

Atomic physics with synchrotron radiation

A powerful probe of the structure and dynamics of atoms promises not only to help solve problems in applied physics, but to test our understanding of quantum electrodynamics, relativity and many-body phenomena.

Bernd Crasemann and François Wuilleumier

Atomic and molecular science has grown dramatically in the last two decades. Today, the combined membership of the APS Division of Electron and Atomic Physics and Division of Chemical Physics rivals that of the society's largest Division, Condensed Matter.

The renaissance of atomic physics can be traced to two developments: the advent of new research tools and the increased relevance of atomic physics to important interdisciplinary problems. The primary new research tool for atomic theorists is clearly the large computer. It would have been impossible to reach the present state of the science without elaborate numerical techniques and the means to carry them out. On the experimental front, lasers have opened possibilities for extraordinarily precise spectroscopy. Yet lasers operate at relatively long wavelengths and can be tuned only over quite narrow intervals, limiting their use to investigations of outer electrons with low binding energies. Lasers are now complemented by

synchrotron radiation, which extends from laser wavelengths down to those of x rays or even gamma rays. As we will see in this article, synchrotron radiation is bringing about a revolution in the study of inner-shell processes.

Significant scientific problems whose solutions are beginning to come within reach motivate the current vigorous pursuit of atomic physics. On the applied side, these problems range from plasma diagnostics and atmospheric reactions to energy conversion and laser and accelerator technology. With a view toward such other fields as surface physics, condensed-matter physics and materials science, we should note that a thorough understanding of the properties of an atom in the free state is required for interpreting the complex structures observed in the solid state.² In the most fundamental research, atoms are the ideal proving ground for our understanding of quantum electrodynamics, relativity and many-body phenomena. Such effects as the screening of the self energy, the influence of the Breit interaction on multiplet splittings, and aspects of the electron-electron Coulomb correlation are subjects of intensive current investigations that are likely to yield results of broad generality.

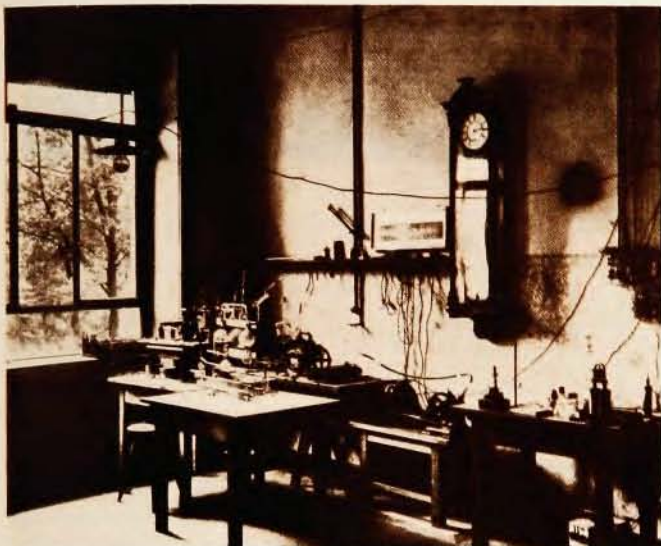
History. The impact of x rays upon science, medicine and technology has been profound, starting almost immediately upon their discovery by Wilhelm Conrad Röntgen during the Christmas holidays of 1895. Applications to military medicine were among the first. For a British expedition in Africa in

1896, the medical department of the War Office ordered two sets of Röntgen-ray apparatus to be sent up the Nile, to help army surgeons locate bullets in soldiers. In the battle of Tsushima during the Russo-Japanese war, the renowned cruiser *Aurora* carried a laboratory in which the naval doctor V. S. Kravchenko is said to have used x rays for diagnoses on 40 wounded sailors.

The key role of x rays in the development of modern physics is well known. Highlights include Max von Laue's 1912 discovery of x-ray diffraction; William Lawrence Bragg's application of x rays to the analysis of crystal structure, also in 1912; Henry G. J. Moseley's unequivocal determination of atomic numbers of the elements through his 1913 law relating those numbers to x-ray spectra; Pierre Auger's discovery of radiationless transitions and Arthur H. Compton's conclusive demonstration of the particle nature of photons, both in 1923.

The primary limitation on the use of Röntgen-tube x rays as a tool for research in atomic physics is that their intensity is appreciable only at a few fixed frequencies characteristic of the anode material. The advent of synchrotron radiation facilities (figure 1) in the last few years has vastly enhanced the potential for the use of photons, from the vacuum ultraviolet to hard x rays, for investigations of atomic structure and transitions. In the first article in this series on synchrotron radiation (June 1983, page 48), Arthur Bienenstock and Herman Winick detail the

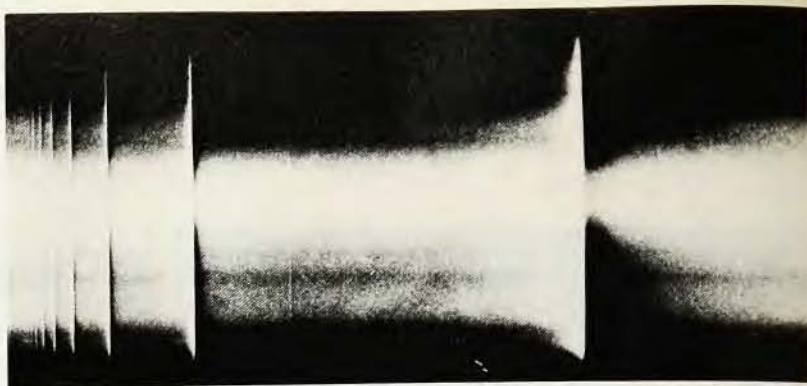
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Evolution in the generation of x rays. Inset is a photograph of Wilhelm C. Röntgen's chambers in the Physical Institute of the University of Würzburg, where he discovered x rays 89 years ago. Below is the storage ring ACO at LURE in Orsay, France. Between bending magnets 2 and 3 in the foreground, an undulator is operated on occasion. (Photograph of Röntgen's laboratory from Otto Glasser, *Wilhelm Conrad Röntgen und die Geschichte der Röntgenstrahlen*, Springer-Verlag, Berlin, 1931.) Figure 1



Atomic absorption spectrum of helium near 60 eV, and corresponding densitometer trace. Observation of these resonances in the helium absorption continuum led to the discovery of autoionizing two-electron excitations. Five are labeled. (Figure courtesy of Keith Codling, University of Reading.) Figure 2



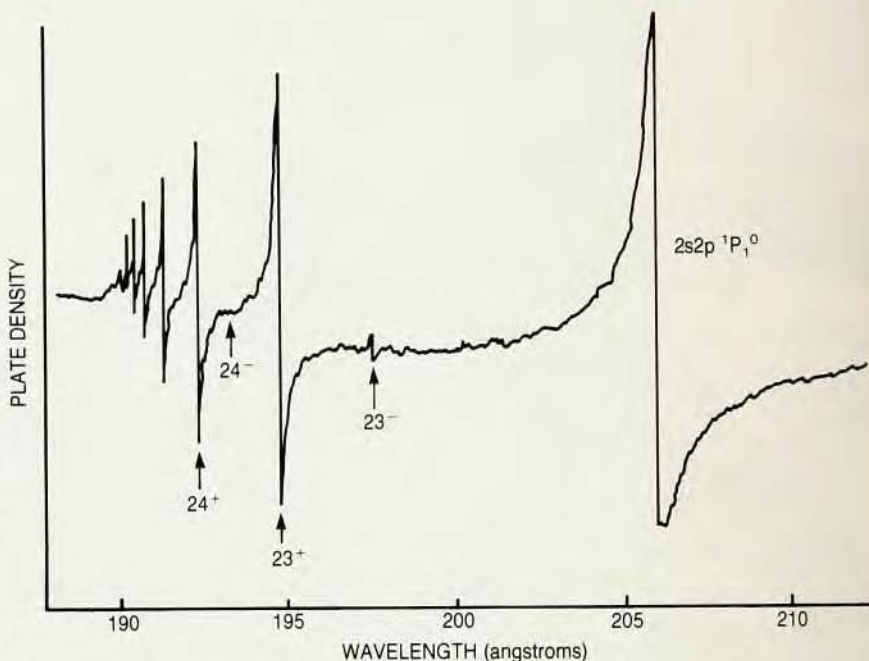
characteristics of these powerful new sources and describe their wide range of application.¹ In the present article, we look at the use of synchrotron radiation in a specific field—the study of free atoms.

After a brief discussion of synchrotron radiation itself, we examine some of the spectroscopic techniques that use it to study the electronic structure and dynamics of atoms. Then we look at some of the fundamental physics problems that are yielding to these techniques. Among the problems we discuss are many-body effects in atomic structure, the dynamics of atomic excitation and deexcitation, resonant Raman scattering, the effect of Auger emission on a slowly receding photoelectron, and photoionization of excited atoms.

Synchrotron radiation

The electromagnetic radiation emitted by relativistic electrons under transverse acceleration in storage rings has unique properties that make it an ideal probe of atomic structure. Intensity and tunability are the most important of these properties.

Free atoms generally need to be studied in the gas phase, and the low density of the target must be offset by a high incident photon flux. Electrons traversing the bending magnets of storage rings radiate considerable power; it is proportional to the fourth power of the electron energy and inversely proportional to the radius of curvature of the orbit. Thus, 100 mA of 3-GeV electrons confined to a circular path of 10-m radius emit more than 70 kW of electromagnetic radiation. The electrons radiate in the forward direction in a tight cone of root-mean-square aperture mc^2/E , or 0.2 milliradians for electrons of energy $E = 3$ GeV. Thus, one can bring substantial flux to ex-



perimental apparatus well removed from the ring. A 5-milliradian segment of the orbit in the foregoing example emits more than 3×10^{13} photons/sec within a 2-eV bandwidth at 1.8 keV, sufficient radiation for a typical experiment. Formidable as this flux is, insertion devices can enhance it yet further by several orders of magnitude, as described below.

The spectrum of synchrotron radiation from the bending magnets of a storage ring is made up of many harmonics of the fundamental frequency of revolution of the electrons. The individual harmonic lines are smeared out into a continuum that extends from the visible through the vacuum-ultraviolet and soft x-rays into the hard x-ray regime. The spectrum is characterized by a "critical photon energy" ϵ_c . Half the radiated power is contained in photons of energy less than ϵ_c , and half above. The photon flux peaks at $0.4 \epsilon_c$ and falls off nearly exponentially above ϵ_c . The critical photon energy is proportional to the magnetic field and to the square of the energy of the stored

electrons.

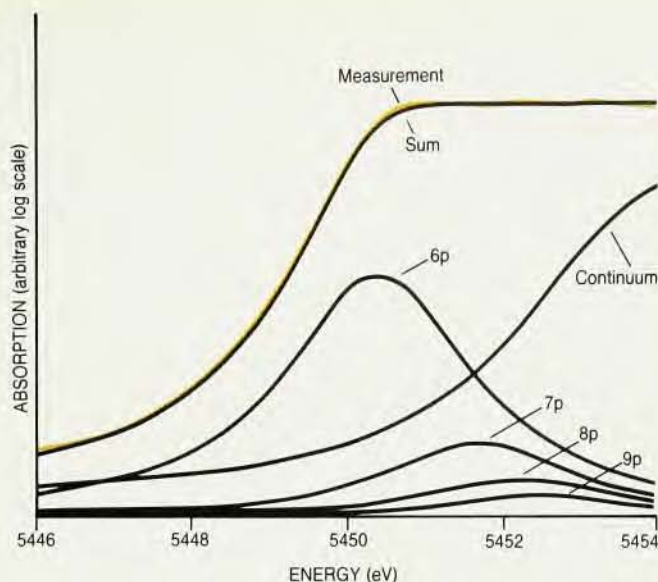
For virtually all applications, experimenters select a monochromatic slice of the synchrotron-radiation spectrum, using diffraction gratings at the longer wavelengths and Bragg diffraction for x-rays. These methods give resolution better than 1 eV at 5 keV and 0.1 eV at 100 eV, and they allow computer-controlled tuning of the photon energy over a wide range of the spectrum.

The intensity of synchrotron radiation can be greatly enhanced, and its spectral distribution modified, through the use of insertion devices—wigglers and undulators—that are now coming into widespread use.³ These devices are periodic arrays of dipole magnets or bifilar-wound solenoids inserted in a straight section of a storage ring. They can be made with conventional or superconducting electromagnets or power-saving and compact permanent magnets. The spatially alternating magnetic field causes the electrons to oscillate about their straight paths, and, therefore, to radiate.

Last year we saw the first report⁴ of

Absorption edge. Colored curve is the L_1 absorption edge of xenon, measured under high resolution with synchrotron radiation at the Stanford Synchrotron Radiation Laboratory. The analysis shows contributions from partial cross sections for the excitation of 2s electrons to np states; the limit of the Rydberg series leads to an accurate L_1 energy level. (From reference 6.)

Figure 3



the generation of stimulated radiation from the interaction of a circulating electron beam and the spatially oscillating magnetic field of an undulator. This successful operation of a storage-ring free-electron laser makes it reasonable to predict the realization, within the next few years, of a free-electron laser that is continuously tunable from about 1800 Å to the infrared.

While intensity and tunability are the most important properties of synchrotron radiation for the purposes of atomic investigations, there are other extremely useful features.³ These include a sharp time structure—typically, nanosecond pulses separated by as much as a microsecond. (Dennis Mills describes this in his article on time-resolution studies using synchrotron radiation, April, page 22.) This makes it possible to conduct time-of-flight electron and ion spectrometry. Synchrotron radiation has a very high degree of polarization, reaching 100% in the orbital plane at the critical energy. This property has become particularly useful for angle-resolved photoelectron spectrometry. It may be possible to superimpose radiation from two undulators to produce circularly polarized x rays, which would open intriguing additional possibilities for experimentation.

Spectrometric techniques

Absorption spectrometry is the simplest and oldest method for determining atomic energy levels. It was first used with ultraviolet synchrotron radiation in a pioneering program at the National Bureau of Standards in the early 1960s. This work led to the discovery of a wealth of discrete resonances in the photoionization continua of rare gases. The first atomic absorption spectrum obtained with synchrotron radiation, that of helium in the

200-Å region, revealed⁵ a series of autoionizing states, as figure 2 shows. This discovery is of crucial importance for the understanding of electron-correlation effects because it represents the simplest possible example of the highly correlated motion of two excited electrons.

More recently, x-ray absorption measurements have revealed the structure of inner-shell edges in rare gases, leading to precise determinations⁶ of hole-state energy levels. (See figure 3.) Total photoabsorption cross sections, such as the one in figure 4, reflect the deviation of the atomic potential from the Coulomb potential, and they demonstrate the inadequacy of independent-particle central-field models by clearly showing the influence of correlations and extra-atomic effects. The scope of absorption spectrometry is limited, however, in that the spectra normally reflect the undifferentiated response of the atomic electrons as a whole. Clearly, if we are to understand the complex gross features,⁷ we must make measurements that enable us to distinguish between the various channels through which the excited atom can decay.

Photoelectron spectrometry provides the necessary discrimination between final states. Using tunable incident light, this technique has proven an extremely valuable tool for the study of electronic structure and dynamics.⁸ To exploit fully the unique properties of synchrotron radiation, it is important to measure the photoelectron intensity as a function of three independent variables: the wavelength of the incident radiation; the photoelectron kinetic energy, which is characteristic of the atomic level structure; and the angle of emission. In addition, one must monitor the polarization of the incident radiation if one wants to determine the

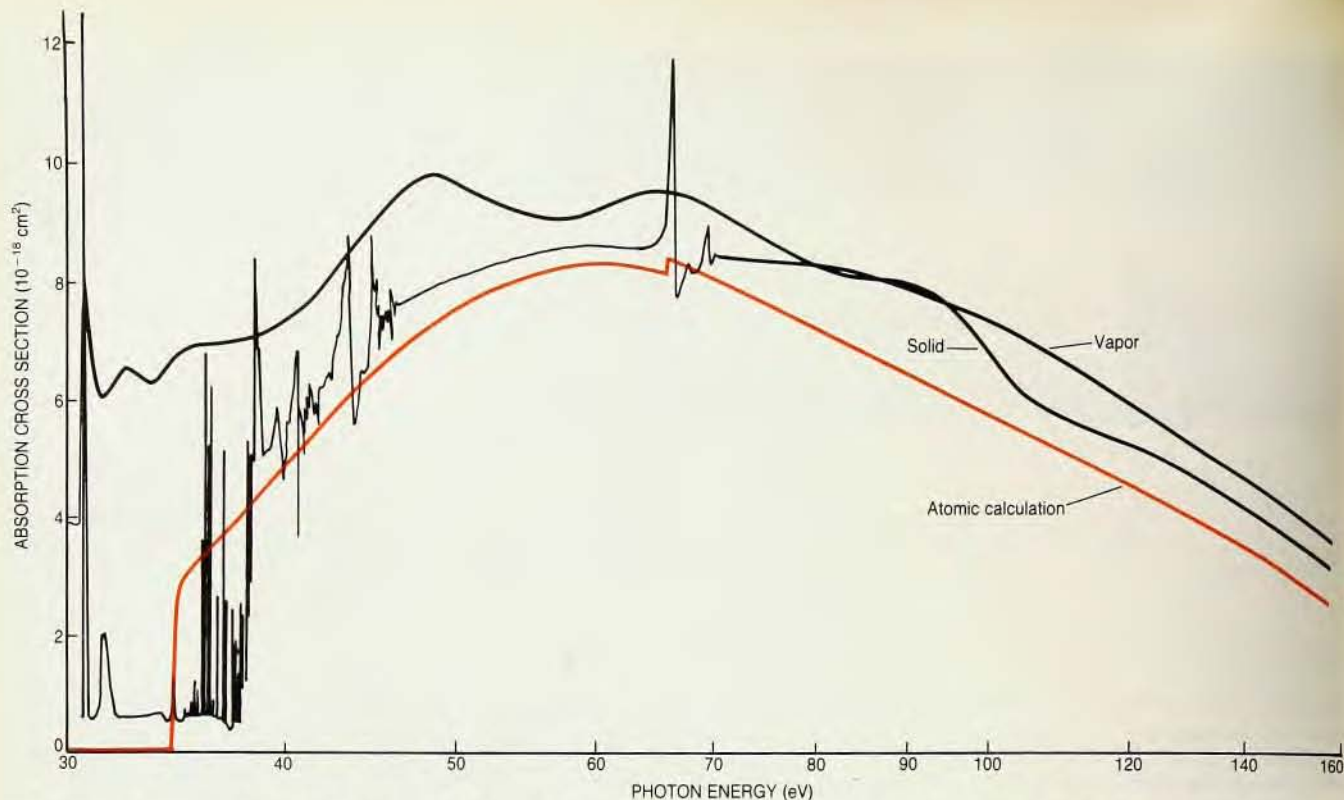
energy-dependent asymmetry parameter from the angular distribution. This parameter is important to the complete description of the photoionization process because it is very sensitive to many-body effects such as coupling between channels of excitation. A number of synchrotron radiation centers in the United States, France, Germany (see figure 5), England and Japan can perform photoionization experiments in which all three variables are measured.

A last step toward the complete characterization of an atom's final state is to determine the spin polarization of the photoelectron. Thus, experiments at the Bonn 2.5-GeV synchrotron have used circularly polarized ultraviolet light to study the variation of the spin polarization with the photoionization cross section; this variation is strong as the energy of the incident photon sweeps through autoionization resonances.

Other techniques that can take advantage of synchrotron radiation include Auger spectrometry and fluorescence spectrometry, both of which permit study of the details of atomic and molecular deexcitation following photoexcitation. Finally, one can use x-ray scattering to probe the distribution of charge in atoms. Using synchrotron radiation, x-ray scattering, in its inelastic part, becomes a sensitive indicator of Coulomb correlation in few-electron systems.⁹

Many-electron effects

We now turn to some interesting problems that physicists are studying with synchrotron radiation and the spectroscopies mentioned above. One goal of much experimental work is to understand the role of many-body effects in atomic structure and atomic transitions, to guide theoretical ap-



Sodium's absorption spectrum. These data on solid and gaseous sodium (black curves) were taken at DESY. They illustrate the inadequacy of independent-particle central-field models, which give the curve shown in color. (From Bernd Sonntag, Universität Hamburg.)

Figure 4

proaches to describe these effects. The many-body problem pervades all of physics, to be sure, but the atom is an ideal system in which to investigate it because here the potential is known, and the dominant charge of the massive nucleus makes the central-field model quite a good first approximation. Thus, one can study and treat many-electron effects selectively—often by perturbative techniques—and gain insights that carry over into the much more difficult situations encountered in nuclear matter and elsewhere.

Many-body effects often manifest themselves very distinctly, not only in outer-shell photoionization cross sections, but also in the photoelectron spectra of shallow core levels. For example, the photoionization cross sections of transition-metal 3d levels go through an enormous excursion near the energy needed to excite an inner 3p electron to an empty orbital. Such a phenomenon is not at all predicted by independent-particle models of the Hartree-Fock type; it arises from interference with the 3p to 3d excitation, which is activated at this energy. The interference causes autoionization, thereby enhancing the photoionization cross section.¹⁰

A striking example of the power of tunable synchrotron radiation to elucidate many-body effects is found in recent measurements of the asymmetric angular distribution of xenon 5s photoelectrons, conducted at the DORIS storage ring in Hamburg and at the

Tantalus I ring in Wisconsin.¹¹ The scarce data that were previously available seemed to indicate that the relativistic random-phase approximation adequately predicts the angular distribution if the correlation between channels of excitation is taken into account. However, the new measurements show that more extensive many-body effects come into play and that one must take into account channels of excitation that involve two electrons, a process that goes beyond the capabilities of the relativistic random-phase approximation calculations in their present form.

"Correlation satellites" in photoelectron spectra arise when a single incident photon causes the promotion of two atomic electrons, at least one of them into the continuum or to an autoionizing state imbedded in the continuum. We have already mentioned (figure 2) the discovery⁵ of such autoionizing doubly-excited states in the absorption spectrum of helium. For atoms of higher atomic numbers, core-level photoelectron satellite spectra can yield unique information about the correlation of the atomic electrons in the ground state. Use of the "sudden" approximation has led to progress¹² in calculating some double-excitation electron peaks, although experiment is well ahead of theory.

Dynamics of excitation

The excitation of atoms and their deexcitation through emission of photons or Auger electrons is usually

considered a two-step process. The decay is separated from the excitation by the intervening relaxation of the surrounding electrons, and the photon and electron emissions are treated as independent processes by means of first-order time-dependent perturbation theory. This two-step model of atomic excitation and deexcitation has the advantages of simplicity and tractability, but it breaks down in particular near the thresholds of inner-shell excitation and when interference arises between competing channels of deexcitation. An important advance in the fundamental understanding of atomic transitions will be brought about by a description that unifies the formation and decay of autoionizing and inner-shell vacancy states.

Several investigators have already made substantial inroads into the difficult problem of creating a unified theory of atomic excitation and deexcitation. Teijo Åberg and his collaborators¹³ at Helsinki University of Technology have drawn upon the fact that any excitation process that gives rise to emission of particles with characteristic energies, including photons, is a resonance scattering process. Hence, Åberg's group has worked toward understanding the electron-emission decay of autoionizing states and the photon-emission decay of inner-shell vacancy states from a unified point of view, by using time-independent multi-channel scattering theory. Theorists have predicted effects due to interfer-



Synchrotron-radiation beam lines fanning out from the storage ring BESSY in West Berlin.

Figure 5

ence between radiative and radiationless decay modes of excited atomic states, because the decay channels are not in fact independent. Several attempts are under way to arrive at a theory that can encompass relaxation and interference effects in a consistent and systematic way.¹⁴ Most of these difficult theoretical efforts are still at a conceptual stage where quantitative predictions are not easily extracted, and where the guidance of experimental data is of the essence for further elaboration. Synchrotron radiation is providing the means for measuring the threshold-excitation and interference effects that signal the breakdown of the two-step model.

The resonant Raman effect represents the epitome of the failure of the two-step approximation: Here a single matrix element describes both excitation and deexcitation. Resonant Raman scattering can be thought of as the inelastic analog of resonance fluorescence; both the radiative (x-ray) and radiationless (Auger) versions of the process have now been seen¹⁵ with synchrotron light. In the resonant Raman effect, a photon of exactly the right energy (within the lifetime width) promotes an atomic electron to an excited state, which decays through an x-ray or Auger transition, all of this occurring in a single step. The emission line exhibits linear dispersion and sub-natural linewidth at resonance.

In the other extreme, a photon with large excess energy can remove an

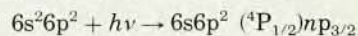
atomic electron rapidly and send it into the continuum. The atomic wavefunctions adjust themselves to the altered potential, and subsequent deexcitation occurs in a quite separate step.

The two opposite regimes, resonant Raman and wholly distinct excitation and deexcitation, are not separated by a drastic discontinuity. As one might expect, there is a phenomenon that links the two and leads gradually from one into the other.

Post-collision interaction is the mechanism that bridges excitation and deexcitation to a varying extent, feeding energy from one to the other and smoothly linking resonant Raman scattering with the opposite two-step extreme. Post-collision interaction arises near the threshold of photoionization, when the slowly receding photoelectron is still within, or near, the residual singly-charged ion as the decay occurs. If this decay consists of the emission of an Auger electron, the ion suddenly becomes doubly charged, and the receding photoelectron finds itself in a deeper potential well. Because the photoelectron's kinetic energy cannot increase instantaneously, the energy difference is transferred (through a virtual photon) to the Auger electron. The Auger electron thus gains energy, and the photoelectron, slow to start out with, may end up bound in a Rydberg orbit. By slowly tuning highly monochromatic x rays through an atomic inner-shell threshold and observing the energy of a suitable Auger peak, one can trace the

resonant Raman effect and its continuation in the form of post-collision interaction.¹⁶

Not only are atomic excitation and deexcitation tied together through such mechanisms as resonant Raman scattering and post-collision interaction, but, as we have indicated, the various channels are not truly independent and can interact and interfere with one another. Manfred O. Krause and his coworkers at the Wisconsin Synchrotron Radiation Center demonstrated¹⁷ this effect in the excitation of a Pb 6s electron to a Rydberg-like np state. The excited state undergoes radiationless decay into either of two single-electron channels, $[6p] \ ^2P_{1/2}$ or $[6p] \ ^2P_{3/2}$ (square brackets denote hole states). These same final states can, however, also be reached through direct photoionization of a 6p electron. Analysis of the electrons emitted in the decay that follows the photoexcitation of lead in the resonance series



shows surprisingly strong constructive interference of the autoionization resonance with the $[6p] \ ^2P_{3/2}$ direct ionization channel, and destructive interference with the $[6p] \ ^2P_{1/2}$ channel. This most striking demonstration of the interaction between channels is illustrated in figure 6 for $n = 7$.

Photoionization of excited atoms

Until now, the vast majority of photoionization measurements has been on

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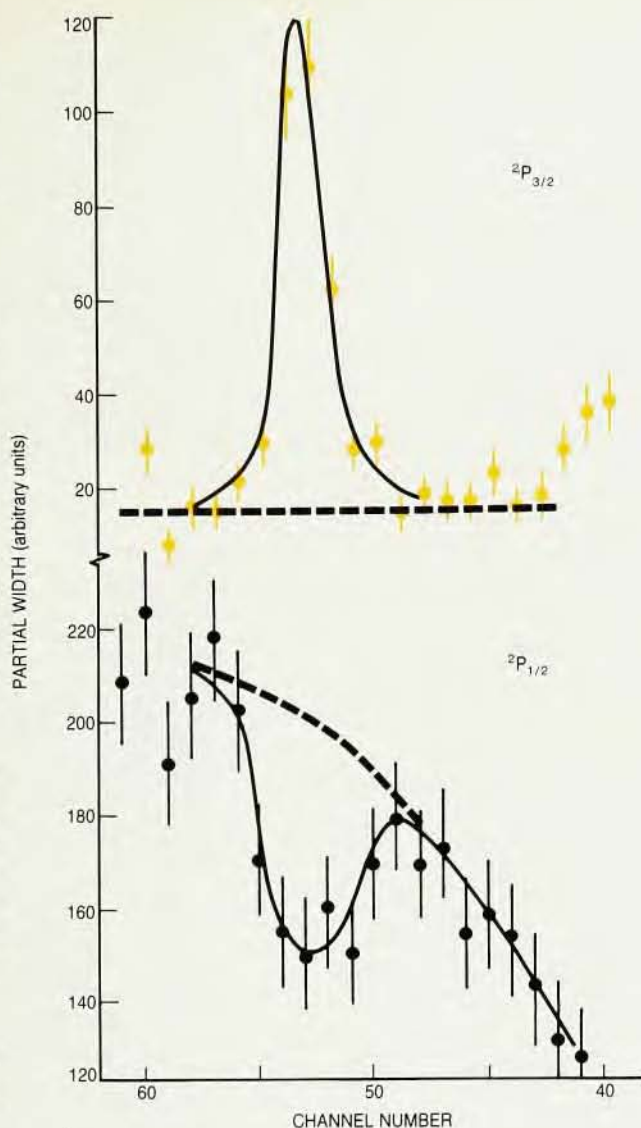
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Decay of lead [6s]7p states into [6p] $^2P_{1/2}$ and $^2P_{3/2}$ final states. Constructive interference between autoionization and direct photoemission channels enhances the $^2P_{3/2}$ final state (top), while the final state $^2P_{1/2}$ exhibits the effect of destructive interference (bottom). (From Manfred O. Krause, Oak Ridge National Laboratory.) Figure 6



atoms and molecules in their ground states. As noted above, these measurements have yielded very valuable information. Yet the field is limited: Dipole selection rules restrict the class of final states, and the initial states often are ensembles of near-degenerate levels (particularly in open-shell atoms), which complicates the interpretation. If one could prepare atoms in an excited initial state for photoionization, then one could circumvent these limitations. The knowledge to be gained on the behavior of excited atoms and molecules is of great potential importance in such fields as plasma physics, astrophysics and photochemistry.

In a few cases, experimenters have successfully prepared atoms in specific initial states through the use of high-power, tunable lasers, and they have photoionized these laser-excited atoms with synchrotron radiation. Available laser energies and ranges of tunability have restricted such experiments to outer electrons of alkali and alkaline-earth atoms.

Physicists at Orsay¹⁸ recently took advantage of the high intensity and broad spectrum of synchrotron radiation to extend these excited-state photoionization measurements. Light from the ACO storage ring, monochromatized by a toroidal grating, illuminated a weakly collimated metal-vapor beam in a volume monitored by an electron-energy analyzer. To the same volume, a tunable ring dye laser delivered up to one watt of 5000 Å to 6000 Å radiation. Figure 7 shows some typical results from this experiment. In the absence of laser radiation (top frame), the main photopeak arises from ionization of the $1s^2 2s^2 2p^6 3s$ 2S ground state of sodium. Synchrotron light transmitted in second order through the monochromator produces additional electron lines.

As the lower frame of figure 7 shows, two new features stand out when the laser is turned on. The first is a photoelectron peak that appears at a binding energy of 40 eV, corresponding to direct ionization of 2p electrons in laser-excited $2p^3 3p$ 2P sodium atoms. Because screening of the 2p electrons by the outer electron is reduced in this excited state, the 2p binding energy is greater than in ground-state atoms. The ratio of the amplitudes of the peaks at 40 and 38 eV binding energy indi-

cates the proportion of atoms in the excited state.

The second new feature apparent in the lower frame of figure 7 is the intense electron peak at 3.03 eV binding energy. This line arises from the decay of an even-parity autoionizing state excited by the stepwise absorption of two vacuum ultraviolet photons from a laser. The intensity of this autoionization peak is a very sensitive indicator of the density of excited states in the medium.

Outlook

In the two decades since Robert Madden and Keith Codling demonstrated the power of synchrotron radiation in atomic spectroscopy through their discovery of the autoionizing states of helium, use of the "light fantastic" from storage rings has expanded greatly. Yet, despite the progress that we have described in this article, synchrotron light is not being used nearly as widely in atomic physics

as in other areas such as surface science, solid-state physics and biology. The reason is primarily that gas-phase targets must be employed for the study of free atoms. The lack of density of these targets entails a need for very-high-flux photon beams. Only with the use of undulators and wigglers is the signal-to-noise ratio for many experiments using hard x rays becoming acceptable. It takes elaborate pumping arrangements to isolate gaseous targets from the ultrahigh vacuum of storage rings—unless the photon energy exceeds the 3.2 keV cutoff of beryllium windows. The size of the source and the horizontal angle of emission limit the degree to which one can monochromatize the radiation from bending magnets.

These difficulties, which make synchrotron light somewhat marginal for many research applications in atomic physics, will be resolved once more insertion devices are installed in existing machines, and once the next

Spectra of photoelectrons ejected from sodium atoms in the ground state (top, laser off) and in the excited $2p^63p\ ^2P_{3/2}$ state (bottom, laser on). Synchrotron radiation transmitted through the monochromator in first and second orders has energies 31.78 eV (right) and 63.56 eV (left). Figure 7

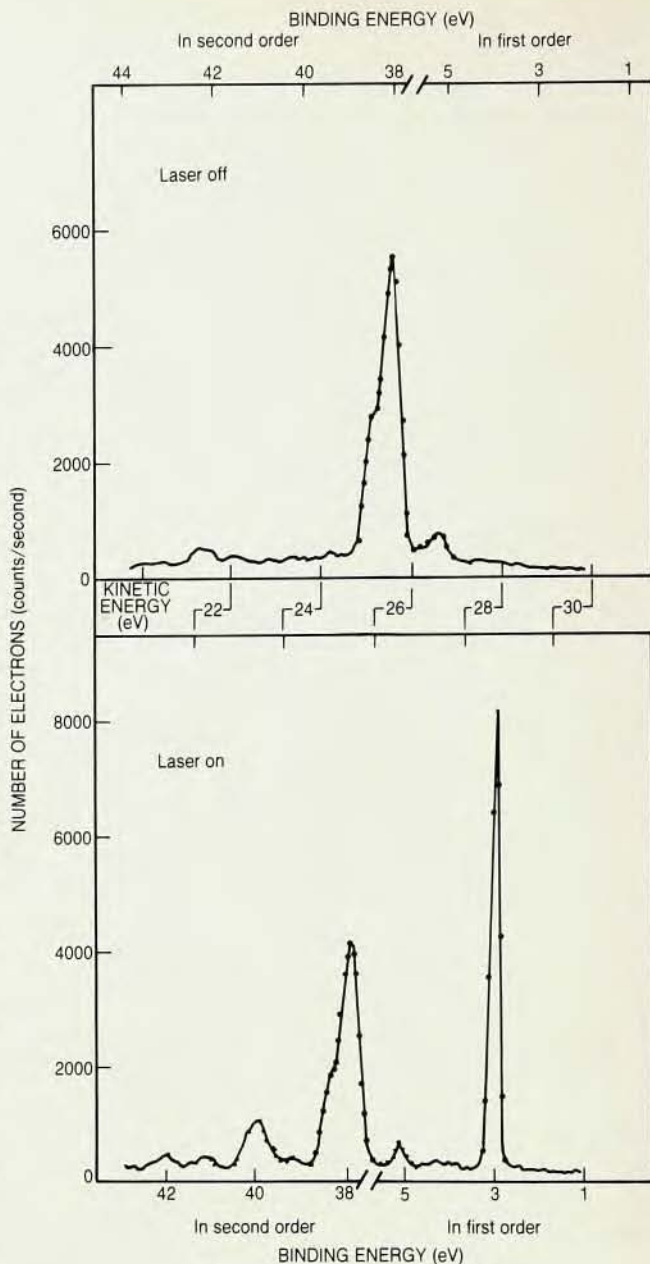
generation of synchrotron radiation facilities is born, starting with Super-ACO in Orsay in 1986, and, one hopes, followed soon by other proposed low-emittance, high-brilliance, all-insertion-device machines in the US and elsewhere.

* * *

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