An Assessment of the National Need for Facilities Dedicated to the Production of Synchrotron Radiation (1976)

Pages
85

Size
5 x 8

ISBN
0309346568

Panel to Assess the National Need for Facilities Dedicated to the Production of Synchrotron Radiation; Solid State Sciences Committee; Assembly of Mathematical and Physical Sciences; National Research Council

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Dedicated to the Production of Synchrotron Radiation
Solid State Sciences Committee
Assembly of Mathematical and Physical Sciences
National Research Council

NATIONAL ACADEMY OF SCIENCES
Washington, D. C. 1976

NOTICE

The project that is the subject of this report was approved by the Governing Board of the National Research Council, whose members are drawn from the Councils of the National Academy of Sciences, the National Academy of Engineering, and the Institute of Medicine. The members of the Committee responsible for the report were chosen for their special competences and with regard for appropriate balance.

This report has been reviewed by a group other than the authors according to procedures approved by a Report Review Committee consisting of members of the National Academy of Sciences, the National Academy of Engineering, and the Institute of Medicine.

The Solid State Sciences Committee is pleased to acknowledge the support of the National Science Foundation and the Energy Research and Development Administration for the conduct of this study.

Available from
Solid State Sciences Committee
National Research Council
2101 Constitution Avenue
Washington, D.C. 20418

PANEL TO ASSESS THE NATIONAL NEED FOR FACILITIES DEDICATED TO THE PRODUCTION OF SYNCHROTRON RADIATION

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PREFACE

Intense electromagnetic radiation is an inevitable by-product of the acceleration of relativistic electrons in the storage rings used in elementary-particle research. Although this synchrotron radiation is only a nuisance to the high-energy physicist, it can be a powerful tool for many other scientists if proper access to the radiation can be provided. Until recently, few scientists have had access to such radiation, and so the great promise of synchrotron radiation as a research tool is only now being generally recognized.

This study was initiated by the Solid State Sciences Committee (SSSC) because of a general concern that development of synchrotron radiation facilities in this country had not kept pace with the research opportunities such radiation offered. Consequently, in January 1976 the SSSC proposed to the Executive Committee of the Assembly of Mathematical and Physical Sciences that a broad assessment of the need for synchrotron radiation facilities in this country be undertaken. A panel of the SSSC was appointed in April to carry out this task. Because the SSSC viewed the need for action as pressing, it asked the Assessment Panel to complete its work as quickly as possible--preferably by the summer of 1976.

In addition to a most demanding schedule, the planners of this study faced other problems. One difficulty concerned the charge to the Panel--to focus its assessment on a technique rather than a field. The scientific users of synchrotron radiation do not belong to a pre-existing and clearly defined group. Indeed, most physical and biological scientists, as well as applied scientists, may be affected in the future by the use of synchrotron radiation, and so the group formed for the assessment had to represent several scientific disciplines.

The time schedule and the newness of the field presented an even more difficult problem. An informed assessment could be made only with the direct participation of scientists now involved in using synchrotron radiation. Information inevitably had to come from enthusiasts and advocates. The planners of this study compensated for this by including in the membership of the Panel scientists who were not directly involved in the issues and by forming an Advisory Group of senior scientists who could ensure that the Panel's judgments were given a broad and realistic perspective.

Finally, my own selection as Chairman of the Assessment Panel was due to the desire of the SSSC that the Chairmanship be in neutral hands. Although I have remained neutral, I hope that the reader of this report does sense genuine enthusiasm in it for the future promise of synchrotron radiation. Certainly this enthusiasm is one that I now share even though my professional interests are now far afield.

The Panel's objective was to assess the overall national need for facilities devoted to the production of synchrotron radiation. The time period considered extended over the next ten years. The Panel undertook to estimate the overall quantitative need as well as the qualitative characteristics that should be available within the total mix of facilities. No attempt to recommend a specific inventory of facilities and their characteristics was made. Not only would this have been a highly speculative and controversial undertaking because of the limited evidence available, but also there is no unique answer--the general needs can be met by any of a number of alternatives. In any case, from the outset it was not the intent of the Panel to answer questions concerning specific facilities, sites, or proposals.

In assisting the reader in interpreting the Panel's recommendations, I would say that most of us would visualize the need for facilities recommended here as being satisfied by three geographically distributed major facilities, at least two of which would have x-ray capability. All three, of course, would have XUV capability, as might other smaller facilities. Specific choices will have to be made after a critical review of concrete proposals. Potential users should play a role in such decisions.

This report is but one step in what I hope will be a continuing effort to develop a national program to exploit the powerful tool of synchrotron radiation. The Panel recognizes that there are important tasks in setting up such

a program that we have not adequately addressed. One such task is the definition of the management structure of national facilities and the role of scientific users in that management. Certain points are clear. National facilities require a management framework that reflects national scientific interests and not local ones. Also, it is not too early to think about means to involve outside scientific users in the planning of new facilities. On this issue, I would emphasize that the problems are *not* entirely analogous to those confronting the high-energy physics community. Synchrotron radiation user groups will come from widely differing fields of basic and applied science. Most groups will be small, and for many the involvement with synchrotron radiation will only be occasional. Thus the interaction between the facility and the outside user is complex and will require particularly sensitive attention if synchrotron radiation is to have the broad impact on science that this report anticipates.

The question of future funding also deserves comment. The Panel has estimated both the capital and operating costs of the facilities themselves but has made no attempt to estimate the cost of the science involved. There are several reasons for this. The funding of most of the science performed at such facilities would be the responsibility of the using scientist; since most such scientists would be using these facilities only part time to meet external scientific objectives, it is not apparent what part of the scientific costs (if any) should be associated with the facilities program. Moreover, it is quite clear that the new science that will be associated with new radiation facilities will not necessarily require commensurate increases to future science budgets. One would expect that much of the future science will evolve from the reorientation of current activities. In short, an attempt *by the Panel* to estimate the science budgets associated with new facilities would have to be based on so many arbitrary assumptions that it would have no significance.

Finally, let me express my personal appreciation to all the participants in this study for their hard and conscientious work. I share their hope that this report will provide the foundation for the development of a national program that can meet the scientific promise that the Panel describes in this report.

Robert W. Morse, *Chairman*
Panel to Assess the National Need
for Facilities Dedicated to the
Production of Synchrotron Radiation

1
INTRODUCTION

The radiation emitted by relativistic electrons subjected to acceleration transverse to their motion is called synchrotron radiation. Electron storage rings are particularly useful sources of such radiation. The attributes of synchrotron radiation from storage rings include continuous spectral distribution from the far infrared to the x-ray region, high intensity and brightness, high stability with well-determined flux, high degree of polarization, subnanosecond pulse structure, and production of radiation in an ultra-high vacuum. As an indication of the unrivaled intensity of such sources, the 4-GeV storage ring at the Stanford Linear Accelerator emits in normal operation a total radiation flux of about 100,000 W, whereas the most powerful available x-ray tube emits a total radiation flux of about 10 W.

The use of synchrotron radiation as a research tool in biology, chemistry, and physics is rapidly growing. Since its initial utilization in the United States at Cornell University in 1956 followed by ongoing work beginning in 1963 at the National Bureau of Standards, two national facilities have been established--the Stanford Synchrotron Radiation Project (SSRP) and the Synchrotron Radiation Center (SRC) at the University of Wisconsin. Programs here and abroad have demonstrated considerable richness in the scientific applications of this unique and powerful source of electromagnetic radiation. The initial demonstration of the scientific importance of synchrotron radiation has created a greatly increased user demand on the existing facilities. In view of this rising demand, questions have arisen concerning the long-range impact of this new radiation source and the facilities that will be necessary to utilize its potential effectively.

Approximately five years are required to construct any new synchrotron radiation facility and to put it into operation. This requirement, together with rapidly increasing scientific importance and concomitant increased user interest, makes it critical that decisions be made on the steps that should be taken to provide the United States with adequate facilities to meet the challenges of this new field now and in the 1980's. The objective of this study is to provide part of the basis for those decisions.

At the foundation of this assessment is the scientific and technological impact of synchrotron radiation described in Chapter 3. A broad and significant impact is documented, which strongly suggests that many important frontiers in physics, biology, chemistry, materials science, and technology have been and will be further advanced by synchrotron radiation research. An estimate of the present and future utilization of synchrotron radiation has also been made and is included in Chapter 4. It is shown in these two chapters that current U.S. synchrotron facilities are not adequate to meet the needs of this field. In an effort to lay the groundwork for decisions about existing and future storage-ring sources, a review of the capabilities, design, and operation of storage rings was undertaken in Chapter 5. Special attention was paid not only to the most advanced storage-ring technology but also to matching the properties of such rings to the instrumentation that will be employed in scientific and technological investigations using synchrotron radiation.

The conclusions and recommendations of this study are intended to provide a basis for a ten-year plan that will provide synchrotron facilities that are powerful, flexible, and well matched to their intended task, thereby fulfilling the scientific and technological promise of synchrotron radiation.

2
CONCLUSIONS AND RECOMMENDATIONS

CONCLUSIONS

■ The scientific and technological impact of synchrotron radiation research has enormous breadth, spanning many disciplines and encompassing many techniques. The most promising experimental techniques at the moment are x-ray diffraction, photoelectron spectroscopy, and absorption studies throughout the ultraviolet and x-ray spectral range. These techniques will advance the frontiers of knowledge in a number of fields. In surface science, basic information will be obtained concerning the structural and electronic properties of surfaces and heterogeneous catalysis. Techniques of structural analysis (x-ray diffraction and x-ray absorption) should provide insight into the organization and function of complex systems of importance in chemical and biological processes such as nitrogen fixation, photosynthesis, and the oxygenation of blood. Our understanding of liquid and polymer-bound catalytic reactions should also be improved. Various techniques will contribute to our knowledge of the properties of materials under a variety of chemical and physical conditions. For example, new insights into the effects of radiation and defects on the strength of materials can be expected.

Improved understanding of catalytic reactions, nitrogen fixation, and radiation damage can easily be seen to have a significant impact on energy-related technology. Elucidation of the functioning of complex biological systems and the effect of impurities on them will play a significant role in medical- and environmental-related technologies. Finally, x-ray lithography should directly affect the development and manufacture of microminiature electronic devices, which will be a cornerstone of the technological society of the future.

■ The use of synchrotron radiation is growing rapidly, with concentrations of active participants centered around existing facilities. It is difficult to foresee the future because of the rapidity of this growth and also because x-ray synchrotron radiation has been available only recently. A conservative analysis of future requirements based on scientific potential and user interest leads to the conclusion that by 1986 there will be a need for approximately 60 x-ray stations serving users requiring radiation in the range 1-50 keV (12-0.25 Å) and 40 soft x-ray/vacuum ultraviolet (XUV) stations serving users requiring radiation in the range 0.01-1.0 keV (1200-12 Å). Currently, there are 7 x-ray and 17 XUV stations. Existing facilities, even if fully expanded and dedicated, would only meet one third to one half of the increased need (measured in terms of stations only) conservatively estimated to be required by 1986.

■ A storage ring has never been built in the United States explicitly to optimize synchrotron radiation experiments. Yet the experience gained in building storage rings for high-energy physics research leads the Panel to conclude that the physics, engineering, and technological expertise necessary to create optimized storage rings does exist. These sources could provide, in addition to more stations, at least one to two orders-of-magnitude enhancement in usable synchrotron light intensity compared with existing facilities as currently operated. New developments such as the helical wiggler (a special magnetic field device to generate short-wavelength radiation) may offer an even greater enhancement in radiation intensity.

■ A lead time of approximately five years is necessary to create new synchrotron radiation capabilities. The importance of the scientific impact, reflected by the advanced status of foreign efforts, leads the Panel to conclude that action is required now to increase U.S. synchrotron radiation capabilities.

■ The efficacy with which scientific and technological frontiers will be advanced is influenced by many factors. Important factors and their effects are the following:

(a) Major future national synchrotron radiation research facilities must be dedicated in order to provide the operational flexibility and continuity necessary to perform diverse and sophisticated experiments. Experiments

utilizing synchrotron radiation will impose exacting specifications on the energy, current, and lattice configuration of machines. Failure to conform to such specifications will seriously inhibit scientific and technological progress.

(b) A balanced geographical distribution of synchrotron radiation capabilities is important to develop the potential of synchrotron radiation efficiently and to serve the larger number of active participants. The existence of several different centers of scientific investigation is highly desirable.

(c) Synchrotron radiation facilities should have a strong in-house scientific staff to provide high-quality experimental instrumentation and to perform prototype experiments. This will increase the number and productivity of participants and develop specialized experimental technology and talent. The operation of such facilities should not, however, be dominated by the in-house staff.

RECOMMENDATIONS

The Panel finds that there exists outstanding scientific and technological justification for a greatly expanded synchrotron radiation capability. Both construction of new dedicated facilities and expansion and dedication of existing facilities are required to meet national needs over the next ten years. Because new facilities require five years to plan and construct, the projected need requires the expansion of existing facilities during the next five years. Needs for synchrotron radiation beyond the next five years can be satisfied only if a commitment to the construction of new dedicated facilities is made now, because full expansion and dedication of existing facilities would satisfy less than one half of the national need projected for 1986. Even with an immediate commitment to the expansion of existing facilities and the construction of new facilities, there will be a chronic undercapacity that will become most acute five years from now.

The Panel *recommends* that, in response to national synchrotron radiation needs, an immediate commitment be made to construct new dedicated national facilities and to expand existing facilities so that optimized XUV and x-ray capabilities are provided with the following features:

- Significant x-ray capability with a synchrotron radiation critical wavelength (λ_c) between 1 and 3 Å and significant XUV capability with λ_c between 10 and 30 Å should be provided for substantial numbers of stations. An emphasis should be placed on achieving maximum source intensity and brightness throughout the x-ray and XUV ranges.

- The capability to provide for a variety of spectrometers (and associated optics elements) that are designed to match the sources is essential.

- A balanced geographical distribution of facilities is important to serve the U.S. scientific community effectively and to ensure that the full scientific and technological potential of synchrotron radiation research is realized. In choosing locations, consideration should be given to proximity to centers of scientific activity, in part to minimize travel time and costs.

- Provision should be made to incorporate special radiation source elements including superconducting wigglers, helical wigglers, and such promising, although speculative, devices as free-electron lasers.

- Consideration should be given to the scope of activity of local laboratories. The presence of biological and chemical laboratory facilities would be desirable.

- An adequate in-house research staff is essential. This staff should have liaison with local laboratories and other scientific institutions and would have several important roles including in-house research and development of facilities (e.g., spectrometers and optics), as well as collaboration with and provision of assistance to visiting users.

The Panel views action on these recommendations as essential to assure a needed increase in U.S. capability and stresses that commitments should be made to follow them as soon as possible.

3

SYNCHROTRON RADIATION SCIENCE AND TECHNOLOGY

INTRODUCTION

The response of materials to electromagnetic radiation has traditionally served as a primary indicator of both the structure and the reactivity of matter. The introduction of each new technique in this area of science has not only broadened its scope but has uncovered new layers of understanding in well-studied fields as well as bringing new fields above the threshold of viability for study. Historically, full scientific and technological exploitation of a given region of the electromagnetic spectrum has become feasible only after the development of a convenient, continuously tunable, intense radiation source. In this chapter we are not, therefore, examining the merits of a new technique but rather the practical utilization of a very wide range of the electromagnetic spectrum--from 10 eV (1240 Å) to 10^5 eV (0.124 Å)--as provided by a source that in brightness, tunability, time structure, and polarization characterization represents many decades in improvement over previously existing sources.

The best laboratory sources of photons in this energy range have been resonance lines in gas discharges and characteristic x-ray lines. Consequently, only widely separated, nontunable, discrete energies were available. One consequence of this limitation has been an artificial separation of the vacuum-ultraviolet (VUV) and x-ray fields--a gap that only synchrotron radiation can bridge. The spectral range between roughly 20 and 1000 eV photon energy--the XUV range--remained essentially closed to analysis until the advent of conveniently usable synchrotron light sources in 1963. The first plate taken at that time indicated the existence of a new quantum number. In the next several years, the gross features of absorption

coefficients were mapped throughout this range and the ubiquitous occurrence of resonances was demonstrated. Much work has been done since these early experiments, but this intermediate-energy range still remains relatively unexplored.

Before beginning the detailed discussions given in the following pages, let us briefly review the types of phenomena implied by the interaction of 10-10^5 eV synchrotron radiation with matter. All valence orbitals in atoms, molecules, and solids can be probed by photons of energy 50 eV, which slightly exceeds the binding energies of 2*s* electrons in neon and in fluorine compounds. The K edges of the very important second-row elements C, N, O, and F are reached at photon energies between 280 and 700 eV. By extending this range to 870 eV, the neon K edge is reached. At least one core level in every element is then accessible to photoabsorption and photoelectron spectroscopy experiments. Higher photon energies, up to 1500 or 2000 eV, are required to study extended fine structure in absorption experiments or the sudden-approximation regime in photoemission experiments. The K edges of heavy elements fall at energies in the 2×10^3 to 5×10^4 eV range. The extended x-ray absorption fine structure (EXAFS) method can be applied throughout the Periodic Table only if continuum radiation around this energy is available.

Radiation with x-ray energies and consequently with 0.2 to 12 Å wavelengths is used to determine the detailed structural arrangement of atoms in molecules and materials. Questions of interest range from the determination of the local arrangement of a few atoms ligating a specific ion through the study of the macroscopic structure of crystalline lattices. Included in this broad ranging scale are studies of periodicities and fluctuations that span one to hundreds of angstroms. Synchrotron radiation can so extend the power and versatility of the methods used to study these problems that its effect may well be to revolutionize fields in which structural information is of paramount importance. Structural studies using synchrotron radiation will have a dramatic impact in biology, chemistry, and the physical sciences as well as on research and diagnostic applications relevant to the nation's energy, environmental, and communications technologies.

Knowledge of local order, for example, is essential to an improvement of our understanding of areas such as heterogeneous catalysis, solution chemistry, enzymatic function, factors controlling the properties of amorphous materials, chemistry on solid surfaces, and the behavior

of dilute impurities in the gaseous, liquid, and solid state. There are unique opportunities for synchrotron radiation studies to contribute to the study of catalysis, dilute impurities, and biology, with application to energy problems, environmental monitoring, and the study of photosynthesis and muscle contraction, respectively.

Synchrotron radiation greatly facilitates dynamic structural studies by x-ray diffraction. Examples of processes that such studies can elucidate include nerve and muscle functions, growth of crystals from melts, and the mechanical failure of materials. Intense synchrotron radiation from facilities of the next generation also promises to make possible experiments in nonlinear optics at x-ray wavelengths.

This chapter treats the potential scientific and technological impact of synchrotron radiation. It includes a brief description of the generic goals of the numerous types of experiments that use photons whose energy lies between 10^1 and 10^5 eV and the connection of those generic goals to important scientific and technological issues. The breadth of the impact of synchrotron radiation is so large that it has been impossible to go into great detail for all the affected areas. Rather, our approach has been to focus on the subset of areas that in the opinion of the Panel will be most fruitful. The science and technology sections, together with other documentation, are then used to estimate the number of scientists who by 1986 will be actively interested in probing the new frontiers opened up by synchrotron radiation. That estimate is then translated into an evaluation of the number of synchrotron radiation experimental stations needed by 1986, a quantity of importance in planning for the country's future synchrotron radiation facilities.

X-RAY PHOTOABSORPTION SPECTROSCOPY

X-ray photoabsorption studies have provided some dramatic examples of the usefulness of synchrotron radiation in understanding electronic and molecular structure. This spectroscopic technique can determine the local atomic arrangement surrounding specific atoms in complex crystalline and noncrystalline environments. In practice, one measures the x-ray absorption in the material over a broad range of photon energies, starting at an absorption edge of the specific element whose atomic environment is of interest. Typically, the absorption is measured over a range

of about 1000 eV above the absorption edge of that element. Through analysis of the structure in the absorption behavior above the edge, it is possible to determine the distances to the atoms coordinating the absorbing atom. Progress in analysis of x-ray absorption fine structure has made possible determination of radial distances to an accuracy of about ±0.05 Å.[1] For those cases in which one shell of heavier near neighbors dominates the effect, accuracy as good as ±0.01 Å may be obtained. Furthermore, the position of the absorption edge gives information about the chemical state of the absorbing atom, and structure at energies just below the edge provides information about bound states associated with the atom and, indirectly, about the symmetry of its environment.

Synchrotron radiation has revolutionized our capability to obtain these absorption fine structure spectra. The absorption spectrum of Cu metal shown in Figure 1(a) required two weeks to obtain with the best conventional x-ray source--a 60-kW rotating anode tube. The Cu metal spectrum shown in Figure 1(b) was taken in 20 minutes, and the signal-to-noise ratio is better by an order of magnitude than that obtained with a conventional source. With synchrotron radiation it is possible to record spectra of atoms at dilutions down to 10 parts per million and of

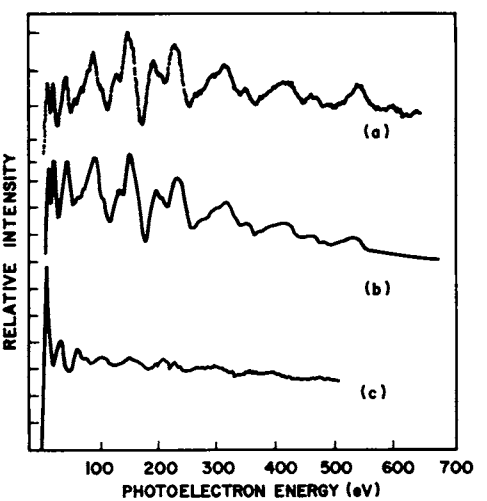

FIGURE 1 X-ray absorption spectra: (a) Cu spectrum using conventional sources; (b) Cu spectrum using synchrotron radiation; (c) thin superconducting Nb_3Ge film spectrum using synchrotron radiation.

atoms in thin films. An example of the latter is the spectrum of a thin superconducting film, which is shown in Figure 1(c). In this case, local order was shown to be correlated with the film's superconducting transition temperature.

The ability to investigate the local environment of a specific metal atom in a metalloprotein *in vivo* portends great advances in the understanding of the roles of such atoms in biological processes. An interesting case history is that of the structure of rubredoxin, a protein with a molecular weight of 8000, which catalyzes oxidation-reduction reactions important to energy transfer in biological processes involved in respiration, metabolism, bacterial photosynthesis, and nitrogen fixation. The molecule contains an Fe atom surrounded by four S atoms; and from earlier x-ray crystallographic results,[2,3] it had been concluded that three of the four Fe-S bonds in rubredoxin are of normal length, approximately 2.34 Å, while one seemed anomalously short, 1.95 Å. The electron transfer function of the molecule had been associated with this short bond. In recent x-ray absorption studies,[4,5] it was shown, however, that all four Fe-S bonds are equal in length, 2.24 Å, within the uncertainty of 0.1 Å. With subsequent improvement in the resolution, crystallographic methods also yielded the result that all four Fe-S bonds are the same.[6] Comparison of absorption measurements also showed that the reduced form of rubredoxin is characterized by an Fe-S distance about 0.05 Å greater than in the oxidized form and that the molecule in solution and in crystalline form has the same Fe-S distances (within ±0.02 Å). The work on rubredoxin demonstrates the potential that x-ray absorption spectroscopy has for determining chemical structures of compounds in their physiological environments. Other subjects of biological interest such as the binding, transport, and activity of physiologically relevant divalent cations like Ca^{+2}, Zn^{+2}, Cu^{+2}, and the Fe site involved in the oxygen transport of hemoglobin can easily be studied.

Applications of x-ray absorption spectroscopy to study problems in environmental and energy-related sciences are numerous. It has been possible, for example, to do *in situ* experiments on functioning heterogeneous catalysts.[7] These studies aid in interpreting the dispersion and mechanism of activity of important bimetallic systems such as Cu-Ru. Modifications of the material to improve catalytic activity have been suggested by the results of the studies.

The chemical forms of the various metals that occur in automobile exhausts and stack gases are largely unknown. Zn, Pb, and other metals in air samples have been examined; the results may yield information on how such atmospheric pollutants are washed out and dispersed. It is important also to know the details of the processes responsible for utilization and release of the vital gases in the atmosphere. Consider, for example, fixation of nitrogen by the enzyme nitrogenase, which contains 2 Mo atoms and 24 Fe atoms and which has a molecular weight of approximately one-quarter million. Crystals of the enzyme large enough for diffraction studies have not yet been successfully prepared. X-ray absorption studies have for the first time determined the oxidation level of the Mo atoms and have indicated that the Mo atoms are probably coordinated by several S atoms.[8] The detailed understanding of the nitrogen fixation process gained in such studies may lead to the development of chemical fixation processes that do not employ enzymes.

Using conventional methods, it has so far been impossible to confirm the suspected involvement of Mn in green plant photosynthetic oxygen evolution. Preliminary spectra of the Mn in the chloroplasts of an actual leaf have been obtained. The spectrum of Mn may be studied as a function of the physical, chemical, and physiological state of the chloroplasts--in the dark, illuminated, and as a function of light flash number--thereby providing insight into the role of the Mn in this critical biological process.

X-ray absorption spectroscopy is also an ideal method for structurally probing amorphous, alloy, and composite materials. Glasses are materials with unique mechanical and thermal properties, and understanding how to control these properties depends critically on a knowledge of structure around specific elements. Electrical properties of glasses are dependent upon specific metallic impurities, and an understanding of the role of these atoms is important in developing inexpensive glassy materials for solar-energy conversion. It should also be possible to determine whether impurity atoms occupy substitutional or interstitial sites in a crystal and to obtain information about the vibrational modes of such impurities. Such information will provide further valuable information on the mechanical and electrical properties of materials.

Usually this class of absorption measurements is insensitive to the nature of the surface unless the sample is highly dispersed (i.e., unless the surface-to-volume

ratio is high). The experiment can be made surface-sensitive by observing the Auger decay yield of the core hole being excited. This kind of experiment has been described theoretically but at this time has not been performed. Such an experiment can surely be done if sufficient photon flux is available. Surface x-ray absorption experiments could produce a much needed diagnostic tool for surface structure determination.

X-RAY CRYSTALLOGRAPHY

A synchrotron source, with its very high brightness and flexible time structure, is well suited to x-ray diffraction studies. This applies to elastic scattering, which includes Bragg and diffuse scattering processes, and inelastic scattering, which includes Compton scattering. Of particular importance is the broad continuous spectrum, which allows the wavelength at which a diffraction pattern is taken to be chosen at will. Every aspect of the investigation of the structure of matter is enhanced by the opportunity to select varying wavelengths at significantly increased intensities.

An important step in the determination of crystal structures is the evaluation of the relative phases of the scattered x rays. For several reasons, including the fact that in general there are relatively fewer experimental data for the more complex materials, heavy atoms are introduced into such structures to facilitate the determination of the phases. Conventional protein crystallography relies on the "isomorphous" introduction of heavy atom markers into the crystals. The preparation of such heavy atom derivatives is often difficult, and determination of the heavy atom positions is often tedious. The problem is especially difficult for many molecules of intermediate molecular weight (1000-5000), where heavy atom substitution, when it can be done, may severely distort the crystal lattice. The important peptide hormones fall into this molecular weight class.

Both the phase and amplitude of an x ray scattered from an atom are affected as the x-ray wavelength approaches an absorption edge of an atom. This phenomenon is called anomalous scattering. In principle, it is possible to determine completely the phase of a protein derivative (or the native crystal of the scatterer is already present) by taking data at several wavelengths on either side of the absorption edge of a heavy atom.[9]

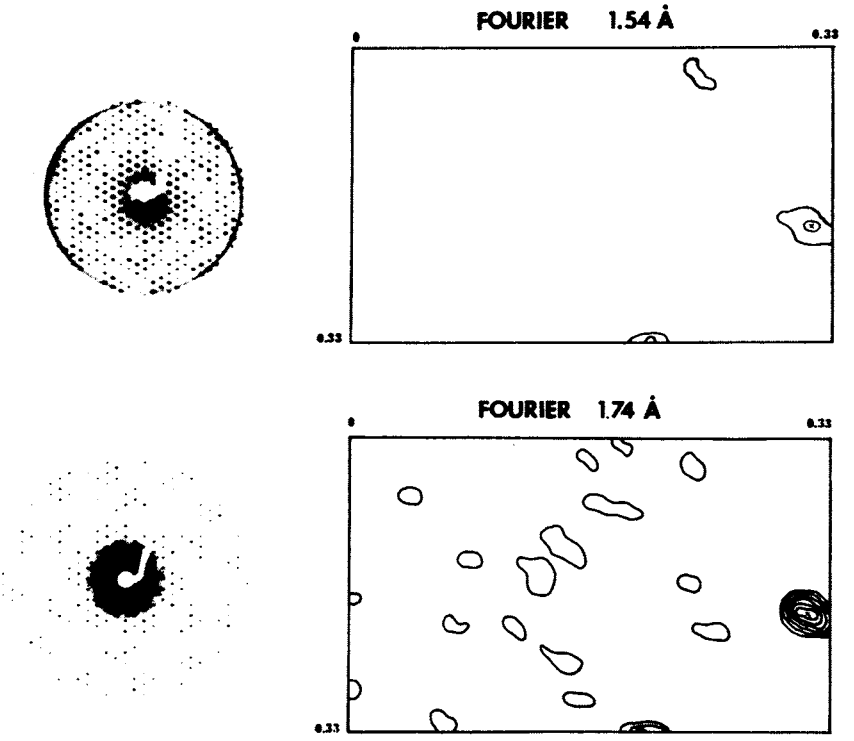

FIGURE 2 Precession diffraction photographs (*hk0* zone) on the same crystal of the protein rubredoxin along with *hk0* projection anomalous Fourier maps of electron density calculated for each zone. The maps were calculated using the intensities from each film and phases from the refined protein structure. The source for the upper photograph was copper K_α radiation from a 1200-W x-ray tube. The exposure time was 12 hours, and the resolution was 3 Å. The source for the lower photograph was synchrotron radiation from SPEAR (the storage ring at the Stanford Linear Accelerator). The exposure time was 15 minutes, and the resolution was 2.7 Å. The site marked with an X is the position of the iron atom. In the map from data taken just above the iron K edge (in energy), where f'' is maximized, the signal-to-noise ratio at the iron site is 6 to 1. For the map calculated at 1.54 Å, the signal-to-noise ratio at the iron site is 2 to 1.

Synchrotron light makes it possible to employ wavelengths close to the absorption edge without the restrictions imposed by the limited number of characteristic lines available from x-ray tubes. This method also has the advantage that the intensity measurements are made on the same crystal. Higher accuracies are thus possible because of the elimination of nonisomorphism effects. Results demonstrating the enhancement of anomalous scattering obtained by tuning synchrotron radiation to an absorption edge of Fe in the protein rubredoxin are shown in Figure 2. In the future, effects of this kind could allow determination of phases using any heavy atom whose absorption edge can be reached, as well as providing a method of determination of phase by using some of the naturally occurring heavier atoms in biological structures such as Fe, Cu, Zn, and possibly even P or S.

Synchrotron light offers substantial gains in data collection rates and in resolution over experiments done with high-powered, fine-focus, x-ray tubes.[10] The increase in rate of data acquisition will allow dynamic studies to be performed, as will be discussed shortly. An example of the increase in intensity can be seen in Figure 2, where precession photographs taken using the same protein crystal with synchrotron light and with a conventional x-ray tube are compared. The high natural collimation of the synchrotron light has improved spot resolution dramatically compared with a conventionally collimated x-ray source.

Crystals of suitable size are required for x-ray studies. The history of biomolecular crystallography is, in large part, the history of success or failure of making such samples. For example, the important enzyme nitrogenase has not been studied by diffraction because of a lack of suitable crystals. The high intensity and small source size available from an optimized synchrotron source should allow diffraction studies of small crystals--perhaps even in the "microcrystalline" domain of around 20 μm.

DYNAMIC DIFFRACTION STUDIES

Perhaps the most important application of synchrotron radiation to diffraction studies involves exploiting the increase in the speed of data acquisition to perform dynamic studies of time-dependent processes.

Dynamic diffraction experiments on contracting living muscle tissue are already being carried out. Several

groups have used synchrotron radiation to record in milliseconds[11] the complete low-angle diffraction pattern from stimulated muscle. Low-angle studies may be made of dynamic processes in chemical and materials systems and of macromolecules in solution. Changes in the size and shape of macromolecules are induced by a number of external stimuli. A muscle is induced to contract by a sudden influx of Ca^{+2} ions that have been released by the sarcoplasmic reticulum. Light, electrical impulse, pressure, heat, or mixing two solutions by flow can cause changes that reveal the function of macromolecules in biological processes.

Dynamic studies of single crystals are now becoming feasible. For example, carbon monoxide bound to hemoglobin can be released by an intense flash of light. The resulting conformational changes in the protein could be determined by recording the x-ray diffraction pattern at various intervals after the light flash. This process could be repeated as soon as the CO rebinds and as often as the crystal will withstand the flash pulses. The synchrotron pulse can be precisely timed with any repetition rate by moving the electron beam (and thereby the source) up and then down so that the x rays pass through a slit when desired. Such dynamic experiments will add a new dimension to our understanding of the functioning of macromolecular systems.

One cannot fail to be excited by the prospect of new diffraction techniques, or new variations in old methods, that specifically exploit the unique characteristics of synchrotron light. For example, the continuous character of the spectrum suggests strongly that the Laue method be re-examined. This method involves scanning the energy spectrum of scattered x rays at some fixed diffraction angle from a crystal with an energy-sensitive solid-state detector. Preliminary results[12] suggest that this method will prove most useful for dynamic studies of crystals. When modern diffraction techniques are combined with optical holographic processing, as recently developed by Stroke and collaborators,[13] one will indeed begin to approach in a practical way the goal of holographic reconstruction of crystallographic models from observational data.

LOW-ANGLE DIFFRACTION

Low-angle diffraction probes organization in materials over tens and hundreds of angstroms. Organization on this scale is encountered in lamellar and fibrous samples, such as retinal rods, muscle fiber, and biomembranes, and in dense multiphase systems, such as porous catalysts. Synchrotron light is well suited for experiments employing high-resolution, low-angle, point focusing cameras. The available intensity is important for several reasons. The complexity of the materials of interest results in a reduction in the average intensity of diffraction. Speed of data collection is also of essence because, even in the absence of radiation, many biological samples have limited *in vitro* lifetimes (often less than 24 hours). Experiments that are marginal even with the largest rotating anode x-ray tubes can be greatly facilitated by the use of synchrotron radiation. A practical example is the study of radiation-induced voids in reactor vessel materials, which appears to be feasible using synchrotron light but has been marginal with conventional sources.

The enhanced anomalous scattering associated with tuning the x-ray wavelength to the absorption edge of some constituent atom is also useful for low-angle scattering.

DIFFUSE X-RAY SCATTERING

Quasi-elastic diffuse scattering[14] occurs when scattered waves from different atoms in the crystal are not in register because of density fluctuations such as those caused in a material by normal thermal vibrations, random small voids, clustering, or the onset of a phase transition. In general, the technique provides information concerning atom-atom correlations out to about the fiftieth coordination shell of neighbors. Future synchrotron radiation sources will offer the possibility of increased resolution, which may make apparent the presence of longer range correlations. Thermal diffuse studies should lead to an improved understanding of the effects of radiation damage on materials such as occurs when materials are used as structural elements in thermonuclear reactors. Structural changes accompanying solid-solid phase transitions (for example, martensitic transitions) can be studied. The understanding of such instabilities in crystals is important in designing materials for use in low-temperature applications such as superconducting power lines.

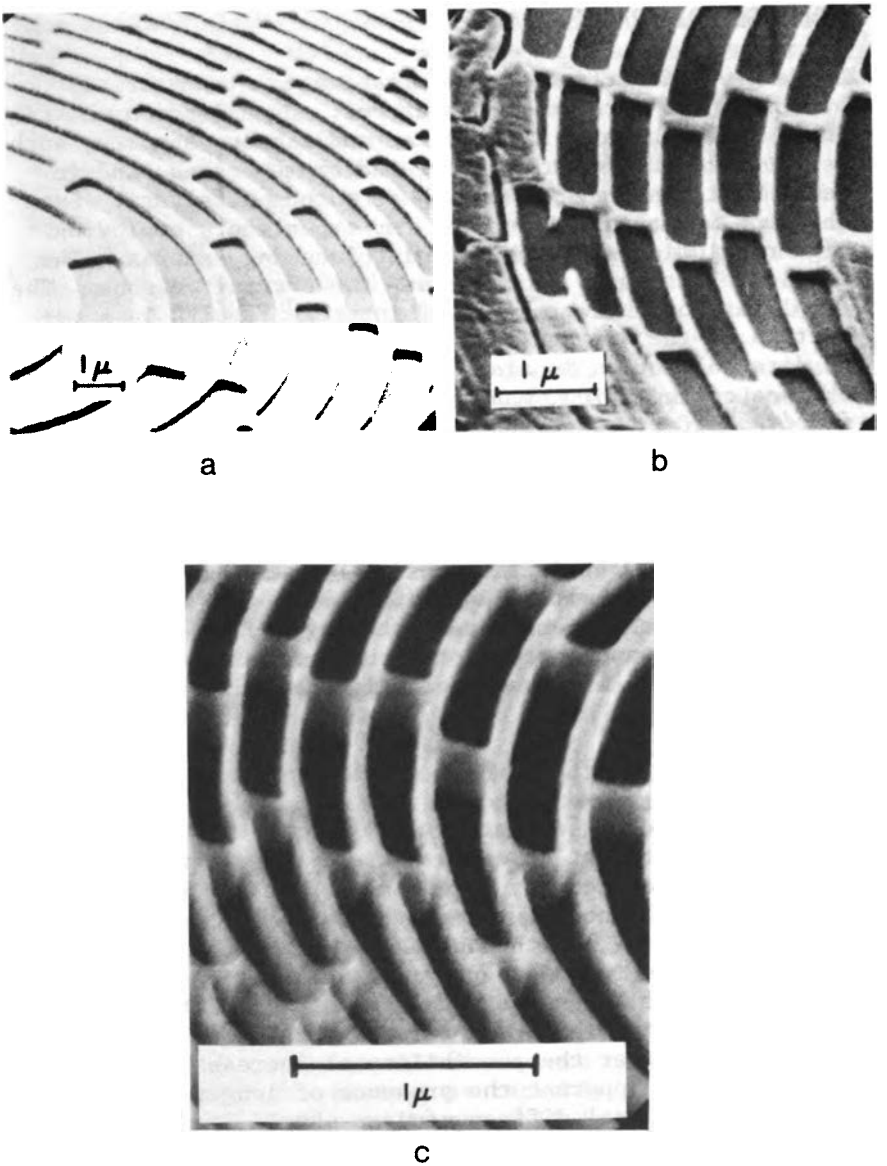

FIGURE 3 Lithography done with synchrotron light. a,b, Views of two different regions of a Fresnel zone plate replica with individual lines as narrow as 0.07 μm; c, a similar structure with high vertical walls.

X-RAY LITHOGRAPHY, MICROSCOPY, AND TOPOGRAPHY

X-Ray Lithography

Synchrotron light is particularly suited for x-ray lithography and related techniques of microscopy. In the lithography process, a mask provides a pattern to be replicated. The mask is placed over a photoresist material, the system is illuminated with x rays, and, finally, the photoresist is developed. Spiller and co-workers have shown[15] that x rays in the vicinity of 10 to 50 Å provide the best contrast with masks made to reproduce details of a pattern at minimum size; synchrotron light is the most intense source of such radiation. Further, the high degree of collimation of the light eliminates loss of resolution caused by penumbral blurring, so that submicrometer patterns have been accurately reproduced on photoresist material with masks as much as a millimeter distant from it.[15] Examples of lithography done with synchrotron light appear in Figure 3. The applications of the technique to integrated circuit fabrication are very promising.

X-Ray Microscopy

In a process similar to that employed in x-ray lithography, a photoresist exposed by x rays transmitted through the sample under study (a cell, for example) is developed and then viewed with a scanning electron microscope. Given a dedicated synchrotron radiation source, it appears[16] practicable to investigate biological samples with resolutions near 100 Å and exposure times much less than a second. This resolution is lower than that which can be attained with electron microscopy, but the x-ray technique is less surface-sensitive and, by variation of x-ray wavelength, offers the prospect of selective enhancement of structural features. Unlike electron microscopy, the x-ray scans may be performed *in vivo*. The two microscopies are thus complementary in a variety of ways.

Horowitz and Howell[17] have experimented with another kind of x-ray microscopy in which the synchrotron light transmitted through a pinhole was used to scan the specimen. An x-ray spectroscopic detector was used to measure the specimen fluorescence, resulting in a chemically sensitive microscopy with great depth of field but lower resolution than the scheme described above. Submicrometer-scale quantitative analysis is practicable with the design of Horowitz and Howell.

Holography, with its three-dimensional capability, is in some sense the most exciting form of microscopy. The first accomplishment in short-wavelength holography (λ < 3000 Å) was the interference pattern obtained by Hellstrom in 1932 using 8 Å radiation in a Lloyd's mirror apparatus. More recently, a holographic Fourier transform reconstruction was achieved using hydrogen Lyman radiation (1215 Å), again using a Lloyd's mirror. The use of Gabor's idea of obtaining magnification by a factor λ_1/λ_2 by making the hologram at λ_2 and reconstructing it at λ_1 is another promising avenue of investigation. Developments in Fourier transform holography and the possibility of improved detectors providing digital methods for reconstruction together with the high brightness of synchrotron radiation opens up exciting prospects in short-wavelength holography.

Synchrotron radiation also promises to advance biomedical applications and radiographic techniques. Significant improvement in detail and contrast may be expected in tomography (scanning radiography), because increased photon counting rates would improve a gray-level distinction. Dichromography is a method whereby contrast is enhanced by making measurements at two wavelengths. The broad spectrum of synchrotron radiation would permit experiments at any set of wavelengths and near any absorption edge.

X-Ray Topography

X-ray diffraction topography permits visualization of many types of crystal defects such as dislocations, voids, and phase boundaries.[18] It has been used with notable success in fundamental studies of defect conditioned properties of materials. At the same time, some of the techniques are sufficiently simple that they are applicable to diagnostic studies.

With synchrotron x rays, elementary forms of single-crystal topography can be performed with increases over methods employing conventional sources in defect sensitivity and in spatial resolution. When simpler approaches are used, the broad spectrum of incident synchrotron radiation can be used to examine crystals having appreciable curvature.

As with other classes of research, it is synchrotron radiation's potential for dynamic studies that is most interesting. For example, the strengths of crystalline materials and their behavior near the elastic limit are largely determined by dislocation motion. Until now this motion has been only studied by interrupted creep tests.

With an optimized x-ray synchrotron source, it should be possible to follow these motions in real time, perhaps with the aid of an electronic imaging system.

INELASTIC X-RAY SCATTERING

Apart from some Compton[19] and resonant Raman[20] scattering experiments, no inelastic scattering experiments have been performed using synchrotron radiation, because the flux currently available is too low to do them readily. A dedicated synchrotron storage ring source with a 1-A current employing an improved magnetic lattice and monochromator should provide factors of 10^3-10^4 enhancement in flux and should thus allow successful high-resolution Raman,[21] Compton,[22] and nonlinear scattering experiments to be done.[23] New frontiers should be opened up using these probes of electronic properties, with perhaps the most significant advances in the study of nonlinear x-ray phenomena. By analogy with long-wavelength nonlinear optics, one would expect through such studies to learn about optical properties on an angstrom scale (e.g., the detailed microscopic nature of the excited-state charge distribution caused by the absorption of a visible photon).

XUV ABSORPTION, REFLECTIVITY, AND FLUORESCENCE SPECTROSCOPIES

Absorption, reflectivity, and fluorescence experiments measure the intensity of the transmitted, reflected, or re-emitted radiation relative to the incident radiation intensity as a function of photon energy. For radiation in the XUV range (i.e., roughly less than 1 keV), these measurements probe the electronic structure of all phases of matter, from atoms or molecules to solids or liquids in various geometrical configurations and purities.

These experiments will continue to have a direct impact on a wide range of scientific and technological applications. Absorption studies of gas-phase molecules are important in providing basic data needed in such areas as diffusion of radiation in stars, the interpretation of astronomical data, studies of aeronomy, radiation processes in plasmas, studies of chemical structures, and condensed-phase studies of molecular adsorption, chemisorption, catalysis, and organic solids. Optical constants over a wide range of photon energies are needed for materials

used for practical applications such as the materials used for the walls of controlled thermonuclear reactor (CTR) chambers.[24] High-resolution measurements can and will provide valuable information about, for example, electronic states, impurities, and localized excitations of commonly used or new materials of interest for device purposes.

Most of the XUV synchrotron radiation work reported to date has involved total photoexcitation cross-section measurements of photoabsorption, reflection, and photoionization.[25] This trend is a consequence of the fact that such measurements in general require only low-to-moderate radiation intensity but depend critically upon the availability of a wide range of continuously variable wavelengths. Synchrotron radiation has many obvious and often unique advantages over alternative sources for these measurements. It is the only intense continuum source, particularly above ~20 eV, and thus the only photon source capable of high-resolution energy-dependent measurements.[26]

Modulation spectroscopy of solids in the ultraviolet wavelength regime is a second-generation experiment that requires a stable source of radiation of high intensity with a continuous spectrum into the far ultraviolet. Modulation spectroscopy involves the application of a small alternating perturbation, e.g., an electric field, a stress, or a temperature to the sample and the measurement of the small component of the reflectivity oscillating at the frequency of the perturbation. This change in the reflectivity can be small--of the order of 10^{-5} of the reflected signal. High incident intensity is especially important in the vacuum ultraviolet because sample reflectances are usually very low. Conventional sources have been used to 6 eV frequently and to 8-10 eV occasionally, but a storage ring is clearly superior above 5 eV and has in fact made modulation spectroscopy possible beyond the LiF edge in the vacuum ultraviolet.[27]

Modulation spectroscopy extended into the vacuum ultraviolet will become increasingly important because it allows one to investigate transitions from core levels localized on specific constituents of the solid. For example, Lynch[28] has recently used synchrotron radiation to measure the electroreflectance from GaP (a commercially important semiconductor) in the photon energy range of the core-level transition from the Ga $3d$ level (~20 eV). More information will be obtained when third-generation modulation experiments are extended to even higher photon energies that take advantage of the polarization of the synchrotron radiation to analyze the symmetry of electron

states participating in optical transitions. For example, with the intensities that will be available with the recommended facilities, measurements could be made of the electroreflectance while a uniaxial stress is applied to a single-crystal sample. These measurements in the photon energy range of core-level excitations (with fixed polarization with respect to the crystal orientation) will yield new information about deformation potentials, which describe how electronic states are modified by crystal deformations. There is no hope of doing these experiments with conventional light sources.

An appropriate example of the utilization of the continuous spectrum of synchrotron radiation in gas-phase absorption is given by the work at DESY (German electron synchrotron) by Eberhardt *et al.*[29] on the fine structure of the carbon K edge in vapors of simple hydrocarbons. In these experiments, the yield of low-energy secondary electrons was measured as the photon energy was swept through the carbon K edge in various hydrocarbons. This technique, which is known as yield spectroscopy, is convenient to use for thick solid samples for which the absorption of light is large.[30] The number of low-energy secondary electrons is related to, but not quite proportional to, the absorption cross section. The curves for the yield of electrons as a function of photon energy for CH_4, C_2H_6, C_2H_4, C_6H_6, and C_2H_2 are shown in Figure 4. In all cases, the primary excitation process is localized on a carbon atom; hence the different spectral features in the curves depict the various excited states that have transition amplitudes from the carbon 1*s* state. This illustrates the unique capabilities of synchrotron radiation to pick an electron localized on one atomic species and excite it to any kinetic energy. The experiments shown here are limited by the combination of intensity and resolution in the primary photon beam so that they permit only the most preliminary interpretation at this time. Experiments of this type probe the reaction of the molecule to a very specific test disturbance--the injection of a well-defined amount of energy in the center of a carbon atom. This creates a localized electron vacancy and an electron of specific but tunable energy. Ultimately, these spectral features should be relatable to the polarization of valence electrons toward the K hole and hence to the chemical reactivity of a specific site. One may look forward to increased resolution, to alternative or simultaneous observation of ionic fragments, and possibly also to analogous experiments with oriented molecules, which could take

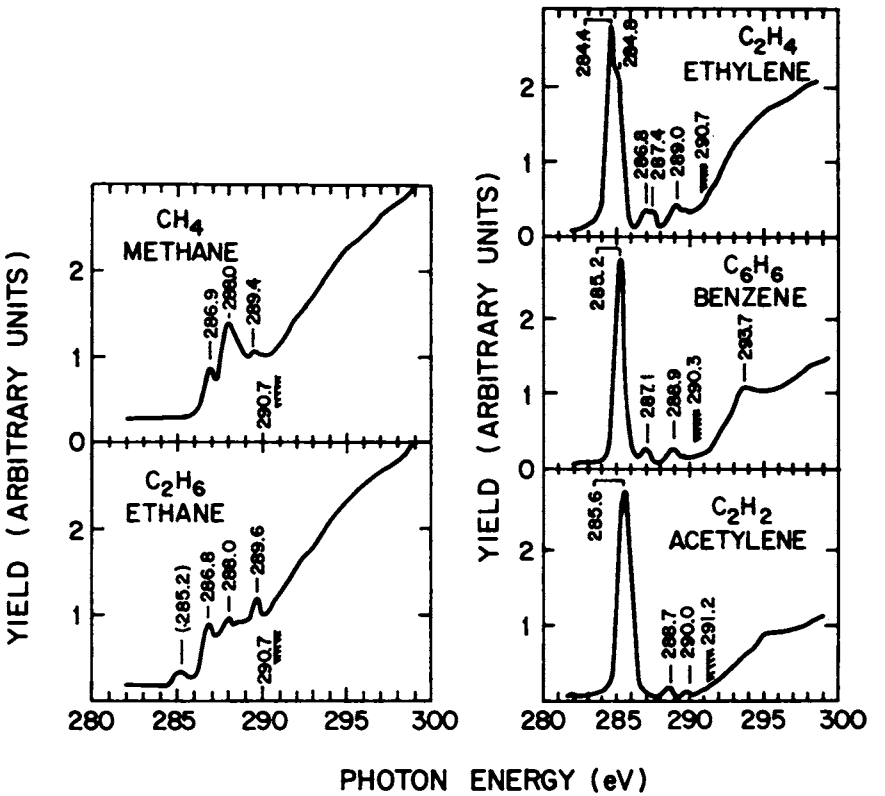

FIGURE 4 Electron yield spectra of gaseous methane, ethane, ethylene, benzene, and acetylene in the range of the C absorption.

advantage of the polarization characteristics of synchrotron light.

Because the K edges of the chemically important second-row elements C, N, O, and F lie between 280 and 870 eV, it is important that this range be covered by synchrotron radiation sources. Desirable coverage would, of course, require that photon flux be available throughout the extended fine-structure range, i.e., up to 1500 eV or higher. It is important in designing new facilities that adequate provision be made for this high-energy end of the XUV range.

There are two other quite different kinds of experiment of this general category for which the properties of synchrotron radiation could be useful: (1) a wide variety

of fluorescence experiments on atoms and molecules can be visualized in which the time structure and polarization properties are exploited. The simplest of these involve lifetime measurements and determination of excited-state spins. (2) Synchrotron radiation furnishes a much brighter source in the far infrared than is presently available. A 0.5-A machine is several orders of magnitude brighter at an energy of 0.001 eV than a 6000 K blackbody and could be useful for special broadband spectroscopy applications. Eventually, however, one would expect that tunable lasers would become available in this region.

PHOTOELECTRON SPECTROSCOPY

In photoelectron spectroscopy, monoenergetic photons are absorbed by a sample and the distribution in kinetic energy of the emitted electrons is measured. This electron energy distribution reveals the energy and relative population of the allowed ionic states of the system, yielding both different and more detailed information about the system being studied than an absorption experiment. Photoemission experiments conducted with both ultraviolet radiation and x rays have proven to be useful in determining the electronic properties of all forms of matter.

The most sophisticated class of photoelectron spectroscopy experiments measures the number of electrons emitted per incident photon as a function of the kinetic energy of the electron, the photon energy, the direction and type of polarization, the direction of the emitted electron with respect to the incident light, and the orientation of the sample. From the information obtained in such an experiment, one can map out the distribution of electrons in energy as well as the spatial and momentum characteristics of each electronic state. Since nearly all physical and chemical properties of matter directly depend upon the detailed nature of the electronic states, these experiments have an immense significance. These are difficult experiments requiring a very bright source of photons with a specific polarization that can be tuned continuously over a wide range of energies.

The following few subsections illustrate the potential of such photoemission techniques by a few selected examples from work on solids, solid surfaces, and gas-phase molecules. This field is in its infancy and will blossom when more intense synchrotron radiation sources are made available.

Solids

When the sample being studied has a fixed orientation in space as does a single crystal or single-crystal surface, the angle of the polarization of the light with respect to the sample becomes important for probing spatial anisotropies of the sample. Similarly, for a solid sample, there is an anisotropic emission of electrons that depends upon the energy and polarization of the photon as well as the orientation of the sample. The uniqueness of synchrotron radiation lies in the ability to tune the photon energy continuously over a wide range (especially above 20 eV) with a fixed polarization and with sufficient flux to make the measurements feasible.

Figure 5 shows spectra from an angle-resolved study of the copper 3*d* bands, which exhibit significant anisotropic effects up to a photon energy of 170 eV.[31] In these measurements, the emission normal to two different

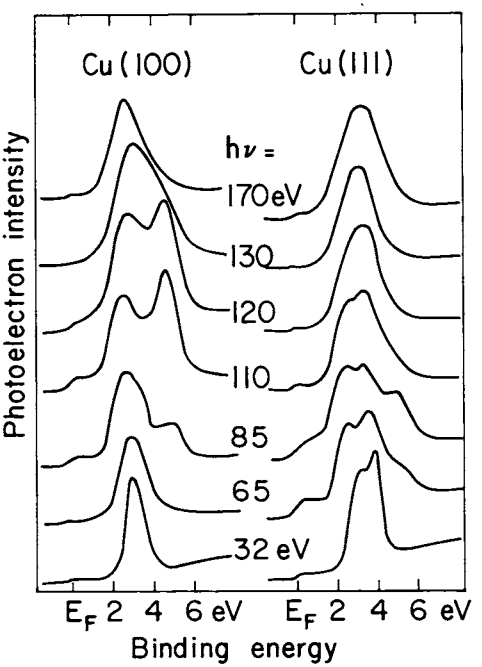

FIGURE 5 Angle-resolved photoelectron energy distributions from two different faces of Cu as a function of photon energy.[31] Only the electrons emitted normal to the surface are measured.

single-crystal surfaces was measured as a function of photon energy. These data illustrate dramatically how anisotropic the emission is with respect to the crystal orientation and how sensitive the angle-resolved emission is to the photon energy. This kind of experiment, when extended to many angles of emission and when suitably analyzed, can determine the energy-momentum dispersion curves for both the occupied and unoccupied electronic states of the solid. Such energy-momentum dispersion curves represent a detailed description of the basic electronic structure of solids, i.e., the fundamental electron-ion and electron-electron interactions.

Solid Surfaces

Strong inelastic electron scattering in solids makes photoelectron spectroscopy of solids a surface-sensitive technique. Electrons that have been photoexcited inside a solid sample have a very short escape depth (4-20 Å) for the photon energy range covered by ultraviolet or soft x-ray radiation. This escape depth usually has a minimum in the 30- to 100-eV kinetic energy range. Therefore, the spectra shown in Figure 5 were taken in the range of maximum sensitivity. Further refinements and interpretation of such experiments should yield surface density of states and allow mapping of electronic charge distributions near the surface. The electronic configuration of a solid surface is one of the most fundamental properties of a surface, since it dictates how it interacts with its environment. Therefore, understanding the properties of clean, well-characterized surfaces is a prerequisite for understanding surface reactions, adsorption processes, and interfacial properties, all of which are directly related to modern technological problems, especially in the areas of energy research and catalysis.

The surface states of semiconductors (both the occupied and unoccupied states) are important for the design of practical devices. These surface states have been studied for some time using photoelectron spectroscopy, but recent measurements using the tunable photon energies available from synchrotron radiation have had a major impact on the field. Knapp and Lapeyre[32] have used their angle-resolved energy analyzer exploiting the variability of photon energy, to find the occupied surface states in GaAs and map out their energy-momentum dispersion. We can look forward to this type of experiment being carried

out on a wide variety of both semiconductor and metal surfaces. The wavelength tunability also enables one to probe the unoccupied surface states of a semiconductor by a technique called partial yield spectroscopy.[33] With this technique, the absorption cross section for excitation from a core state to an occupied surface state is recorded as a function of photon energy by observing the Auger decay of the hole. The scope of these experiments will be expanded to include an investigation of various core states (using different photon energies) with different orbital character so that the orbital character (e.g., s, p, d) as well as the spatial localization of the surface states can be determined. Such experiments require a wide range of photon energies as well as an intense source.

There is a very active theoretical interest in these surface photoelectron experiments. It should soon be possible to make detailed spatial plots of surface states and of dangling bonds by appropriate analyses of the photon energy and polarization dependences of angular-resolved electron emission data.

Photoelectron spectroscopy from solids is a real-time experiment in that the spectrum changes as the surface becomes contaminated. Even in an ultra-high vacuum an experiment must be completed in several hours or less. This sets a requirement upon the necessary photon flux that depends upon the partial photoionization cross section of the electronic level being observed, the kinetic energy of the emitted electron, and the chemistry of the surface being studied.[34] A basic problem is that photoelectron experiments should be conducted with constant-energy bandwidth ΔE. As the photon energy is increased from the 20-eV range to the 100-eV range and above, a combination of factors--available flux, photoemission cross section, and acceptance into the analyzer, to name three--all conspire to reduce the spectral intensity. Thus intense sources and efficient monochromators and electron spectrometers are required.

Surface Adsorption Systems

There is a rapidly growing interest in fundamental studies of surfaces and surface reactions, stimulated in part by the advent of new experimental techniques such as photoelectron spectroscopy and in part by the relevance of research on surfaces to very practical problems facing our technological society. These problems are found in areas

such as heterogeneous catalysis, corrosion, embrittlement, and the development of photocells and many solid-state electronic devices. It is also clear that the new techniques in this field are of practical use as diagnostic tools even before basic understanding has been achieved. In this section we address the use of synchrotron radiation as a new diagnostic tool for surface studies.

The fundamental question relevant to adsorption on surfaces is, "What is the nature of the bonding configuration?" It is likely that synchrotron radiation will play an important role in answering this question. Close coupling between theoretical and experimental programs on photon-dependent angle-resolved surface photoemission using synchrotron radiation should lead to a technique capable of determining the energy, momentum, and spatial characteristics of the bonding orbitals, as well as the local geometry.[35,36] Theoretical studies of surface adsorption are severely limited by a lack of structural information. Conventional techniques such as x-ray scattering are not sensitive to the state of surfaces; and low-energy electron diffraction, even though it has made tremendous progress, is limited to specific well-ordered systems.

The detailed understanding of the photoionization excitations of gas phase molecules, like that shown in Figure 6 (to be discussed in the next subsection), can be used successfully to determine bonding configurations of adsorbed molecules. For example, a comparison of the frequency dependence of the partial photoionization cross section of adsorbed CO and gas phase CO led to the correct identification of the energy levels of adsorbed CO.[37]

The surface fixes the orientation of the molecule relative to the incident light and electron direction, so that if the angular dependence of the signal from each molecular orbital is known, the bonding geometry can be determined. The relevant gas-phase information is now available for CO and N_2 and will surely be forthcoming for larger molecules. Several experimental groups are now in the process of interpreting their angle-resolved photoemission data for adsorbed CO in terms of the gas-phase data. An example of the capabilities of this approach is given in Figure 7 from the work of Smith *et al.*[38] for CO adsorbed on Ni(100). The three curves are photoelectron energy distributions for normal emission with different polarization directions at a photon energy of 28 eV. The two peaks marked 1 and 2 are from the 1π and 4σ levels of CO, respectively. Curve 1 is for a mixed polarization

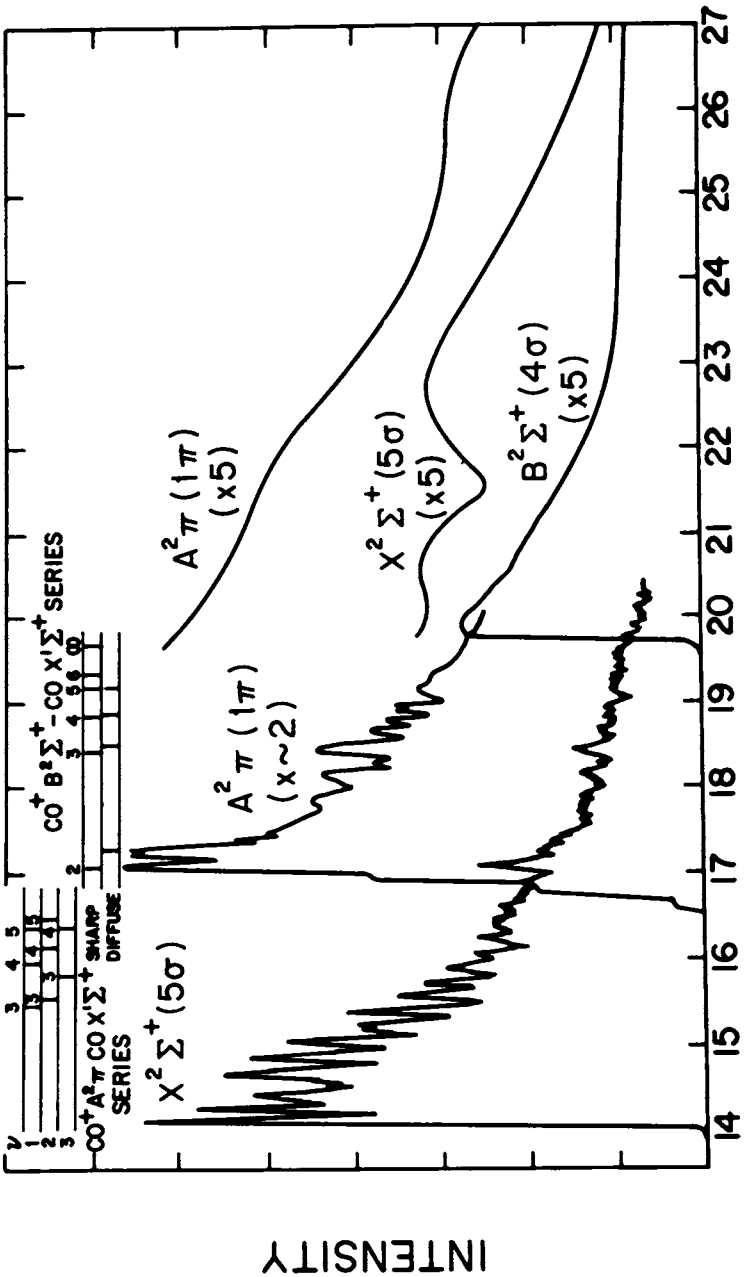

FIGURE 6 Partial photoionization cross-section curves for the three lowest energy ionic states of CO^+. These curves were swept continuously as a function of photon energy with 1.6 Å slits and 0.4-eV electron analyzer resolution.

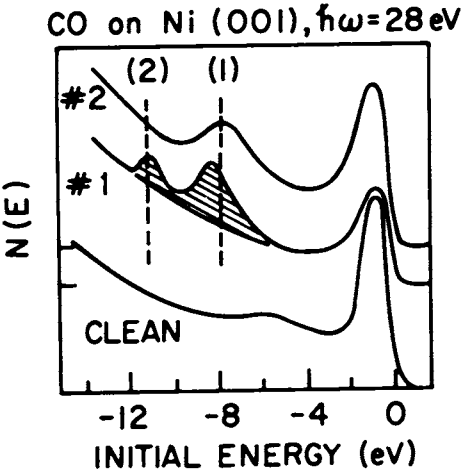

FIGURE 7 Angle-resolved photoelectron spectra from CO adsorbed on Ni(100),[38] at $\hbar\omega$ = 28 eV. The bottom curve is for clean Ni, while curve 1 is for CO adsorption with a polarization vector out of the surface plane. Curve 2 is the same adsorption system with the polarization vector in the surface plane. Only electrons emitted normal to the surface are collected.

with components of the electric field in the plane of the surface and perpendicular to the plane, while curve 2 is for polarization totally in the plane of the surface. It is known from the gas-phase work on CO that the cross section for excitation of the 4σ level is small for a polarization perpendicular to the molecular axis, and that there is no photoemitted current in the direction parallel to the molecular axis for this polarization. Therefore, the fact that the 4σ level is not present in curve 2 is consistent with CO standing up on the surface with the carbon end down. Much more work at other photon energies and different angles must be completed before a positive conclusion can be reached, but this figure clearly illustrates the potential of angle-resolved, polarization-dependent measurements.

Atoms and Molecules

An atom or molecule in the gas phase has no preferred orientation with respect to the light polarization or detector direction, so that many of the angular effects discussed for solids are averaged out. The angular dependence is a function of the angle between the polarization vector and the direction of detection, so that the measured photoelectron energy distribution is a function of the photon energy, the kinetic energy of the electron, and this angle. There is a multitude of ionic states whose energy can be measured by the difference between the photon energy and the kinetic energy of the emitted electron. The cross section as a function of photon energy for each allowed ionic state is called the partial photoionization cross section. Partial photoionization cross sections are of major interest since they contain much more detailed information and are easier to interpret than total cross sections. Together with theoretical analyses, they will yield a more complete understanding of the electronic structure of atoms and molecules. A basic understanding of various photoionization processes and products involving molecules is critical for many areas of science.

The intense light continuum of synchrotron radiation is ideal for studies of the photon energy dependence of the partial photoionization cross section. The partial photoionization cross section for any state can be measured by changing the photon energy with the electron energy analyzer acceptance energy, fixed to the photon energy offset by a voltage equal to the binding energy of the state. These curves can be obtained for any kind of ionic state, one-electron excitations, vibronic excitations, or many-electron excitation, i.e., any excited state of the ion. Figure 6 illustrates this approach for the three lowest energy electronic states of CO^+.[39] In principle these spectra could be obtained in individual laboratories using various light sources, especially in the photon energy region below 20 eV. In practice, synchrotron radiation, with its smooth intense continuum, is a qualitatively superior source for photon-dependent partial-photoionization cross-section measurements that opens up many new applications. It is likely that more of these measurements have been made during the last year using synchrotron radiation than have ever before been made to date using laboratory sources. For the photon energy range above approximately 20 eV, synchrotron radiation is the only feasible source at present.

The curves shown in Figure 6 are for the three lowest energy electronic states of CO^+. The 5σ state is the ground state of the ion that results from removing an electron from the 5σ orbital with a binding energy of ~14 eV. The 1π state of CO^+ is the first electronically excited state, with an electron missing from the 1π orbital (16.7-eV binding energy). The 4σ state of CO^+ is the second electronically excited state, which corresponds to an excitation of an electron from the 4σ level of CO with a binding energy of ~19.7 eV. A wavelength resolution of 1.6 Å and an analyzer resolution of 0.4 eV were used in these measurements. The rapid oscillations near threshold (below 19 eV) are due to autoionization. Autoionization via electronic and vibronic interactions is clearly observed, as is the coupling of different ionic states to a specific autoionizing level. This detailed information can be used to determine the shape of the potential energy curves for the ionic states as well as the excited states of the neutral molecule, which are the intermediate states in autoionization. Above approximately 21 eV, the shape of the cross sections is primarily due to ionization into the continuum. This portion of the spectrum has been explained quantitatively by recent theoretical calculations for N_2 and CO.[40,41] For example, the broad peak in the 5σ state near 23 eV is due to a scattering resonance in the final state. The outlook for this field is very exciting, especially considering the prospects for comparing the data and calculations over a wider photon energy range (up to 1000 eV) and for achieving better photon resolution.

Such measurements will require greatly increased source intensities as well as improved monochromators and electron spectrometer systems. It is important to note that the maximum flux desired will always exceed what is available and will be dictated by the higher end of the energy range that one wants to measure.

Future experiments will extend combined photoelectron and mass spectrometry measurements to determine not only the distribution of ionic states but also the final photodissociation products as a function of photon energy. An interesting experiment has been completed on the photoionization spectra of CO_2 dimers formed from a supersonic molecular beam. A comparison of the photoionization spectrum for mass-to-charge ratio of 88, the dimer, and for mass-to-charge ratio 44, the monomer, revealed a shift in the appearance potential for the dimer ion of approximately 0.37 eV. In addition, a great deal of the autoionization

fine structure was either missing or broadened in the dimer compared with the monomer. These types of experiments when combined with photoelectron spectroscopy will provide a link between the gas phase and the liquid or solid phase of matter necessary for understanding nucleation and condensation.

RELEVANCE TO NATIONAL TECHNOLOGICAL NEEDS

We have seen that synchrotron radiation substantially enhances our ability to use light to probe the structure and behavior of matter. Some applications relate directly to the fundamental properties of the systems under study; others, more applied, are diagnostic in character. Both classes of investigation are of direct relevance to national needs. In the following subsections, examples will be given in the areas of energy technology, medicine, electronic devices, and environmental science.

Energy Technology

One of the areas of growth in synchrotron radiation uses has been the application of x-ray absorption spectroscopy to catalysis. Only one research group in the United States employed this technique for any purpose in the period prior to the availability of synchrotron light; but in 1975 at least six U.S. chemical and energy companies invested money and manpower for studying catalysis in this way. Using x-ray absorption spectroscopy, it is now possible to monitor the local environment, activity, and degradation of dispersed catalytic particles in working models of reaction chambers and fuel cells. Also, information about basic mechanisms in catalysis can be obtained by using such techniques as low-angle scattering and photoelectron spectroscopy.

X-ray absorption spectroscopy is a particularly useful method for investigating disordered systems, which have many important applications such as the development of glasses for solar-energy collecting devices, amorphous semiconductors, and superconducting thin films. Also, one can study the local environment of dilute atomic impurities that affect the mechanical, thermal, or electrical properties of materials. As an example, defects, impurities, and their ordering behavior in high-temperature insulators are important for the utilization of such

insulators in magnetohydrodynamic power generation. A further understanding of structural ordering behavior in superionic conductors will improve the prospects for engineering new high energy density batteries.

Metal atoms are critical for the activity of many biological molecules. This is the case for certain molecules in the leaf chloroplasts responsible for photosynthesis and for nitrogenase, the bacterial enzyme that catalyses the conversion of atmospheric nitrogen into ammonia. Nitrogenase and chloroplast function can be effectively studied by x-ray absorption spectroscopy, which can furnish structural information not otherwise available because of the extreme dilution of the metal atoms. The study of the catalytic role of nitrogenase is part of an effort to devise more efficient methods of producing synthetic fertilizer.

Various applications of synchrotron radiation should assist in the development of fusion as an energy source. For example, in order for the emission spectra of ions such as N or O, present as impurities in the thermonuclear plasma, to be employed for diagnostics, more complete and accurate spectroscopic data are needed, especially on the high-lying states. Also, a significant source of contamination of a plasma is gas desorption from container walls induced by electrons, ions, and photons, and thus more data on photon-induced desorption are needed. Finally, there are formidable materials problems associated with the first vessel walls; one is production of voids by radiation. Low-angle diffraction study of these voids with high-powered x-ray tubes is at best marginal. With synchrotron radiation there is a real prospect of obtaining useful information by this technique.

Materials problems also arise in magnetohydrodynamic generators, fission reactors, coal conversion systems, and hydrogen storage materials, to name but a few. Both static and dynamic information are needed concerning materials problems involving impurities as well as defects and transformations produced by radiation, heat, and chemical reactions. The usefulness of techniques such as topography, microscopy, and low-angle and diffuse scattering in obtaining such information is greatly increased when conventional photon sources are replaced by synchrotron radiation.

Electronic Devices

X-ray lithography using synchrotron radiation has great potential for a central role in the development and manufacture of integrated circuits of submicrometer dimensions (semiconductors, Josephson junction magnetic bubbles, integrated optics). Although integrated circuits have not yet reached the submicrometer level, a trend toward smaller, higher-density circuits with higher performance and at lower cost, has been the major development of the electronics industry in the past decade. As we have seen in Figure 3, high-quality structures with 700 Å element widths have already been fabricated by x-ray lithography; however, before there can be widespread industrial usage, significant problems must be solved, e.g., the registration of multiple layers of submicrometer structures required for most devices.

The ability to produce by replication devices and structures having submicrometer dimensions will have many important technological and scientific uses in addition to those mentioned above, i.e., applications to microwave devices, acoustic devices, injection laser display devices, one-dimensional superconductor structures, Fresnel zone plates for x ray and XUV optics, and submicrometer transmission gratings.

Medicine

As Lewis Thomas has eloquently indicated,[42] fundamental advances in the technology of medicine depend upon advances in the knowledge of disease mechanisms. Synchrotron radiation can make unique contributions to this knowledge through its use in microscopy, radiography, low-angle scattering, and x-ray absorption spectroscopy. The binding, transport, and activity of physiologically relevant divalent cations, such as Ca^{2+}, can be studied with the last-named method. From the differences found between cation behavior in normal and diseased cells, it may be possible to shed light on questions of molecular defects in muscle function, such as those that occur in muscular dystrophy. Combined absorption and low-angle scattering studies will be useful to determine how these divalent cations react with biomembranes. These ions have been linked with sickle cell anemia, and their behavior has been exploited in its treatment.

Environmental Studies

The broad questions addressed in the environmental area are primarily those questions of determining chemically the fate or consequences of various materials that are introduced into our environment. Synchrotron radiation provides the opportunity to model the upper atmospheric environment with respect to solar irradiation and permits the study of the reaction of various chemicals with ozone. Necessary to these studies are gas-phase absorption, ionization, and dissociation measurements, which are facilitated by the continuum properties of synchrotron radiation.

Of relevance to pollution control is information concerning the local environment and chemical state of atoms of species that are pollutants or are normal constituents of materials that have been polluted. Through the use of synchrotron light, obtaining such information is greatly facilitated. For example, applying x-ray absorption spectroscopy to ascertain the chemical state of dilute metal impurities, such as lead or zinc, in samples taken from the atmosphere will elucidate the mechanisms of dispersal and, in turn, aid in developing methods of control. The identification, location, and determination of the chemical state of impurities on the surface of a solid, as well as the chemistry of the surface atoms in proximity of the impurity, will advance understanding of the effects of air and water pollutants on the properties of the material in question, e.g., corrosion associated with atmospheric sulfur on metal and mineral surfaces.

CONCLUSION

The Panel concludes that synchrotron radiation research will continue to expand and to have a major and unique impact on a wide variety of scientific and technological disciplines. It has been reasonably straightforward to describe the effect of synchrotron radiation on various currently practiced techniques and thus on the fields and frontiers affected by those techniques. While it is impossible to predict with certainty, the most exciting things are probably yet to be discovered, since synchrotron radiation represents a dramatic increase in experimental capability and since the research will be carried out in a very strongly interdisciplinary environment.

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4

PROJECTED UTILIZATION AND NEEDS FOR SYNCHROTRON RADIATION THROUGH 1986

INTRODUCTION

In order to assess the national needs for synchrotron radiation facilities it is necessary to project the growth in the use of such radiation for no less than ten years. This is about the time required for a new facility to be built and brought to full utilization. If, however, the scale of future facilities (which must be planned now) is to be estimated on the basis of scientific needs of the next decade, the circular nature of the problem must be recognized at the outset. Future demands for the use of synchrotron radiation obviously depend upon the availability of facilities and their geographical distribution. It is essential, therefore, to define the criterion for meeting future needs.

The Panel has taken the national needs for facilities to be those required for this country to exploit *the most promising* scientific and technological opportunities provided by synchrotron radiation. This is a subjective and judgmental criterion. However, it is clearly not the same as defining needs to be those that meet the total possible demand for facilities. With the latter criterion, utilization is not constrained by the availability of facilities. The Panel accepts as inevitable (and perhaps desirable) that facilities remain the principal constraint on the use of synchrotron radiation. At the same time, the Panel recognizes the need for careful planning for the next decade to ensure that this constraint does not unduly limit scientific and technological progress.

Projecting scientific and technical opportunities a decade ahead is very difficult in the early growth period of a promising new technique. It is especially difficult for fields affected by synchrotron radiation because there

is such a wide range of applications. For most facilities (e.g., oceanographic ships or telescopes) it is possible to define a specific population within the scientific community and to say with confidence that they will be the only scientists who will employ the tool over the next ten years. Such boundaries are not easily defined in the case of synchrotron radiation. Our present identifications of the most relevant scientific groups are based on present work. We are confident, however, that new and important applications not here identified will emerge long before ten years have passed.

In spite of the fact that most of the uncertainties about future projections of scientific opportunities of synchrotron radiation would tend to increase estimates of fugure needs, the Panel has deliberately taken a *conservative* position in estimating the future needs. In estimating growth we have not postulated any new or unknown applications. We have not assumed, as can in fact be persuasively argued, that a few of the present applications of synchrotron radiation can have such a dramatic impact on an important field that scores of new research groups will require access to the radiation within the decade.

The Panel estimated the scientific and technological needs for synchrotron radiation in an essentially judgmental way. Although opinions regarding future growth were widely sought from knowledgeable scientists, the Panel rejected at the outset making a survey of the scientific community in order to identify future users. Such a broad survey, we believed, would not be translatable with meaningful numbers. Our approach was first to assemble as much evidence as possible bearing on the future utilization of synchrotron radiation. This evidence included the present utilization and demand on synchrotron radiation facilities, recent growth in publications using synchrotron radiation, and an examination of the scientific and technical areas most affected by synchrotron radiation. On the basis of this evidence the Panel tried to identify the most promising scientific and technical areas that will be most affected by synchrotron radiation. The Panel then made numerical estimates of the future users of radiation who would be attracted by these new opportunities. These numbers were then tested by other criteria for their reasonableness; for example, what fraction of the number of people working in the field did our number represent? Naturally, such predictions lead to a range of numbers. In every case, we have used the more conservative (i.e., smaller) end of the estimated ranges.

TABLE 1 Minimum Predicted Growth in Synchrotron Radiation Facilities

	Present (Dec. 1976)		1986	
Use	Users	Stations	Users	Stations
X radiation	85	7	675	~60
XUV	120	16	480	~40

It is necessary for our purposes to translate the number of future "users" into the number of experimental stations at a facility that would be required to meet their needs. We define a "user" as an advanced-degree, U.S.-based scientist who is directly involved with experiments requiring the use of synchrotron radiation. We define a "full-time user equivalent" as a user or group of users who result in one man-year of experimental research per year using synchrotron research. We have made the somewhat arbitrary definition that three average users are equal to one full-time equivalent user (FTE). This approximately represents the current nature of synchrotron radiation usage. In order to translate the number of FTE's into the required number of experimental stations, we have used the ratio of 4 full-time users to 1 experimental station. Thus we have postulated a factor of 12 between the number of users and the number of stations.

Using this formula, a summary of our predictions for the minimum need for synchrotron facilities over the next decade is given in Table 1. As can be seen from this table the prediction is for a need of approximately 8 times more stations producing x radiation and 3 times more XUV stations than now exist. This conclusion will be discussed at length in the following subsections.

Utilization of Synchrotron Radiation Sources as of January 1, 1976, and Indicators of Growth

At present, there are three electron storage rings and one synchrotron that are serving as sources of synchrotron radiation for research. These are SURF II (240-MeV storage ring) located at the National Bureau of Standards in Gaithersburg, Maryland; the Cornell Synchrotron (15 GeV); TANTALUS I (240-MeV storage ring) operated at the

University of Wisconsin; and SPEAR (4-GeV storage ring) located at Stanford as part of the Stanford Linear Accelerator Center. Two facilities at electron storage rings--the Synchrotron Radiation Center (SRC) at Wisconsin and the Stanford Synchrotron Radiation Project (SSRP)--are funded by the National Science Foundation, and the NBS facility is funded by the Department of Commerce. Two of the facilities (NBS and SRC) are dedicated to synchrotron radiation use. At two of the facilities (SSRP and Cornell), the synchrotron radiation research is parasitic on high-energy physics research.

The physical location as well as the available energy spectrum has influenced the number and type of users (defined as advanced-degree scientists who have utilized synchrotron radiation as part of their research efforts). SURF (about 16 VUV users) was the first facility, and publications based on its use started in 1962. It has primarily drawn scientists from the Washington, D.C., area. The Cornell synchrotron effort has been small (about 5 x-ray users) and has drawn primarily on local users. The SRC group at Wisconsin started publishing in 1969, and as of January 1976 had an active XUV user group of 62. These have come from about 18 states--primarily Wisconsin (17), Illinois (10), New York (5), New Jersey (5), and Iowa (4). The effort at SSRP is the most recent, and that group started publishing in 1974. As of January 1976, the SSRP had about 80 active users (about 45 x-ray and 35 XUV) with the bulk (41) from California. There are users from eight other states--primarily New Jersey (11), Washington (10), Oregon (4), Tennessee (2). These figures exclude graduate degree candidates and technicians.

One source for evaluating current interest is provided by the multidisciplinary User Group meetings held at the two NSF-funded facilities, the Wisconsin SRC and the California SSRP. The eighth annual meeting at the Wisconsin SRC drew 86 national scientists and graduate students in October 1975, whereas the second annual meeting for SSRP at Stanford one day later drew another group of 99 national scientists and graduate students. In addition, about 20 foreign scientists attended both meetings. The mailing lists for these two annual meetings (eliminating duplicates) contain the names of 494 advanced-degree scientists, 154 other scientists (including graduate students), and 167 foreign scientists.

In order to examine the growth of the sciences that utilize synchrotron radiation on a worldwide basis, we have examined the number of refereed publications for

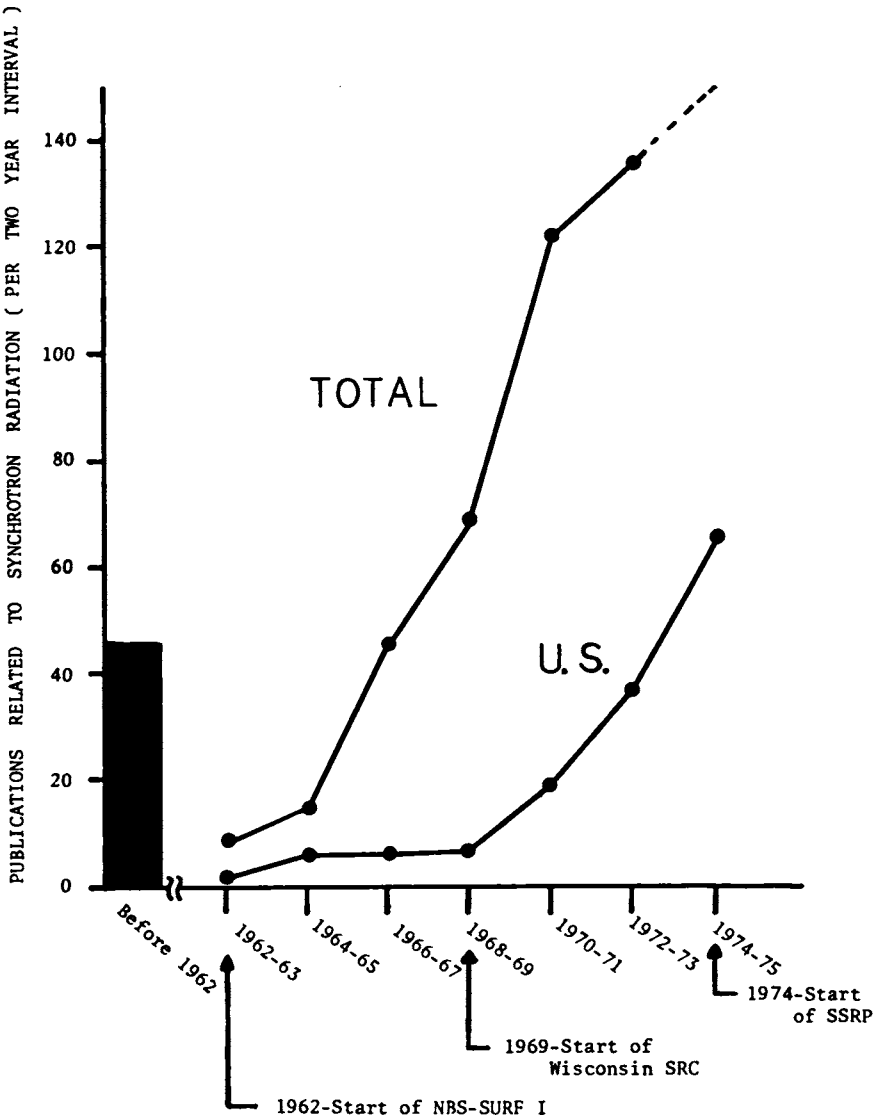

FIGURE 8 Number of publications as a function of time.

which synchrotron radiation was used. This growth is displayed in Figure 8, where the data of Marr *et al.*[1] are utilized to give the world output through December of 1973, and data from the SURF, SRC, and SSRP facilities are used to yield the U.S. contributions. As can be seen from these data, the publication rate is strongly accelerating and the U.S. contribution is rising. The lag of U.S. science behind foreign efforts can be attributed entirely to a lag in the availability of U.S. synchrotron radiation facilities. The date on which each facility began operation is noted on Figure 8. For the years 1962-1974, essentially all the publications cited from the United States are concerned with results from XUV experiments; the results from x-ray experiments began to appear in the early part of 1975, with a significant number in that year (5) reporting results on x-ray absorption-edge fine-structure (EXAFS) measurements. Not shown on the graph are the publication results for 1976. Preliminary reports for the first 3 months of 1976 show approximately 21 papers appearing or accepted for publication (12 with x-ray results and 9 with XUV results).

Although publications data are indicative of growth over a number of years, they do not show what research areas can be expected to produce that growth. Experience shows that research interest in a given area is stimulated considerably by a few publications that indicate to others the power of a given approach to solving problems. Examples of U.S. papers of this type are those of Eastman and co-workers on the examination of surface adsorbed materials by variable wavelength photoelectron spectroscopy[2]; core-level electroreflectance and thermoreflectance modulation spectroscopy as described by Lynch and others[3]; fine structure on the photoelectron spectra of silicon near the $L_{II,III}$ core level by Brown and co-workers[4]; the recent EXAFS measurements by Eisenberger and co-workers at Bell[5] and by Shulman *et al.* and Sayers *et al.*[6]; and the angular resolved photoemission studies and photoemission measurements with either constant initial or final state energies by Lapeyre and co-workers.[7]

Sources of Growth and the Science Areas Affected by the Availability of Synchrotron Radiation Sources

We wish to identify the science areas that will be affected by the availability of synchrotron radiation and to estimate the number of expected users in these areas. We can

categorize users into three groups. Some of them will be full-time in the sense that their primary commitment is to studies for which the synchrotron radiation is essential. Another group will be part-time in that several techniques might be brought to bear on a particular problem, and synchrotron radiation would be one of these. Finally, there will be a very large third group that would be interested in the results of one or several samples or a single experiment. In principle, at least, this latter group could include up to one fourth of all the scientists in this country! In order to make meaningful projections for facility use, however, we have chosen to estimate the numbers of users in only the first two categories. The number we seek is the full-time-equivalent user. As noted earlier, we define the number of full-time-equivalent users (FTE) as one third of the numbers of scientists whom we can identify in the first two groups mentioned above.

Listed below are the areas of science that we have previously identified to be strongly influenced by the utilization of synchrotron radiation. Also included are the estimates of FTE users of the two energy regions of synchrotron radiation. As previously discussed, these energy distinctions are made primarily on a historical basis.

Life Sciences and Related. Thousands of conventional x-ray tubes are in use in development and research laboratories in the United States. The American Crystallographic Association (ACA) has approximately 1800 members, and of these about 300 belong to a division concerned with biomolecular crystallography. Among the crystallographers there are at least 50 or more groups that have computer-controlled diffractometers for automated collection of intensity data. There is another subgroup of the ACA with about 75 members who use low-angle scattering in their research on chemical or biological systems (for example, in studying such structures as biopolymers, porous catalysts, and amorphous materials). The research experience at the German synchrotron DESY and at the SSRP strongly suggests that the research of this group with interests in low-angle scattering experiments will be strongly affected. The synchrotron source could also prove to be invaluable for solving certain classes of problems of interest to the large-angle scattering community as a whole. It is also clear that many scientists using diffraction techniques may employ x-ray absorption spectroscopy to complement their diffraction research. The

several hundred scientists who are now using such techniques as nuclear magnetic resonance and electron spin resonance in their studies of biological function will find that x-ray absorption spectroscopy data will be of substantial value to them. X-ray absorption measurements at other absorption edges (e.g., that of C at 285 eV) may also be employed. In applications requiring microscopy, the advantages of using x rays from synchrotron radiation have also been demonstrated.

We summarize all of these uses into a prediction of approximately 100 FTE users of x rays by 1986 and 10 FTE users of XUV.

Surface and Bulk Materials Science. Earlier we summarized just a few of the problems in surface science and solid-state science that are amenable to examination using photon physics. In these areas of science there are laboratory sources that can be employed for some of the research. Thus there is a problem attempting to determine when synchrotron radiation will provide the only source for a given experiment. However, growth in the use of synchrotron radiation in this area has been strong. We note that under titles "Electron Spectroscopy I. Ultraviolet Photoexcitation and Electron Spectroscopy II. X-ray Photoexcitation," the Fundamental Reviews compiled every two years by *Analytical Chemistry* list 315 papers in the 1972 review, 760 in the 1974 review, and 923 in 1976. The reviews in 1974 and 1976 also included electron excitation, but these papers are excluded from the quoted numbers. These numbers may increase even more if carbon-edge EXAFS measurements at ~285 eV become feasible for carbon compounds and polymers.

A striking change in research techniques for probing surface properties is evident from the papers presented at the Solid State Section of the American Physical Society national meetings. In 1970 and 1971, there were approximately 10 papers presented each year, and nearly all were concerned with low-energy electron diffraction (LEED). In 1973, there were 53 papers, of which 5 reported work using photon probes and none reported work on molecules. By contrast, in 1976, there were 167 papers, of which 49 involved photon probe studies and 15 involved atom, molecule, or molecular cluster studies. A similar pattern exists for other major national meetings involving surface science such as the Physical Electronics Conference and the fall American Vacuum Society meeting.

With respect to future x-ray utilization, we note that about 50 of the group concerned with low-angle scattering in the American Crystallographic Association are mostly chemists and work with lamellar systems, polymers, etc. There are also approximately a dozen diffuse scattering and inelastic scattering groups concerned with materials problems. We estimate that of the 1000 or more laboratories that use x-ray topography for diagnostic purposes, approximately 50 or more would benefit by the use of synchrotron radiation. The use of EXAFS in various materials science disciplines such as solid-state physics and chemistry will be part of a large number of research programs. Inelastic x-ray scattering experiments--Raman, Compton, and nonlinear--are expected to become active when the next generation of synchrotron sources becomes available.

For these scientific areas we estimate 95 FTE users of x rays and 95 FTE users of XUV by 1986.

Molecular and Atomic Studies. New machines or new configurations for synchrotron radiation promise increases in intensity of roughly a factor of 100 over the whole XUV region where molecular bonding interactions can be explored. This improvement in intensity and consequently in resolution should lead to an addition of at least 30 (10 FTE) or more users to the 20 (7 FTE) already now working with atomic and molecular problems. If the intensity becomes high enough, more extensive ion-molecular reaction studies with control of the reactant ion energy state will be performed, and another 10 (3 FTE) or more users could become involved.

Fluorescence measurements using the storage-ring time structure have been shown to be superior to flash lamps by work at ACO in France. Biological applications of fast photon fluorescence are possible. Within the last few months, a new society called the "Inter-American Photochemical Society" was formed as a consequence of discussions at the VIII International Conference on Photochemistry held in Edmonton, Canada, in August 1975. The initial responses to such a new society would appear to suggest that there will be an initial membership of 400 or more. Approximately 10 percent of these, or 10 FTE users, may be involved in XUV work by 1986.

In the application of condensed-phase absorption to biology, several groups are interested in utilizing the polarization properties, and particularly the circularly polarized light expected from the helical wigglers (see

page 72). The future here is difficult to predict because the first experiments using circularly rather than elliptically polarized light have not yet been performed. It is anticipated that some time-dependent response will be indicative of the conformational properties of large molecules, and that the effect of various chromophores on the observed circular dichroism in the XUV will lead to improved correlations with structure.

We conclude that within these areas there will be ~10 FTE users of x rays and 25 FTE users of XUV by 1986.

Technological Applications. There is a broad range of important technological applications for synchrotron radiation as described in a previous section.

An estimate of future user demand through 1985 for technological applications is difficult because a significant user base for a number of the new applications does not exist and because the present U.S. facilities are not designed to accommodate nearly all of the previously mentioned applications. The number of users can be expected to be affected by certain breakthroughs in demonstrated utility of synchrotron radiation. It might also be expected that certain applications, such as the successful generation of x-ray lithographic integrated circuits, would stimulate certain industries to build their own storage-ring sources.

We estimate that there will be about 20 FTE users in the x-ray region and about 30 users in the XUV photon energy region below 1000 eV at national synchrotron radiation facilities. These estimates, which are uncertain for reasons mentioned above, are based on the assumption that very marginal if any arrangements will exist for industrial work of a proprietary nature. If proprietary work at such facilities were provided for, the technological potential of synchrotron radiation would be more fully exploited and the number of users could increase dramatically.

Summary, Predictions, and Station Requirements. In the previous sections we have attempted to identify the scientific areas where the users of synchrotron radiation will originate. In terms of facility requirements, however, we need a prescription for converting between FTE users and the required number of fully equipped stations that should be available to support the research effort of the FTE users. Because these stations would have photons available for the equivalent of at least 10-12 hours per day, 6 days per week, and because few experimenters

TABLE 2 User and Station Projections through 1986

Use	Users	FTE Users	Stations
X ray	675	225	~60
XUV	480	160	~40

can handle the data that would be produced on this basis continuously, we have chosen the conservative factor of 1/4 to convert the number of FTE users into the number of stations at the synchrotron radiation facilities. Table 2 summarizes the previous projections. It should also be kept in mind that for sophisticated specialized experiments, the apparatus will not be in continuous use and yet no one else will be able to use the station.

Factors That Will Increase or Decrease Utilization

Certain features of national synchrotron radiation facilities can significantly affect the ease with which the appropriate science can be performed and will affect the quantity and quality of research that will be accomplished. Because these features should be recognized in any plans for future facilities, we list some of the pertinent ones here. We note that there is a limited group of scientists in this country that is knowledgeable about the properties of synchrotron radiation, how to exploit it through appropriate instrumentation, how to avoid some of the attendant problems that arise from the continuum properties (scattering, order overlap, etc.), how to maintain vacuum environment, and how to handle the high power and radiation intensities that are available. There is a much larger group that knows a great deal about a particular sample, instrumental system, or technique but is not familiar with the problems of coupling an experiment to a radiation port. Finally, there is the remaining group, which is primarily sample-oriented, where the results from several samples constitute the study. This latter group may not be equipped with any instrumentation. We believe that all three groups should be accommodated at a national facility.

For all three of these groups, it is essential that there be an active and vigorous group of qualified scientists located at the facility, or in close proximity,

so that implementation of new ideas and experiments could be expedited. The regions of the spectrum served by synchrotron radiation lack detailed information on filters, suffer from radiation losses in monochromator designs, and need innovations for detectors and improved methods for data collection and handling. The data on filters and reflectivities are especially critical when both the x-ray radiation and the XUV radiation must be separated for an XUV experiment. In order to expedite support of these scientists and the users, it is essential that the support shops in the machine, electrical, mechanical, and computing areas be of exceptionally high quality and that there be access to libraries and other sources of information to consult.

The problem of dedicated versus parasitic operation is critical to meeting the needs of users. Only when scheduling and machine properties are tuned to meet the needs of the synchroton radiation users is there the optimal match of experimental conditions to answer the desired fundamental questions. The experiences of our European colleagues and others who operate in a mode parasitic to high-energy physics programs give clear evidence that a parasitic mode of operation for future major national facilities is not adequate, since it cannot produce the desired flexibility in experiments and will impose other restraints and conditions that slow research progress. This is particularly true for the experiments that cannot employ windows and must be directly connected to the ring vacuum.

One of the most critical needs for most users is the availability of radiation ports that are fully instrumented through a monochromator stage. A variety of these should be available from the relatively low to the very high-resolution variety. In the x-ray area, this would include instrumentation for moving the samples as well as detection equipment for recording the signal responses.

Because the number of storage rings will undoubtedly always be smaller than the needs or desires of research groups located throughout this country, consideration should be given to geographical distribution and to the difficulty of mounting experiments at remote sites. The costs of transporting equipment and personnel, the costs of subsistence and housing at the site, and the costs of emergency repairs and modifications will inhibit or prevent many users from exploiting the radiation capabilities. If the costs of all of these services exceed the basic grant of a potential user, the experiment will not be

performed unless one or more of the following is provided: additional travel grants, inexpensive site housing, technical support for data collection at the facility, or collaborative arrangements with established users.

If collaboration becomes a predominent mode, the level of support provided by the facility for a full-time user will make a difference in the numbers of users. If experimental chambers can be provided with flexibility in equipment, pumps, counters, and other instrumentation, then collaboration is enhanced.

In the technology applications area, a provision for conducting proprietary research and assigning associated costs appropriately at a national facility will have a decided influence on the number of industrial users who will be encouraged to employ this approach to their research and applications problems. If this group is to be encouraged, a simple device for segregating proprietary research would have to be explored. Finally, one of the factors most conducive to research at a national facility is the interaction of scientists from various disciplines.

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5
MACHINE CONSIDERATIONS AND OPTIONS

STORAGE RINGS AS RADIATION SOURCES

In the world today, there are more than 20 electron synchrotrons and storage rings being used or readied for use as synchrotron radiation sources.[1] (See Table 3.) In addition, there are several proposals and design studies for new facilities. The large increase in worldwide interest and activity, particularly abroad, in synchrotron radiation as a research tool is due largely to the successful recent experience with *storage ring sources*. An electron storage ring is an extremely useful source of electromagnetic radiation. The attributes of such a light source that most interest scientists are

1. Continuous spectral distribution from the infrared to the x-ray region
2. High intensity and brightness
3. Great stability with known flux--a secondary standard
4. Radiation produced in ultrahigh vacuum
5. High polarization
6. Subnanosecond pulse structure

Stripped to its essentials, an electron storage ring consists of a high vacuum chamber that threads through the bending and focusing magnets that guide the electrons. Figure 9 is an artist's sketch of an intermediate (1-GeV) energy storage ring stripped of all of its magnets but one. Not shown is an injection system, which inserts 10^{10}-10^{12} electrons into stable orbits. These electrons circulate for hours--the decay rate is determined by occasional encounters with residual gas atoms. The electrons are accelerated (centripetally) by the magnetic

TABLE 3 Synchrotron Radiation Research Facilities--1976

Laboratory	Energy (GeV)	Current (mA)	Bending Radius (m)	Critical Energy (keV)	Remarks
Storage Rings					
DORIS, Hamburg, Germany	1.5-4.0	~250	12.2	11.6	
SPEAR, Stanford, U.S.A.	1.5-4.0	5-35	12.7	11.1	≥300 mA available in multibunch mode
VEPP-3, Novosibirsk, U.S.S.R.	2.25	200	6.0	4.2	
VEPP-2M, Novosibirsk, U.S.S.R.	0.67	100	1.22	0.54	
ACO, Orsay, France	0.54	100	1.1	0.32	Dedicated to synchrotron radiation
INS-SOR, Tokyo, Japan	0.30	100	1.0	0.059	Dedicated to synchrotron radiation
Tantalus I, Wisconsin, U.S.A.	0.24	100	0.64	0.948	Dedicated to synchrotron radiation
SURF II, NBS, Washington D.C., U.S.A.	0.24	~50	0.84	0.036	Dedicated to synchrotron radiation
DCI, Orsay, France	1.8	500	3.8	3.4	
PACHRA, Moscow, U.S.S.R.	1.3	10-300	4.0	1.1	
Daresbury, U.K.	2.0	500-1000	5.55	3.2	In construction operation 1980, dedicated
ADONE, Frascati, Italy	1.5	60	5.0	1.5	Synchrotron radiation beam lines in construction
Synchrotrons					
Cornell, U.S.A.	12	2	100	38	
DESY, Hamburg, Germany	7.5	10-30	31.7	29.5	
ARUS, Yerevan, U.S.S.R.	6.0	20	24.6	19.5	
NINA, Daresbury, U.K.	5.0	40	20.8	13.5	Terminates operation in 1977
BONN, Germany	2.5	30	7.6	4.6	
INS-SOR, Tokyo, Japan	1.3	30	4.0	1.22	
Frascati, Italy	1.1	10	3.6	.82	
C-60, Moscow, U.S.S.R.	0.68	10	1.6	.44	
BONN II, Germany	0.5	30	1.7	.16	

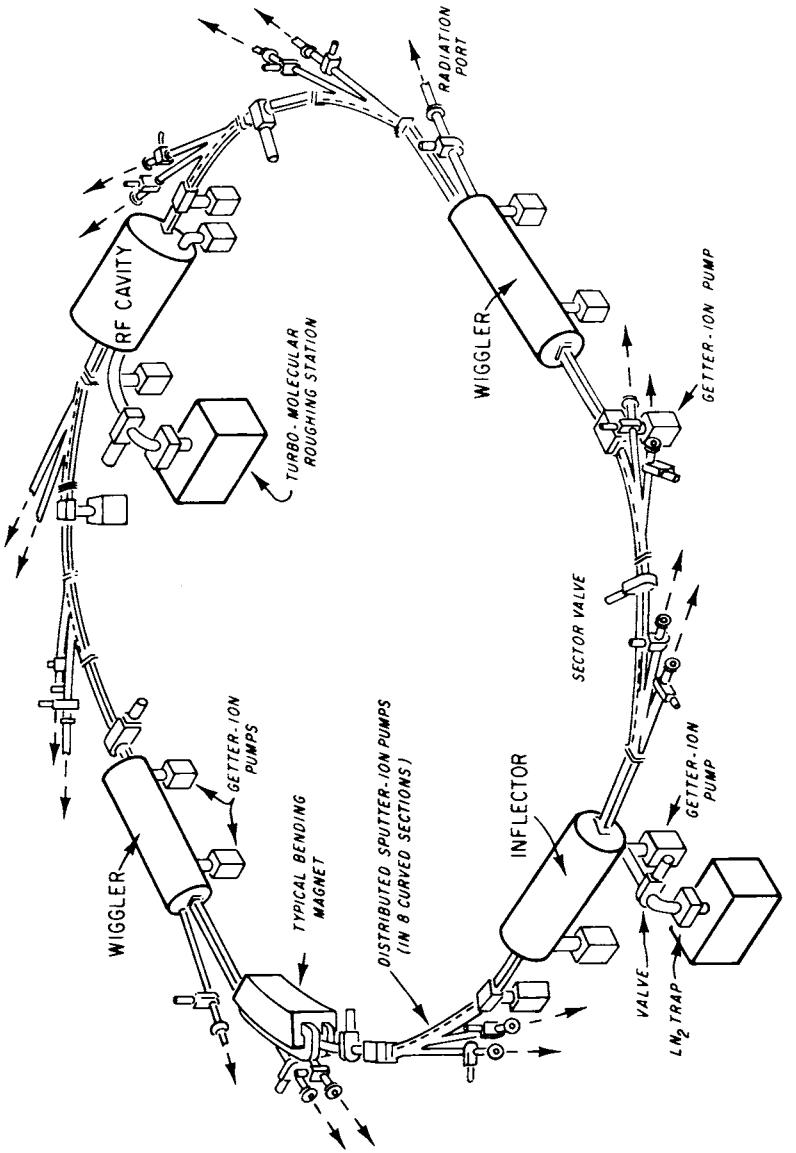
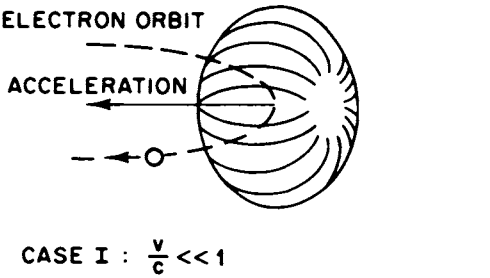


FIGURE 9 Isometric of a small, 0.5-1.0 GeV, high-current storage ring as utilized for synchrotron radiation. Only one bending magnet is shown. Notice the several tangent ports for radiation.

fields of the ring bending magnets or special insertion magnets and emit synchrotron radiation wherever this acceleration occurs. The radiation from each electron is sharply collimated in the forward direction, as indicated in Figure 10. Tangent vacuum pipes convey the synchrotron radiation to the research apparatus. The energy lost by the electrons is continuously supplied by a radio-frequency accelerating system, and provision must be made (especially in the case of large machines) to absorb the radiation on cooled surfaces. The emission of synchrotron radiation can be used to shrink the dimensions of the circulating electron beam to very small values (less than a millimeter in the vertical direction). The horizontal



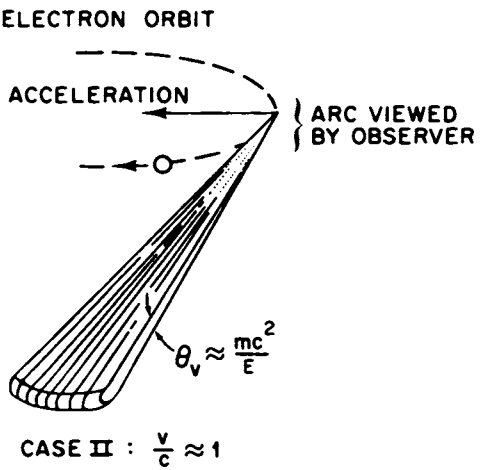


FIGURE 10 Radiation pattern from electrons moving in a circular orbit.

beam size is limited by quantum fluctuations in the emission process; and in a given machine, the limiting size increases with energy. The vertical beam size is largely determined by coupling between horizontal and vertical oscillations. In practical storage rings, the circulating electrons are confined to one or more bunches that can be only a few centimeters long. In these cases, the radiation as viewed in the laboratory is pulsed with a subnanosecond time structure.

Thus, storage rings generate very intense, stable, sharply pulsed, highly collimated, and polarized radiation from a high vacuum source of small transverse dimensions. Although electron synchrotrons offer some of these same properties, they lack stability because of their cyclic nature. Furthermore, the high-energy radiation environment at a large synchrotron is another complication. Because of their clear superiority and advanced state of development,[2] storage rings are the only sources now being considered for new synchrotron radiation facilities.

It is significant that almost all of the machines now in use as synchrotron sources are high-energy physics machines. Indeed, only one machine now in operation (the Tokyo 300-MeV storage ring) and one under construction (the Daresbury 2-GeV storage ring) have been designed from the start as synchrotron light sources. In most of the other machines, the synchrotron radiation research is still a secondary program on machines designed for high-energy physics research. Machine schedules and operating conditions are largely determined by high-energy physics requirements. This results in severe limitations on the synchrotron radiation research program. For example, colliding-beam currents (due to the limitations of beam-beam interactions) are less by a factor of 5 to 50 than possible single-beam currents. Also, the collision of beams enlarges the vertical beam size, reducing source brightness.

Thus the full potential of a major storage ring ($\geq$0.7 GeV) dedicated to synchrotron radiation research has not yet been realized anywhere. Based on experience with existing facilities, it may be confidently predicted that such a facility would offer significantly greater research opportunities, particularly if it were a machine designed for synchrotron radiation use from the start. Not only will such a machine be operated at energies and currents that optimize the synchrotron radiation research program, but the basic design of the machine can offer other advantages. For example, the bending magnet design

can be such as to facilitate the attachment of tangential vacuum pipes, and the magnet lattice can be selected to produce the highest source brightness (smallest electron-beam emittance).

In addition, devices for enhancing the production of synchrotron radiation (wigglers) offer the promise of further extension of research capabilities. Wigglers are magnetic structures inserted into storage ring straight sections. The transverse wiggler has a high field (~50 kG) and possibly several alternating polarity sections. It significantly extends the synchrotron radiation spectrum to higher energies and also increases the flux at all energies. The helical wiggler has a moderate field and produces quasi-monochromatic tunable synchrotron radiation of very high intensity. The characteristics of storage rings and wigglers designed specifically for synchrotron radiation are described in more detail later in this chapter.

PROPERTIES OF THE RADIATION

The basic properties of synchrotron radiation, compared with other sources of electromagnetic radiation, may be summarized as follows.

The normal radiation pattern of an electron in circular motion at low energy is a rather nondirectional dipole radiation pattern. At relativistic energies this pattern is folded strongly into the forward direction (see Figure 10) so that the radiation is emitted in a flat pancake in the plane of the electron orbit. The theory of synchrotron radiation[3] shows that for emission at short wavelengths near the peak of the spectrum, the root-mean-square divergence angle of the radiation cone is given by

$$\psi_{rms} \sim 1/\gamma, \qquad (1)$$

where γ is the ratio of energy to rest mass for the electron, $\gamma = E/m_0c^2 = 1957\ E$ (GeV).

The narrow cone sweeps around the entire closed orbit of a storage ring, but any single observer can "see" only a small fraction of the 2π of arc. Consequently, the total radiation pattern is like a flat pancake with a mean thickness of beam height given by Eq. (1). During one turn, an electron radiates

$$\delta W = 2.66 E^3 H \text{ keV} \qquad (2)$$

of energy (in this report electron energy E will be in units of GeV, orbit radius R in meters, magnetic field H in kOe, and photon energy $h\nu$ in electron volts). At the energies considered here the velocity of an electron, to six or seven significant figures, is equal to the velocity of light. If a storage ring contains I amperes of circulating electrons the power radiated is then $P = 2.66IE^3H$ in kilowatts, and the radio-frequency system must supply this power. Examples of the power at 1 A and an arbitrary magnetic field of 10 kOe are

Energy (GeV)	0.7	2	4	8
Power (kW)	9	211	1703	13,600

These numbers indicate the limits approached in rf power and vacuum chamber cooling, for large high-energy rings, under large circulating current conditions.

The spectrum of photon emission is most readily specified by use of a parameter called the critical wavelength, λ_c, or by the corresponding critical energy $h\nu_c$ (eV) = $12400/\lambda_c$ (Å). Half of the photons are emitted at wavelengths shorter than about $12\lambda_c$, and half the radiation power is at wavelengths shorter than λ_c. The critical wavelength is given by

$$\lambda_c = 5.59R/E^3 = 186/HE^2 \text{ Å}. \qquad (3)$$

It turns out that the photon flux in the synchrotron continuum integrated over all vertical angles is given by a particularly simple expression as follows:

$$N_k(\lambda) = k\lambda F_1 \text{ photons/(sec-mA-mrad arc)}, \qquad (4)$$

where $k = \Delta\lambda/\lambda$ and the spectral function F_1 is a function only of $(\lambda_c/\lambda) = (h\nu/h\nu_c)$. This function, shown in Figure 11, falls faster than exponentially below λ_c, peaks at 3-4 times λ_c, and decreases slowly at the long wavelengths.

Equation (4) indicates that the flux per milliradian at a fixed value of λ/λ_c increases as γ. The function F_1 drops off rather slowly as λ is increased above λ_c. Actually, high-energy machines of large radius emit somewhat greater flux per unit horizontal angle at long wavelengths than small machines. The asymptotic form[3] for the spectral function F_1 shows that at $\lambda >> \lambda_c$ the flux $N_k(\lambda)$ can be written in a form that is independent of γ but increases as $R^{1/3}$.

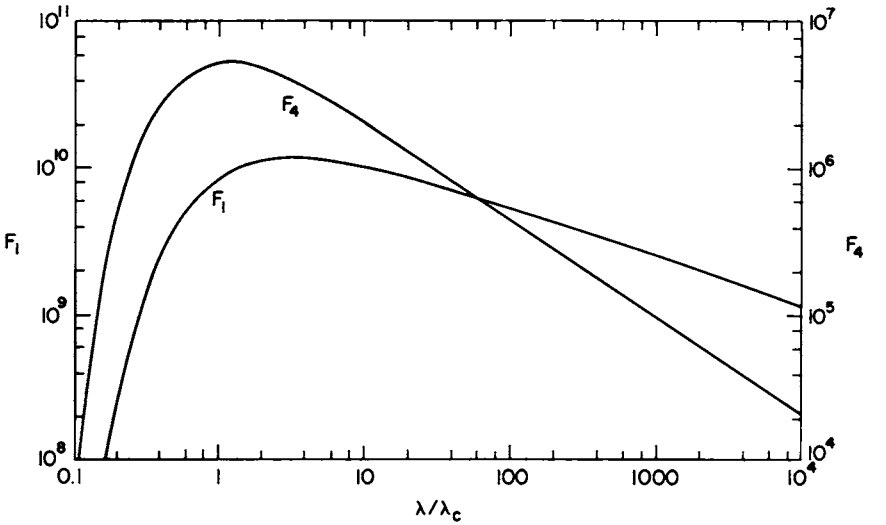


FIGURE 11 Flux functions. $F_1(\lambda/\lambda_c)$: Number of photons/(sec-mA-mrad) at λ is given by $N(\lambda) = k\gamma F_1$, where $k = \Delta\lambda/\lambda$. $F_4(\lambda/\lambda_c)$: Number of photons/(sec-mA-mrad arc-mrad ψ) is $N(0,\lambda) = k\gamma^2 F_4$, where $k = \Delta\lambda/\lambda$ and $\psi = 0$.

Equation (4) gives the flux integrated over all vertical angles ψ. At $\psi = 0$ and wavelength λ the photon flux per milliradian of arc and *per milliradian* ψ is given by

$$\frac{dN}{d\psi} = k\gamma^2 F_4 \text{ photons/(sec-mA-mrad}^2\text{)}, \qquad (5)$$

where F_4 is another universal spectral function of λ/λ_c as shown in Figure 11. This equation shows that at fixed λ/λ_c the flux per mrad^2 increases as γ^2.

In the orbital plane, $\psi = 0$, synchrotron radiation is plane polarized. Out of the orbital plane and at long wavelengths $\lambda >> \lambda_c$, the radiation is increasingly elliptically polarized.[4] This could be important for experimental work such as broadband cyclotron resonance in the far infrared or dichroism experiments at various wavelengths. High-current storage rings should be useful sources of far infrared (100-1000 μm) radiation.[3,5] The angle ψ_{rms} increases at long wavelength, however, and collecting mirrors must be placed close enough to orbit to collect most of the vertical angle and an appreciable fraction of the horizontal angle.

Electrons in a storage ring are distributed about the central orbit both in position and angle. Source brightness is determined by the number of photons per unit angle *per unit area*. The net emission angle spread of the radiation in the vertical direction, $\sigma_{0y'}$, is related to the spread in emission angle by a single electron, σ_ψ, and the vertical angle spread of the electron beam $\sigma_{0y'}$, according to

$$\sigma_{0y'} = \sqrt{\sigma_\psi^2 + \sigma_{y'}^2}. \qquad (6)$$

To minimize $\sigma_{0y'}$ it is desirable to keep $\sigma_{y'} \lesssim \sigma_\psi$. The radiation angle spread σ_ψ is inversely proportional to γ and roughly proportional to the square root of λ/λ_c. Source area is determined by the cross-sectional area of the electron beam and the length of the source. The brightness relationships are complex, but, in general, it is desirable that electron-beam size and angular divergence be small and that the source length be short. These considerations indicate that the light-source storage ring should have as small an emittance (product of cross section and divergence) as possible.

ELECTRON STORAGE RING CHARACTERISTICS

Existing U.S. Synchrotron Radiation Facilities

At present, in the United States there are three storage rings and one synchrotron with synchrotron radiation research programs. Of these, two storage rings--Tantalus I at Wisconsin and SPEAR at Stanford--provide radiation for large numbers of national users. In this section we describe the capabilities of each of these facilities and note the possibilities for providing additional research capability. Table 4 summarizes the operating characteristics of the three U.S. storage ring sources, Tantalus I, SPEAR, and SURF II.

Tantalus I. The 240-MeV storage ring at the University of Wisconsin Synchrotron Radiation Center (SRC) at Stoughton, Wisconsin, has recently been improved with a new injector and a new vacuum chamber. As a result, the stored beam current increased from about 5 mA to about 150 mA.

TABLE 4 Characteristics of Existing U.S. Storage Ring Sources

	SURF II	Tantalus I (SRC)	SPEAR (SSRP) Low-Energy Operation	SPEAR (SSRP) High-Energy Operation
General				
Energy	0.24 GeV	0.24 GeV	1.5 GeV	4.0 GeV
Bending magnet field	9.5 kOe	12. kOe	3.9 kOe	10.4 kOe
Bending magnet radius	0.84 m	0.64 m	12.7 m	12.7 m
Geometrical radius	0.84 m	1.5 m	37. m	37. m
Injection energy	10. MeV	40. MeV	2250. MeV	2250. MeV
Orbital frequency	57. MHz	32. MHz	1.28 MHz	1.28 MHz
Beam lifetime	1-3[a] h	4-6 h	~6 h	~4 h
No. of tangential beam ports in use	3	8	2	
No. of experimental stations (end of 1976)	4	11	8[e]	
Annual operating budget (FY 76)	150 $K	390 $K	531 $K	
Power				
Typical operating current	4-8 mA	80-140 mA	5[c] mA	40[c] mA
Total power consumption	300 kW	115 kW	2000 kW	8000 kW
Radiation in Arcs				
λ_c from bending magnets	340 Å	257 Å	7.2 Å	1.1 Å
Flux at λ_c per sec-mA-mrad at 0.1% $\Delta\lambda/\lambda$	4×10^9 photons	4×10^9 photons	2.4×10^{10} photons	6.4×10^{10} photons
Beam size at 2σ				
Horizontal	1.6 mm	0.72 mm	1.6 mm[d]	4.2 mm[d]
Vertical	0.06 (0.4)[b] mm	0.18 (0.3)[b] mm	0.8 mm[d]	2.2 mm[d]

[a] Lower lifetime with very small beam height--long lifetime with enlarged beam.
[b] Figure in parentheses indicate when beam is enlarged to increase lifetime.
[c] These figures are for single-bunch colliding-beam operation--as much as 225 mA has been achieved in multibunch mode during tests at injection energy 2.25 GeV.
[d] Figures given are for tangential beam-source point during colliding-beam operation. Source sizes would be reduced by a factor of ~3 in single-beam mode. Other source points with smaller source size exist.
[e] Includes one port with 3 x-ray stations to be completed in 1976.

Although originally built to study the physics of colliding beam machines, it has been operating solely as a synchrotron radiation source since 1967.

This highly successful national facility operates the year around on daily 10-hour shifts, providing intense synchrotron radiation to as many as 11 simultaneous users on 8 tangent beam ports, which provide an average of 50 mrad horizontal angle each. The critical energy is 48 eV (258 Å). Experiments have been carried out from the infrared up to 250 eV (50 Å).

Although close to the maximum number of tangent beam ports are already installed, improved research capability is possible on Tantalus I with higher current and increased source brightness, as well as with new improved monochromators. This facility could also serve a larger community of research workers with an increased operations schedule.

SPEAR. The Stanford Synchrotron Radiation Project (SSRP) has been in operation since May 1974, providing radiation from the uv to x rays of 40-50 keV to five simultaneous users sharing a single tangent beam line. SPEAR is one of the most powerful electron storage rings now in operation. Its stored beam energy varies from 1.5 GeV [critical energy = 0.58 keV (21 Å)] to 4.0 GeV [critical energy = 11 keV (1.1 Å)].

The original port provides 11.5 mrad of radiation to two XUV users and three x-ray users simultaneously. A beam line on a second port will be completed in 1976 providing 20 mrad that will accommodate four or more simultaneous users. Three x-ray monochromators are now under construction for this line.

Ten additional beam ports could be added while maintaining compatibility with colliding-beam operation. Also, a total of 27 beam ports is possible if SPEAR were operated as a fully dedicated source.

The present mode of operation of SPEAR calls for simultaneous running of colliding-beam physics (operating costs paid for by the high-energy physics program) and symbiotic synchrotron radiation research during the scheduled high-energy physics running time. Also, up to 5 percent additional running time is available for dedicated synchrotron radiation research if operation costs are fully paid by SSRP. Operation for synchrotron radiation research could increase to 50 percent of the total available SPEAR operations time when PEP (the LBL/SLAC 18-GeV colliding-beam storage ring) is operating for high-energy physics for 50 percent of its available time (expected in 1980).

TABLE 5 Typical Electron Storage Ring Characteristics

Characteristics	Primary Objective		
	XUV	X-Ray + XUV	
General			
Energy (GeV)	0.6-0.9	1.9-2.5	3.9-4.5
Bending magnet field (kOe)	12	8-12	9-12
Bending magnet radius (m)	1.7-2.5	6.5-8.5	11-16
Geometrical "radius," $C/2\pi$ (m)	9	21	32
Orbit frequencies (MHz)	5.3	2.3	1.5
Building (ft^2)	125 × 150	200 × 200	200 × 200
Power			
Estimated max. current (A)	0.5	1	0.5
Total power consumption (MW)	1	3.5	5
Max. radiation in arcs (kW)	6	250	1000
Radiation power in 10 mrad, arc (W)	10	400	1600
Radiation power/cm^2 at 2 m, arc (W)	15	2000	16,000
Min. distance to first mirror (m)	1	2	4
Radiation in Arcs			
λ_c from bending magnets (Å)	18-30	2.5-6	~1 Å
Beam size at 2σ, H (mm)	~0.5	~0.5	~0.5
V (mm)	~0.08	~0.08	~0.08
Insertions			
Free length (m)	4	5	6
Transverse wiggler field (kOe)	20 (normal)	40	40
Wiggler λ_c (Å)	19	1	0.3
Insertion beam size at 2σ,			
H (mm)	0.3	0.15	0.3
V (mm)	0.015	0.01	0.02
(Flux per mrad at λ_c same as arcs)			
Helical wiggler λ at 2.5-cm pitch (Å)	130	14	4
Flux at λ_c/sec, 0.1% $\Delta\lambda/\lambda$ (photons)	2×10^{17}	4×10^{18}	7×10^{18}
(2.5-m helix, 2.5-cm pitch)			
Facility Costs			
Basic building and source ($ millions)	~3.5	~15	~25
Basic operating costs/year ($ millions) (exclusive of science support)	~1.0	~2.5	~3.0
Beam line and instrumentation[a] ($ millions) (total cost over ~5 years)	~4	~8	~8

[a] We have assumed 20 stations on the XUV ring and 40 stations on the x-ray rings, to be developed over ~5 years.

There have been several hours of machine studies of high-current single-beam mode of operation. This experience, plus analysis of the potential of SPEAR as a dedicated synchrotron radiation source, indicate that, if dedicated entirely to synchrotron radiation use, most of the characteristics of SPEAR would be similar to those described in Table 5, columns 2 and 3.

SURF II. The original 170-MeV synchrotron at the National Bureau of Standards was recently upgraded to a 240-MeV storage ring in a collaborative effort by the Physical Sciences Laboratory at the University of Wisconsin (which operates Tantalus I) and the NBS staff. The conversion was completed in 1975, and the new machine now operates regularly as a dedicated synchrotron radiation source with 4-mA stored current (up to 8 mA has been achieved). A research program is now being resumed, and three beam lines are installed. This facility could accomodate about 8 additional beam lines and serve a much larger community of users. The critical energy is 36 eV (345 Å), and thus usable flux is available to 160 eV (75 Å).

Improvements to the injection and radio-frequency systems are likely to result in further increase in stored beam current.

Cornell 12-GeV Synchrotron

One beam port is now in operation, providing 2.5 mrad of synchrotron radiation to a small number of local users during high-energy physics runs. The critical energy at 12 GeV is 38 keV (0.32 Å). Thus radiation up to 150 keV (0.08 Å) is available. This is the only U.S. synchrotron source that provides such high-energy radiation. Although additional research facilities could be provided, at present there is not a large demand, partly because of the limitations inherent in a synchrotron source (as opposed to a storage ring). However, since the Cornell program is relatively new (synchrotron radiation research started in 1974) the scientific demand for high-energy radiation may yet increase as initial research results are published.

Characteristics of Storage Rings Constructed for Synchrotron Radiation Use

Three examples of storage rings in the 0.6-4.5 GeV electron energy range that can be constructed as optimized sources

of synchrotron radiation are summarized in Table 5. The entries in the table are calculated from the physical and engineering relationships. Specific designs will modify the combinations of parameters but cannot change the orders of magnitude. Spectral characteristics of radiation from the bending magnets of these three rings are shown in Figure 12. Also, assumptions used in deriving the rough cost estimates given in Table 5 are described in the last part of this chapter.

At present, it appears that storage rings with energies higher than 4.5 GeV need not be considered as dedicated sources. The extremely short wavelengths (<0.1 Å) that they would produce do not now appear to have a large number of applications. Furthermore, these extremely short wavelengths could be utilized parasitically on high-energy physics machines. Such a program is already under way at the Cornell 12-GeV synchrotron. Provision is being made in the design of PEP, the LBL/SLAC 18-GeV colliding-beam storage ring, to facilitate the installation of synchrotron radiation beam lines. The proposal for an 8-GeV colliding-beam storage ring at Cornell includes several synchrotron radiation beam lines.

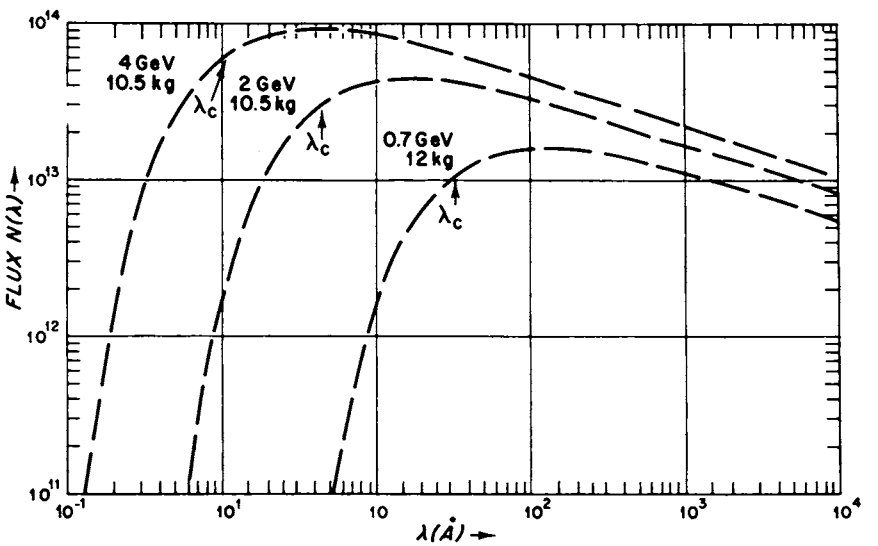


FIGURE 12 Synchrotron radiation spectra for the three rings summarized in Table 5.

Injectors to Storage Rings

Microtrons, linacs, and synchrotrons have been successfully used as storage-ring injectors. The energy, intensity, and emittance are the important injector performance parameters. An injection energy much lower than the storage-ring operating energy is economical and is feasible since the energy can be slowly ramped up in the storage ring after loading. The highest injection energy is most likely to enable the accumulation of large stored current since thresholds for instabilities, which limit stored current, vary inversely as the injection energy or a power of the injection energy. Injection at the operating energy means that the storage-ring configuration need not change between injection and storage. Consequently, the stored beam can be "topped up" in a very short time, providing almost continuous stored beam operation with an average current of 80 to 90 percent of maximum current.

A high-intensity, low-emittance injector will provide rapid fill rates--even if the aperture of the storage ring is limited by the presence of small-bore helical wigglers.

UTILIZING THE RADIATION--BEAM LINES AND MONOCHROMATORS

The basic requirement in utilizing synchrotron radiation is to maximize the radiant power falling onto a sample within the required spectral bandwidth. In practice, the broad-banded radiation is rendered monochromatic by a grating or crystal so that the bandwidth ($\Delta E/E$ or $\Delta\lambda/\lambda$) is 10^{-3} or smaller. Sample area varies widely depending on the experiment; however, optical focusing is often desirable or required. As an example, in an angle-resolved photoemission apparatus one would like to focus a monochromatic beam ($\Delta h\nu \sim 0.2$ eV or less) to a 1 mm^2 area on the sample with a convergence of 3° or less. Focal spots as small as 100 μm square are sought in x-ray biological diffraction work. X-ray microscopy can make good use of still smaller focused beams. Other synchrotron applications benefit from a more parallel beam to achieve high-energy resolution with a large-area detector and crystal monochromator.

There are many ways to collect and make monochromatic the flux from a storage ring. Gratings are used throughout the XUV range, and diffraction crystals are used in

the x-ray region. A block diagram is shown in Figure 13, which summarizes the main factors involved in matching various types of monochromators and optical configurations to synchrotron radiation sources. The monochromatized flux $N_{out}(h\nu)$ is related to the synchrotron source flux $N(h\nu)$ at fixed E, I, and H according to the relation

$$N_{out}(h\nu) = N(h\nu)\ \theta_x \cdot T_B \cdot T_M \cdot \left[(\alpha_x/\varepsilon'_x)\ (\alpha_y/\varepsilon'_y)\right],$$

where

T_B = transmission coefficient of beam-line optics,
T_M = transmission coefficient of monochromator optics,
α_x, α_y = horizontal and vertical acceptances (size times angle) of monochromator
θ_x = horizontal beam-line acceptance angle,
$\varepsilon'_x, \varepsilon'_y$ = horizontal and vertical emittances of beam-line optics at the monochromator,
ε_x^{eff} = effective horizontal source emittance including beam-line acceptance angle,
ε_y = vertical source emittance
$N(h\nu)$ = total source flux per horizontal angle per $h\nu$.

In the above relation, the ratios (α/ε') should be replaced by unity whenever α/ε' exceeds unity; i.e., all the available incident radiation is utilized when the

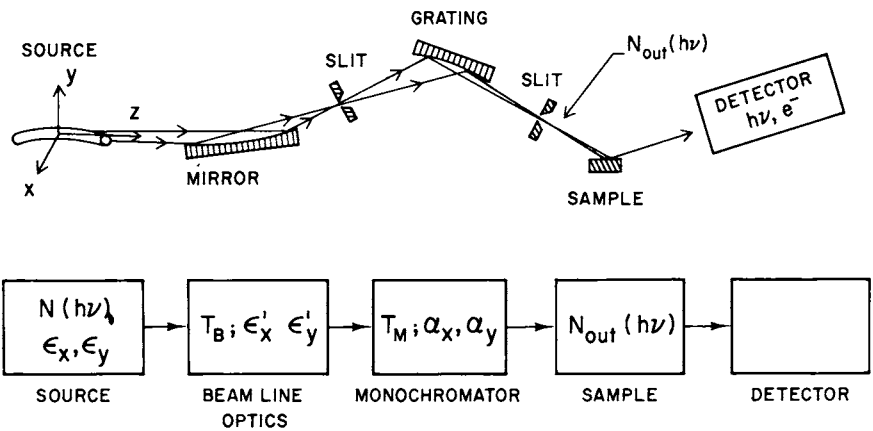


FIGURE 13 Schematic of source, beam-line optics, monochromator, sample, and detector configuration.

spatial and angular acceptance exceeds the emittance. The source emittances ε_x^{eff}, ε_y, are transformed into the ε'_x, ε'_y by the beam-line optics. For example, a focusing mirror can be used to modify the vertical emittance by demagnifying the vertical spot size while magnifying the vertical beam divergence σ_y. Postmonochromator optics (not considered in Figure 13) is sometimes required if a small spot size or well-collimated beam at the sample detector is required.

For each type of monochromator, the most efficient arrangement is obtained by matching the monochromator acceptance to the source emittance and by maximizing the transmission coefficients of the monochromator and beam-line optics. Presently, many different types of monochromator are being used with synchrotron radiation sources. Monochromators with diffraction gratings are generally used below ~500-1000 eV and include the Seya-Namioka type, normal-incidence type, Wadsworth type, and

TABLE 6 Monochromatized Flux (photons/sec) for a 1-m Seya Monochromator and Grasshopper Grazing-Incidence Monochromator Matched to a Typical 0.7-GeV, 0.1-A Storage Ring and for a Channel-Cut Crystal Monochromator Matched to a 4.0-GeV, 0.1-A Ring

λ (Å)	Source Flux	Flux at Sample
A. Seya (30-mrad Horizontal)		
304 (40.8 eV)	4.38×10^{13}	1.6×10^{11}
584 (21.2 eV)	3.77×10^{13}	1.2×10^{12}
1048 (11.8 eV)	3.30×10^{13}	3.1×10^{12}
B. Grasshopper (25-mrad Horizontal)		
44 (282 eV)	3.74×10^{13}	3.6×10^{10}
100 (124 eV)	4.18×10^{13}	2.3×10^{11}
200 (62 eV)	4×10^{13}	2.2×10^{11}
400 (31 eV)	3.5×10^{13}	1.9×10^{11}
C. Channel-Cut Crystal (2 mrad)		
0.4 (31 keV)	7.5×10^{11}	2×10^{11}
1 (12.4 keV)	3×10^{12}	7.5×10^{11}
4 (3.1 keV)	7.4×10^{12}	1.1×10^{12}

Rowland-circle grazing-incidence type. For photon energies below ~50 eV, the acceptances of such grating monochromators can usually be matched to the source emittances. However, for experiments at energies in the 50-1000 eV range, matching such monochromators to current sources becomes much more difficult--in fact, usually impossible--because of the small acceptances imposed by energy resolution requirements. Thus small emittance is very important for this energy range.

Illustrative examples of current design capability in the XUV range are summarized in Table 6. Monochromatized fluxes are given for several energies for a 1-m Seya-Namioka monochromator and a 2-m grasshopper grazing-incidence monochromator matched to a typical 0.7-GeV, 0.1-A storage ring (see Table 5). Comparable fluxes would be obtained by optimally matching these monochromators to the larger storage rings described in Table 4. New geometries and dispersion elements will certainly be employed in the future. For example, new transmission gratings look especially promising.[6]

In the x-ray range, crystal monochromators such as the channel-cut crystal are widely used. Acceptances can be well matched to source emittances with total flux generally increasing with horizontal angular acceptance.

Thus source flux rather than source brightness (flux/emittance) is usually the most important factor in the x-ray region. There are notable exceptions, i.e., when small sample areas are involved. Monochromatized fluxes for a channel-cut crystal monochromator matched to a typical 4.0-GeV, 0.1-A storage ring are given in Table 6.

SPECIAL INSERTIONS

Storage rings can readily be designed with insertions or straight sections having a free space a few meters long. Magnetic lenses at the ends of the insertions match the particle orbits to the adjacent arcs of the ring. These lenses can then focus the electron beam to a very small cross section at the center of the insertion, often referred to as a "low-β point." Wiggler magnets placed in these insertions can supply desirable radiation spectral characteristics and intensities.

Transverse Wigglers

Equation (3) shows that λ_c is inversely proportional to the product of magnetic-field strength and the square of electron energy. Short segments of strong vertical magnetic field alternating in polarity at the low-β focus will provide short-wavelength radiation from a very small and short source. Superconducting wigglers can operate at field strengths of at least 40 kOe. The small three-dimensional source size and large flux will result in high brightness particularly desirable for crystallography and x-ray diffraction. This type of source can be particularly useful for short-exposure diffraction from biological molecules.

Helical Wigglers and the Free Electron Laser

The most recent wiggler development is the "helical wiggler" or "free electron laser" analyzed some years ago by Motz and others and recently tested on the beam from a 24-MeV electron linac.[7] Kincaid[8] has proposed its inclusion in a storage ring, and this possibility has been analyzed by Blewett and Chasman[9] with encouraging results.

In the helical wiggler, the electron beam passes down the axis of a transverse magnetic field whose direction rotates with distance along the beam. Such a field can be produced by a bifilar helical winding. The orbits are tight helices of diameters a small fraction of a millimeter and pitches the same as the pitch of the applied transverse field. A notable feature of this device is that radiation in the direction of relativistic electron motion is modified by interference effects. For a well-collimated electron beam, and not too high a magnetic field, the broad synchrotron spectrum from a long helix is reduced to a line spectrum. Moreover, the radiation from such a device is circularly polarized.

The central wavelength of the wiggler spectrum is given by

$$\lambda_1 = \frac{\lambda_0}{2\gamma^2}(1 + K^2), \tag{8}$$

where λ_0 is the pitch or wavelength of the helical field, $\gamma = E/mc^2$, and $K = \lambda_0 eH/2\pi mc^2$. It turns out[10] that the emission on axis is optimized by adjusting the field so that K is approximately equal to unity, in which case the

central wavelength is simply the pitch divided by γ^2. As an example, consider a 2-cm pitch and 700-MeV electrons ($\gamma \sim 1400$). The peak wavelength would be at 100 Å.

The spectral purity of light from a helical wiggler will depend upon the homogeneity and emittance of the electron beam and the angular acceptance of the light collection optics. A lower limit to the linewidth is determined by the length of the helix; the fractional spectrum width is of the order of $1/N$, where N is the number of periods.

On axis, the flux per unit solid angle at peak wavelength λ_1 increases both as γ^2 and as N^2. This axial flux in a spectral band with $k = \Delta\lambda/\lambda$ is given by[10]

$$\frac{dN}{d\psi}(\lambda) = 2.28 \times 10^7 \, k\gamma^2 N^2 \text{ photons/(sec-mA-mrad}^2\text{)}, \quad (9)$$

where we have optimized by setting $K = 1$. A direct comparison can now be made with Eq. (5), which gives the flux from a bending magnet. Notice that the function F_4 in Eq. (5) is a maximum of 5×10^6 at the characteristic wavelength λ_c. Let us compare a bending magnet spectrum near λ_c with that of a helix at λ_1. Suppose that γ is the same in the two cases. The on-axis flux from a helix ($N = 100$) per unit solid angle per unit $\Delta\lambda/\lambda$ is about a factor of 10^4 greater than the on-axis flux from a bending magnet.

The width of the radiation cone from a helix when $K = 1$ is of the order of $1/\gamma$. When $K > 1$ the pattern opens up into a hollow cone. If, for example, $H = 5000$ kOe on the axis of a helix of 2-cm pitch and the electron energy is 2 GeV, K will be 0.9 and the radiation is contained in a cone with apex angle of about 0.5 mrad. The radiation pattern at the exit slit of a 5-m wiggler will be included in a circle of 2.5-mm radius, the outer radiation coming from electrons at the entrance end of the wiggler. The peak wavelength λ_1 at 2 GeV would be 11.8 Å. In a superconducting wiggler with a cold bore having an inside diameter of 1 cm, if fields as high as 5000 kOe are desired at the axis, Blewett and Chasman have shown that the pitch cannot be less than about 2 cm, and for practically achievable current densities may have to be 2.5 cm or greater.

The speculative possibility of using a helical wiggler to construct a free-electron laser deserves special mention. The same interference effects that reduce the

spontaneous emission spectrum from the normal broad synchrotron radiation spectrum to a line spectrum also provide the principle of operation of a new class of lasers based on the stimulated emission of radiation.[9]

It should be mentioned that while wigglers clearly have special attributes as radiation sources, their use may increase the complexity of storage-ring operation and could affect the other sources on the storage ring. For example, it appears that helical wigglers increase the vertical source size. Methods for avoiding this effect are under study. There are also practical problems, especially at injection, associated with the introduction of small apertures into the storage-ring configuration such as a 1-cm-diameter helical wiggler. Such apertures require more complex low-emittance electron injectors.

DISCUSSION OF OPTIONS AND COSTS

Comparison of Machine Options

As discussed above and in other parts of this report, there is a considerable diversity of research programs over a broad wavelength range that advantageously utilize synchrotron radiation sources. For convenience, we divide the wavelength region into two general spectral regions--the x-ray region above 1 keV and the XUV region below 1 keV--and discuss three general sizes of possible sources as summarized in Table 5.

X-Ray Sources

It seems generally agreed that a substantial capability with λ_c less than 2-3 Å (4-6 keV), as well as a capability with λ_c of about 1 Å (12 keV), is needed to support the x-ray research for which synchrotron radiation is suitable. A suitable radiation spectrum with λ_c less than about 2-3 Å can be obtained from the normal ring bending magnets with a ring energy in the range ~2.5-4.5 GeV.

A radiation spectrum with λ_c about 1 Å can be obtained from the normal ring bending magnets with an energy in the range 3.9-4.5 GeV. It can be obtained from high field wigglers in insertions in a ring with energy 1.9-2.5 GeV. The first example is large and requires large rf power. It would support many x-ray stations with λ_c of about 1 Å. The beam size would be fairly small and, in the horizontal direction, increased by momentum dispersion.

This type of ring would probably have two insertions for special sources in which very small beam sizes would be available.

The second example (energy 1.9-2.5 GeV) would have several insertions, four to eight, and would supply softer x rays from the normal bending magnets for many x-ray stations. High field wigglers in these insertions would produce x rays from very small sources with λ_c about 1 Å for up to 15-20 stations.

XUV Sources

There is no alternative to the use of high electron energy for x-ray production, and the x rays are accompanied by a continuous spectrum throughout the XUV region below about 1 keV. However, the lower energy photons can also be provided by a ring at 0.6-0.9 GeV. First, optical elements can be smaller and placed near the source point to capture large angles of radiation and need not be specially designed to absorb the higher energy x rays. More complete polarization is obtained by working near λ_c. The ring is smaller and requires less rf power and less shielding than larger rings.

This example would be a ring with at least 20 stations for XUV from small sources in the normal bending magnets. Two or four insertions would provide bright sources from specialized wigglers with λ_c as small as about 6 Å.

Further Considerations

It is often desirable, and sometimes necessary, to adjust the operating parameters of the storage ring to fit the needs of an experiment or of a class of experiments. An attempt to satisfy the needs of x-ray users and of XUV users on a single ring may encounter operating conflicts that result in much less than optimal operating efficiency. As an example, for a large storage ring, it may be necessary to schedule a block of time for very high current (~1 A) at low energy (~2 GeV) for maximum uv flux and other blocks of time for lower currents (~0.1 A) at high energy (~4 GeV) for hard-x-ray users. A large class of users may receive close to optimal flux with intermediate conditions (e.g., 3 GeV at 250 mA).

The operating conflict will be particularly serious if the helical wiggler proves to be an important radiation