing and running synchrotron X-ray and neutron sources. This partnership has not always been smooth sailing, but there is no doubt that it has been extremely productive, resulting in user facilities that serve thousands of users per year working in diverse fields, and resulting in discoveries of importance for drug development, energy storage, magnetic information storage and retrieval and many other technologies which benefit our society.

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