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INTRODUCTION

A series of nine research briefing reports were prepared in 1984 under the supervision of the Committee on Science, Engineering, and Public Policy, a joint committee of the National Academies of Sciences and Engineering, and the Institute of Medicine. Prepared at the request of the Office of Science and Technology Policy and the National Science Foundation, the briefings were delivered in both written and oral form to these organizations between September 5 and October 1, 1984. The briefings were also presented to senior officials of other interested federal departments and agencies.

Selected Opportunities in Physics was one of these briefings. Among the topics of this briefing was physics at the laser-atomic frontier. To provide a fuller picture of the scientific opportunities and the role of this research on the national scene the Committee on Atomic and Molecular Science of the National Research Council organized a workshop which was held at the National Academy of Sciences on October 11, 1985. This report presents the papers delivered at the workshop.

Following is the introduction to *Selected Opportunities in Physics,* and the text of the briefing on physics at the laser-atomic frontier. (Reproduced from *Research Briefings, 1984,* National Academy Press, Washington, D.C.)

Physics has been rich in discoveries of fundamental laws of nature. These discoveries have influenced other fields of science from mathematics to biology and medicine and have spawned entire industries. Maxwell's electromagnetic theory, for example, forms the basis for understanding electromagnetic phenomena from waves to plasmas and underlies radio, television, radar, and our industrial power networks and modern communication systems. Quantum mechanics provides a framework for portraying physical reality and underlies all of the natural sciences. It has made possible the invention of the transistor, many of the amazing devices of modern solid state electronics, and the laser. Lasers, in turn, are having a major impact on many areas of science. They are also playing a rapidly expanding role in medicine and broad areas of technology, including communications, manufacturing, and national defense.

The discovery of x rays led to the largest single advance in the history of medical diagnostics. The recently created NMR body imaging technique may mark a comparable advance. From the study of electrical noise in the atmosphere, radioastronomy was born, which in turn provided a dazzling new portrait of the universe. Investigations of the properties of nuclei led to the creation of the nuclear power industry. Numerous other examples could be cited. The benefits to society from basic research in physics have been incalculable.

Recent fundamental advances in physics demonstrate that physics is still in a golden age and has never been more vigorous or more productively interactive with other fields. Progress in physics over the last decade has been remarkable. Puzzles that seemed to present insuperable challenges at the beginning of the 1970's have yielded to powerful and elegant theoretical and experimental techniques. The new insights and accomplishments have not only brought greater unity to the various branches of physics but have also strengthened the ties of

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physics to other areas of science and opened a vast array of new opportunities. Every part of physics has participated in the advance, as the forthcoming report of the Physics Survey Committee will make clear.

The present report focuses on six areas of special opportunity in physics. In order to find areas in which incremental funding may lead to major advances, a planning group (consisting of the Briefing Panel and others listed above) was convened. From twenty suggestions, the following six were chosen: (1) physics at the laser-atomic frontier, (2) relativistic plasma waves, (3) physical properties of deliberately-structured materials, (4) biomolecular dynamics and intercellular cooperativity, (5) cosmology, and (6) nuclear matter under extreme conditions. Large national facilities and well-established programs were explicitly excluded.

The six areas chosen promise to yield fundamental results of great interest. Many of the areas are likely to advance technology and to produce results with impact on the nation's industries; many can be expected to contribute to national security.

All six areas cut across lines of narrow specialization and link different fields. Progress in any one area can thus be expected to stimulate progress in other fields. Cosmology, for instance, connects physics to astronomy and astrophysics. Biological physics interacts strongly with chemistry, biochemistry, biology, medicine, and pharmacology. Deliberately-structured materials relate fundamental aspects of physics to many fields of technology.

1. PHYSICS AT THE LASER-ATOMIC FRONTIER

The laser continues to have a revolutionary impact in the field from which it emerged — atomic physics. The laser-atomic frontier comprises research opportunities generated by joining laser methods with new techniques in atomic and molecular physics.

Opportunities at the laser-atomic frontier include research with trapped particles; femtosecond (10^{-15} sec) spectroscopy and other new forms of spectroscopy; the study of previously inaccessible atomic, molecular, and ionic species; and the development of novel light sources. Advances in these areas will lead to new tests of basic theory and deepen our understanding of the structure of matter at the atomic and molecular level, including the transfer of energy and the nature of chemical reactions. In addition to high scientific interest, many of these topics are likely to yield technological advances. The opportunities selected include the following.

Trapped Particles

Using laser light, it is possible to cool to the millikelvin region ions confined in electromagnetic traps and also to cool and trap neutral atoms. In conjunction with recently developed ultrastable tunable lasers, these developments open the way to major advances in high-precision measurements: frequencies to parts in 10^{16}, masses to parts in 10^{11}, new tests of the isotropy of space, and studies of collective motion in plasmas and gases. Applications include advanced atomic clocks, optical frequency standards, metrology, and communications.

New Spectroscopies

Breakthroughs in the generation of femtosecond light pulses make it possible to take "snapshots" of atoms as they collide or molecules as they react, and to observe fast adsorption and relaxation on surfaces. Femtosecond spectroscopy has applications to fast electron circuitry and high-speed instrumentation. Another new type of

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spectroscopy is carried out with relativistic particle beams. Using the Doppler effect to shift highly stable laser radiation from the visible into the ultraviolet opens the way to new areas of high-resolution spectroscopy of atoms, ions, and molecules. Scientific objectives include studies of relativistic and quantum electrodynamic effects in highly charged ions, high resolution ultraviolet spectroscopy of atoms and molecules, and new types of photoejection measurements.

The Creation of New Species

Lasers can produce new species as well as species that have been inaccessible, such as multiply excited atoms, molecular ions, and clusters. Problems such as correlated electron motion, the evolution of matter from single atoms to the condensed state, and catalysis can be investigated. There are many applications to other sciences and to industrial problems. Studies of ionic species can lead to the design of more efficient combustion engines; catalysis is of enormous importance to chemical processing.

Matter in Intense Fields

Laser methods open the way to the study of matter under extraordinary conditions, such as in intense electric and magnetic fields and powerful radiation fields where nonlinear phenomena and multiphoton processes occur. Studies of multiphoton processes made possible by high-power lasers are expected to reveal new aspects of the interaction of photons with matter, and they may open the way to novel types of photochemical processing and isotope separation.

New Light Sources

The creation of the excimer laser and the generation of extreme ultraviolet light in supersonic atomic beams are two recent examples of light sources from research at the laser-atomic frontier. Other new sources can be expected, with wavelengths from the infrared to the soft x-ray region. Such sources would be valuable to wide areas of atomic physics, chemistry, condensed matter physics, and biology. Using coherent generation of pulsed laser light, for example, it may be possible to provide very short pulses in the soft x-ray region. This technique may provide a laboratory-based alternative to synchrotron radiation light sources for many applications.

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1. THE NEW SPECTROSCOPIES

Richard N. Zare
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INTRODUCTION

Most of what we know about the structure of matter comes from studies of the interaction of radiation and matter, that is, from spectroscopy in its most general sense. In the past decade spectroscopy has undergone a revolution. Spectroscopic techniques based on coherent tunable light sources have achieved a precision and sensitivity that is enormously greater than anything possible with conventional incoherent light sources. They have opened new areas in atomic and molecular physics and changed our ideas about the nature of light and optical processes. The techniques have made it possible to create new species such as Rydberg atoms and high vibrationally excited molecules, and species that are challenging our traditional ideas of structure and of ergodic and non-ergodic behavior. They make it possible to look at familiar species from unfamiliar perspectives. For example, our new ability to prepare and study molecules with a known amount of internal energy is having a dramatic impact on our understanding of chemical reactivity.

Beyond these scientific advances, a new optical technology has emerged. Methods based on non-linear interactions between coherent light and matter promise to provide frequency standards of unprecedented accuracy and a host of non-linear devices. Techniques are being developed to mix different frequencies of light as if they were radio waves, to correct the distortions due to propagation through the atmosphere, and possibly to overcome what was once regarded as the fundamental quantum limit of noise.

This collected enterprise, which marches under the banner of *laser spectroscopy,* is far broader than the conventional meaning of spectroscopy. We describe here a few highlights of the new advances.

ULTRA-PRECISE LASERS

A typical optical frequency, for instance the frequency of green light, is 5×10^{14} Hz. A conventional atomic lamp emits spectral lines with a spread in frequency $\Delta\nu$ that is due chiefly to the motion of the atoms. The resulting Doppler width is

$$\Delta\nu = \frac{v}{c}\nu \sim 5\times10^{9}\ \text{Hz}\,.$$

(v is the speed of the atom; c is the speed of light.)

A commercial dye laser bought off the shelf today can produce light with a spectral width of approximately 5×10^{5} Hz. Not only can the dye laser be tuned to any desired visible wavelength, its linewidth is smaller than that of an atomic lamp by a factor of ten thousand! The state-of-the-art in laboratory lasers is greater yet. Scientists such as John Hall at JILA and Richard Brewer at IBM-San Jose are already achieving stability of a few hundred hertz, representing an additional increase in spectral purity by a factor of one thousand, and another factor of one hundred is being talked about.

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We turn now to some of the scientific advances made possible by advances in lasers.

ULTRA-PRECISE LASER SPECTROSCOPY

The same Doppler effect that broadens the spectrum of light in an atomic lamp also broadens the absorption spectrum of a moving atom. Several techniques have been invented for effectively eliminating the first-order broadening due to the atom's motion. However, there is another effect, the second-order Doppler effect, which cannot be simply eliminated. It is relativistic in origin.

$$\Delta\nu = \frac{\nu}{2}\left(\frac{v}{c}\right)^2$$

and occurs essentially because a moving clock ticks more slowly than a clock at rest. Eliminating this effect is a challenge being faced today and the subject of several of the following talks. Here we describe how this problem is being solved for hydrogen.

The spectroscopy of hydrogen.

In the past, the most precise spectroscopy on hydrogen has involved the optical Balmer series ($n>2 \rightarrow n=2$). However, the 2s→1s transition is about to move into the spotlight. This $l=1 \rightarrow l=0$ transition is one-photon forbidden but it can occur by a two-photon process. Because the two-photon radiation rate is low the lifetime of the 2s state is long, and the intrinsic linewidth, according to the uncertainty principle, is small — only 1.3 Hz. (In comparison, for the Lyman-alpha transition, 2p→1s, the linewidth is 10^8 Hz.) By measuring the forbidden 1s→2s transition it is possible to determine the Lamb shift in the ground state, the Rydberg constant and the electron-proton mass ratio, all to unprecedented precision.

The first-order Doppler effect can be eliminated by a technique called *two-photon Doppler free spectroscopy*. The atoms absorb photons from a standing wave in the cell. Because the standing wave consists of two oppositely directed traveling waves, if the atom moves while it is absorbing radiation the frequency of one wave is increased while the other is decreased by $\nu_0(1 - v/c)$. As a result, the first-order Doppler effect cancels out for the two-photon transition. However, the second-order Doppler effect must also be eliminated.

A proposal by Theodor W. Hansch[1] is to employ an atomic hydrogen "fountain" (see Figure 1.1). Hydrogen atoms from a cold atomic source fly upward and then fall back under gravity. The atoms pass through a laser beam as they ascend, and again as they descend. The atoms absorb radiation near the top of the trajectory where they are almost at rest, nearly eliminating the second-order Doppler effect. The linewidth is calculated to be about 1 Hz: the resolution, $\nu/\Delta\nu$, is about 10^{15}! Such a prodigious resolution can be expected to reveal numerous effects that are generally overlooked. For instance, one will even be sensitive in the slight shift — 0.02 Hz — that comes about from the gravitational red shift within the apparatus.[1]

How does the potential precision for the 1s→2s transition compare with what we know today about the H-atom? Figure 1.2 summarizes the laser spectroscopy of hydrogen. We are talking about extending the resolution from 10^8 to 10^{15}. Historically, as we have increased the resolution of such experiments, we have always learned a great deal. Furthermore, the pursuit of this resolution provides a powerful driving force for the development of ever more precise lasers and can be expected to have a powerful impact on metrology, communications, and optical technology.

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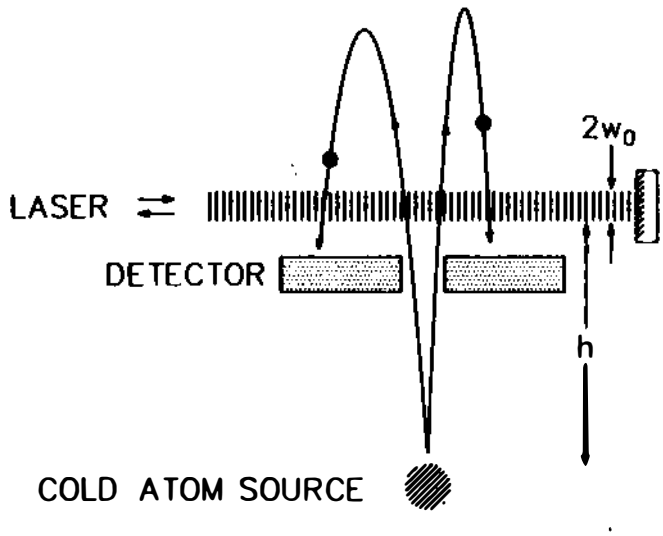

Figure 1.1. Scheme for two-photon optical Ramsey spectroscopy in an atomic "fountain." Taken from Ref. 1.

ULTRA-SENSITIVE LASER SPECTROSCOPY

Lasers make it possible to identify the particular exact quantum state of a molecule, to see how it vibrates, rotates, and translates, and to do this at extraordinarily low concentration levels. One method is *intracavity-laser absorption*. The idea is simple: the species being studied is placed inside the cavity of the laser. The laser behaves as an amplifier if it is run at marginal conditions. As a result, the atomic absorption process is enormously amplified compared to standard absorption. Another method is *laser fluorescence excitation*, in which the desired species fluoresces intensively under the laser light. Using *laser multiphoton ionization*, it is possible to ionize species with high selectivity, and to count individual ions. In *laser optoacoustic spectroscopy*, we can actually listen as molecules or atoms absorb light. The method traces back to Alexander Graham Bell[2] and his "spectrophone," basically a microphone in a cell. If a species in the cell is excited, collisions dump the energy into the medium, deflecting the diaphragm of the microphone and generating a signal. The sensitivity is extraordinary. Bell prophesied that this effect would probably be as important as spectrographs were to spectroscopy, but the technique was simply forgotten after his initial experiments. It is fitting that Kreuzer and Patel[3] at Bell Laboratory would resurrect Bell's technique with the laser. Now the method is being used inside flames and other places where one cannot easily place diagnostic probes.

Multiphoton Spectroscopy.

Single photon ionization is a popular technique for studying the electronic structure of atoms and molecules. For example, radiation at 58.4 nm from a helium resonance lamp is often used to photoionize materials by single photon absorption. The experiments required a windowless discharge and the cumbersome technique of vacuum ultraviolet. An alternative possibility is to photoionize by exciting virtual levels by simultaneously absorbing several photons.

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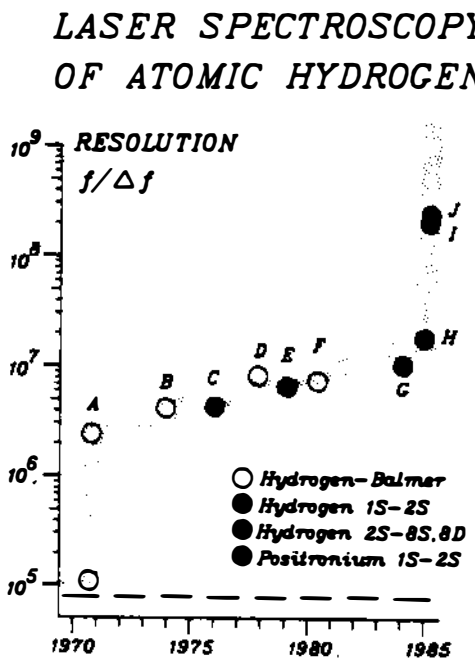

Figure 1.2. Progress in high-resolution spectroscopy of the H atom. The reference key is: A. T.W. Hansch, I.S. Shahin, A.L. Schawlow, Nature *235*, 63 (1972); B. T.W. Hansch, M.H. Nayfeh, S.A. Lee, S.M. Curry, and I.S. Shahin, Phys. Rev. Lett. *32*, 1336 (1974); C. T.W. Hansch, S.A. Lee, R. Wallenstein, and C. Wieman, Phys. Rev. Lett. *34*, 307 (1975). D. J.E.M. Goldsmith, E.W. Weber, and T.W. Hansch, Phys. Rev. Lett. *41*, 1525 (1978); E. C. Wieman and T.W. Hansch, Phys. Rev. *A22*, 192 (1980); F. S.R. Amin, C.D. Caldwell, and W. Lichten, Phys. Rev. Lett. *47*, 1234 (1981); G. S. Chu, A. Mills, and J.L. Hall, Phys. Rev. Lett. *52*, 1689 (1984); H. E.A. Hildum, U. Boesl, D.H. McIntyre, R.G. Beausoleil, and T.W. Hansch, Phys. Rev. Lett. *56*, 576 (1986); I. C.J. Foot, B. Bouillaud, R.G. Beausoleil, and T.W. Hansch, Phys. Rev. Lett. *54*, 1913 (1985); J. F. Biraben and L. Julien, Opt. Commun. *59*, 319 (1985).

With lasers it is now routine to induce non-linear multiphoton processes. It is possible to photoeject two electrons simultaneously, to excite inner atomic shells, and to observe what appears to be an anomalous enhanced absorption in high Z elements. The experimental results are posing provocative questions about collective motion of the electrons. As L. Armstrong's paper describes in Chapter 4, there is much to be done in this area. We describe the application of these ideas to a novel technique called resonance ionization spectroscopy.

Resonance Ionization Spectroscopy.

This process makes it possible to ionize selectively essentially every element of the periodic table. The method is illustrated in igure 1.3. The atom is multiphoton ionized, but by tuning the laser to an intermediate state the rate is vastly enhanced for the desired species. The method is so selective that it is possible to detect a single atom in a dense background. Such enormous sensitivity is essential to problems such as the detection of rare neutrino events and the search for superheavy elements, quark-bound atoms, etc. This technique makes it possible to analyze isotopes at new levels of sensitivity, making it possible to find out something about the primordial universe from looking at the isotopic composition of meteorites. A project is under way at Oak Ridge National Laboratory[4] to keep track of one thousand ^{81}Kr atoms in the presence of 10^7 ^{80}Kr or

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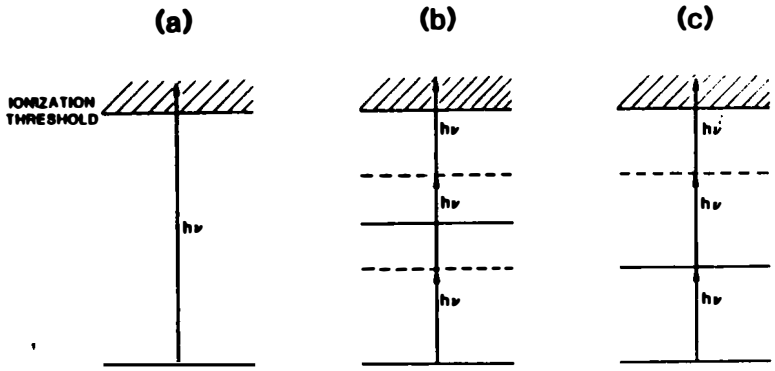

Figure 1.3. Schematic energy diagram for (a) one-photon photoionization, (b) nonresonant multiphoton ionization, and (c) resonant multiphoton ionization. The solid horizontal lines represent real levels, the dashed horizontal lines virtual levels. In the multiphoton ionization process the photon frequencies may be the same (one-color experiment) or different (multicolor experiment). Taken from R.N. Zare, *Science 226*, 298 (1984).

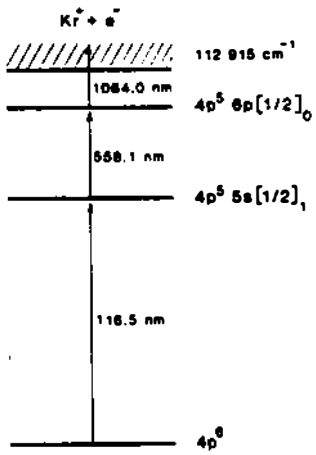

Figure 1.4. Three-photon resonance ionization scheme for krypton.

^{82}Kr atoms, plus a background of 10^{12} other types of atoms and molecules. As Figure 1.4 shows, the selective photoionization scheme requires laser light at 3 wavelengths: 116.5 nm, 558.1 nm, and 1064 nm. Intense radiation at 1064 nm is provided by a pulsed Nd:YAG laser; radiation at the other wavelengths is generated by a complex system of dye lasers, frequency doublers, mirrors and Raman shifters. The final result is a coaxial beam of the three colors; this goes to the apparatus shown in Figure 1.5. Initially the gas is frozen on a cold finger. A light pulse from separate laser desorbs the gas, and then the multiphoton system is fired. The krypton ions are

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gas is frozen on a cold finger. A light pulse from separate laser desorbs the gas, and then the multiphoton system is fired. The krypton ions are collected by a quadrupole mass-filter that sorts them according to mass; the ions are accelerated through 30 keV and driven into a Be-Cu target where they are buried. Each impact knocks out a pulse of electrons, so every ion gets counted. The Be-Cu target can be removed, the krypton baked out, and the whole process repeated, making recounts possible.

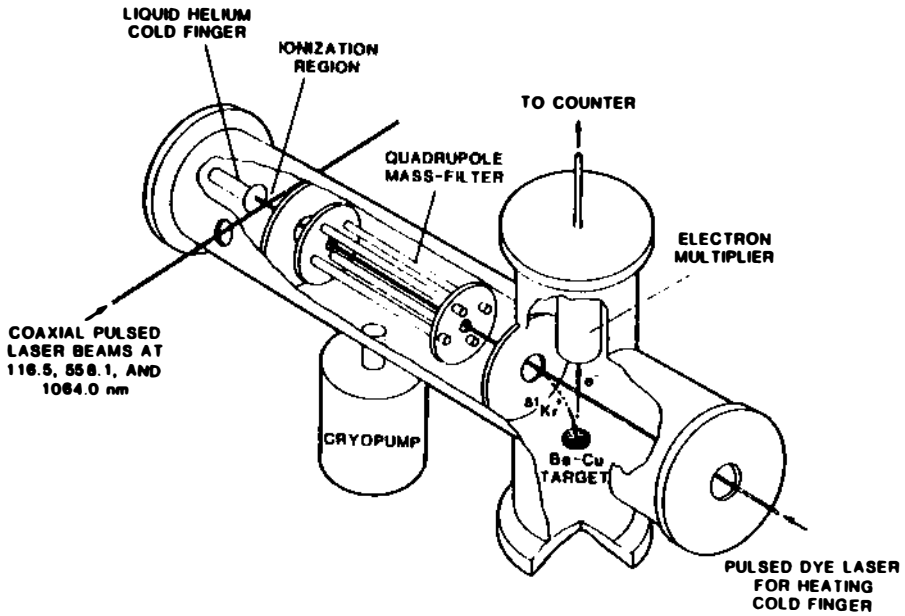

Figure 1.5. Schematic diagram for the sorting and counting of ^{81}Kr atoms using the three-photon resonant multiphoton ionization scheme: Kr $4p^6$ + $h\nu$ (116.5 nm) → $4p^5$ $5s[\frac{1}{2}]_1$ + $h\nu$ (558.1 nm) → $4p^5$ $6p[\frac{1}{2}]_0$ + $h\nu$ (1064.0 nm) → $Ke^+ + e^-$. This drawing is adapted from Chen *et al.* [4]

The particular interest in ^{81}Kr is due to a proposed solar neutrino experiment in which the common bromine isotope ^{81}Br is converted into ^{81}Kr by neutrino absorption. With a detector of about 400 m^3 filled with carbon-tetrabromide, one expects that ^{81}Kr will be produced at a rate of about 2 atoms/day. The plan is to accumulate the atoms for about a half year, and then cycle the system and implant the atoms in a silicon target which can be laser annealed to eject the ^{81}Kr for counting and recounting. It is believed that the number of atoms can be determined with a signal-to-noise ratio of about 100.

ULTRA FAST LASER SPECTROSCOPY

The fastest flash lamp operates in the nanosecond (10^{-9} s) range. For over a decade one has been able to work in the 10^{-12} s range with picosecond lasers, but today one can achieve pulses in the femtosecond (10^{-15} s) region. The record is slightly under 10 fs. In a pulse this short the light only has time for about three oscillations. Such short pulses allow one to "freeze" vibrations, and to witness all sorts of electronic phenomena in solids such as electron lattice relaxation and electron-hole recombination. Understanding these processes ultimately will determine how fast we can make solid state switches and logic circuits and whether it will be possible to make computer

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devices that will be limited only by the speed of light.

Spectroscopy of the Transition State.

A dream of scientists who try to understand chemical transformations from one state to another of matter is to make a "motion picture," a real time picture of what is happening.[5]

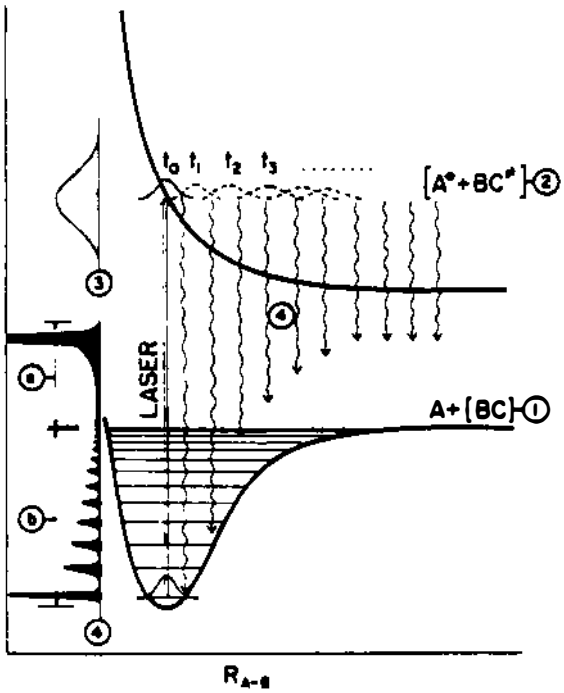

Figure 1.6. A photodissociation experiment. The laser transfers the ground-state wave function to the repulsive excited state where it evolves (dashed wave packets $t_1, t_2 \cdots$, etc.) into A* + BC†. Indicated in the figure by numbers are accessible experimental probes: (1) equilibrium geometry and spectroscopic constants of the final BC product, (2) internal-state, angular, and velocity distributions of the final products, (3) absorption (photodissociation) spectrum, (4) emission spectrum, (a) wing emission, (b) discrete emission. Taken from Ref. 7.

We have a simple intellectual construct for describing the reaction: the potential energy surface. According to the Born-Oppenheimer principle, we can separate the electronic motions from the nuclear motions and think about a representative particle moving across a potential energy surface which follows several final paths. The problem is that the switching of paths from reactants to products in a chemical reaction occurs too fast. However, by using femtosecond time resolution provided by inelastic light scattering, we can now explore exactly how the reaction evolves. The essential idea is to study only the second half of a collision, the outward part of a collision in which a species dissociates. The species absorbs light and then tears itself apart (dissociates):

$$ABC + h\nu \rightarrow [ABC] \rightarrow AB + C \ .$$

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We start by considering an initial ground state with a wave packet at time $t=0$. The absorption of light projects the system onto the repulsive part of a potential surface and starts to evolve in time and moves away, eventually becoming a free particle system. However, while this is happening there is a small probability that the system will re-radiate the energy that was absorbed in the form of light. This scattering process depends on the overlap of the state's wave packet and the ground state wave function. The time evolution during the transformation of the reactant to product leads to the development in time of differing Franck-Condon overlaps of the vibrational levels of the initial electronic surface (see Fig. 1.6).

The theory for this process was created by Eric J. Heller.[6] Without going into details, one can still get a feeling for what is involved. The signal is proportional to the following, as in Raman scattering:

$$\alpha_{if}(\omega) = \int_0^\infty \exp(it\,\Delta\omega) < \phi_f \mid \phi_i(t) > dt.$$

This is half the Fourier transform of the overlap between the nuclear wave functions of the final state and initial states. It depends on the frequency difference $\Delta\omega$ between the scattering light and the frequency needed to cause this process to occur. By tuning off-resonance, i.e., by changing $\Delta\omega$, one can sample the dynamics on a time scale that decreases as $\Delta\omega$ is increased.

An example of this process, from work by Imre, Kinsey, Sinha and Krenos,[7] is shown in Figure 1.7: the breaking of methyl iodide (CH_3I) into a methyl radical and an iodine atom. CH_3I is tetrahedral (a central carbon atom, 3 hydrogens out of the face and iodine).

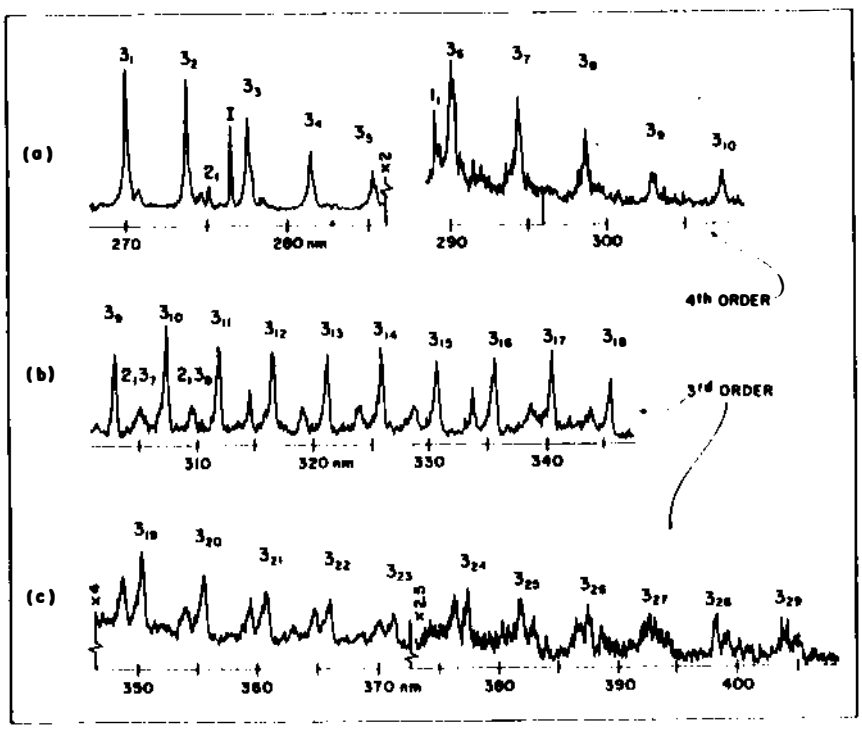

Figure 1.7. (a) CH_3I Raman spectrum obtained with excitation at 514 nm, (b) at 355 nm, (c) at 266 nm. Taken from Ref. 7.

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It dissociates into a planar methyl radical and an iodine atom. At large frequency offsets, i.e. when the CH_3 and I are close in time (Fig. 1.7a), the only frequencies observed correspond to the fundamental stretching vibrations of CH_3I. As the time increases (Fig. 1.7b) one can see a small amount of the C-I stretch vibration. For very small frequency offset, corresponding to very long times — one sees progressions in the stretching vibration and starts to see the "umbrella mode" of the free CH_3 radical (Fig. 1.7c). In short, we now have a means for studying how a reaction takes place on a time scale that corresponds to femtosecond or sub-femtosecond resolution.

SQUEEZED STATES OF LIGHT

As a last topic we turn from how light is increasing our knowlege of matter to how matter is changing our concept of light. As D.H. Auston describes in Chapter 5, it appears possible to create "squeezed states" of light whose photon statistics offer significant advantages in precise metrology, the detection of gravitational waves and in communications. This research area is still in its infancy and only a brief introduction will be presented here, based on the review by Walls[8] and an article by Levenson and Shelby.[9]

We describe a light wave as having an electric field given by

$$E = E_1 \cos \omega t + E_2 \sin \omega t,$$

where ω is the frequency of oscillation and the components E_1 and E_2 are called the quadrature amplitudes. Let us construct a phasor diagram (Fig. 1.8) in which the x and y axes are the components E_1 and E_2.

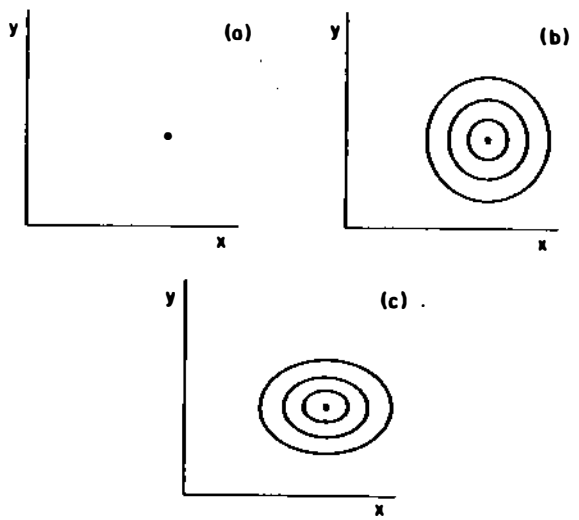

Figure 1.8. Phasor diagrams showing (a) description of a classical light wave, (b) description of a coherent state of light, and (c) description of a squeezed state of light.

A classical state of the light wave is represented by a single point in the phasor diagram since both quadrature amplitudes can be perfectly specified (Fig. 1.8a). In quantum mechanics the quadrature amplitudes are conjugate variables, conjugate in the same sense as the position x and the momentum p_x. Consequently, we must replace our single point in the phasor diagram by

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probability contours. A perfectly coherent state would be represented by circular contours centered at the average value of the field (Fig. 1.8b). Let us return to quantum mechanics; the uncertainty principle states the uncertainty in one conjugate variable times the uncertainty in the other must be greater than or equal to Planck's constant. It follows that one conjugate variable can be determined accurately only at the expense of large fluctuations in the other. A "squeezed state" of light is one having "elliptical" probability contours (Fig. 1.8c), with the minor axes smaller than the radii of the corresponding circular contours of the coherent state. Thus a squeezed state of light has one quadrature amplitude less uncertain than that of a coherent state and by using non-linear optics effects to determine that quadrature amplitude it is possible to make measurements more precise than the ordinary quantum limit.

Suppose we have a squeezed state of light. If we measure the power of the light, for example, with a calorimeter, we will not learn about its nature because energy flux is proportional to the sum of the squares of the two quadrature amplitudes. What we need to do instead is some type of beat experiment, homodyne or heterodyne detection, in which we can make light waves interfere with one another. Several laboratories have or appear to be on the verge of generating and detecting squeezed state of light and D.H. Auston will describe one such experiment in more detail. Already we can glimpse into the future that light-matter interactions will fundamentally alter our conception of both what is light and what is matter.

CONCLUSION

These examples are selected from a large body of new studies made possible by lasers and laser spectroscopy. Many other examples will be found in the following papers. We hope that they provide convincing evidence that the assertion that there has been a revolution in spectroscopy is in no way exaggerated.

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2. LIGHT UNDER CONTROL: A NEW GENERATION OF RADIATION SOURCES

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INTRODUCTION

Electromagnetic radiation from the far infrared through the x-ray regime is the primary tool for probing the structure of atoms and molecules, whether free or bound in solids and liquids. This range makes it possible to study spatial features on scales that extend down to less than the atomic spacing in solids and to explore energies from 100 keV to less than thermal energy at room temperature. The quality of the information depends critically on the quality of the available light sources. (We shall follow common usage by referring to radiation from the infrared to the x-ray regime simply as "light.") Prior to 1960 all light sources were essentially thermal, producing chaotic, incoherent radiation. During the past 25 years a new family of light sources has been created, sources that are controllable and coherent. Comparing the new and old light sources is roughly like comparing a modern radio transmission of Mozart's music around the world to Marconi's first dot-dash signal across the Atlantic using the electrical noise from gigantic sparks. This revolution in light sources is by no means complete and today we have new opportunities for improving the quality of radiation and expanding the spectral region of our sources.

The quality of a light source is determined by the degree of temporal control, which is closely related to the concept of spectral purity, and by the degree of spatial control, which is related to the source's directionality. Temporal control requires producing a well-defined frequency that maintains a fixed relative phase for extremely long times, and also being able to switch radiation *on* and *off* in short times. Spatial control involves maintaining a uniform phase and amplitude across the wavefront of a light beam. This determines how well a beam can be focused, propagated in waveguides, or used in applications such as holography.

The overall control of light is best described by the parameter of *brightness* or specific spectral intensity. Brightness is the amount of energy passing through a unit area in a unit time, unit solid angle and unit frequency interval. A source with only a modest power can be extremely bright if the radiation is carefully controlled: the one milliwatt helium neon laser used in a supermarket checkout counter is brighter than the light from a one megaton nuclear bomb! Within the narrow bandwidth and single direction in which it radiates, the helium neon laser provides more energy per second than a thermal source at 10^{12} K.

This paper is about controlled light sources, how much control now exists and what is being done to extend it.

NEW SOURCES OF VISIBLE RADIATION

Lasers are our best controlled sources of light, and their control is greatest in the visible region. The essential element of control is the cavity formed by the laser's end mirrors. Most of the radiation in a laser is reflected back and forth through the gain medium to be amplified again

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and again. (The output beam is created from a small fraction of the circulating power that is allowed to escape through one of the end mirrors.) The cavity provides a memory that establishes the temporal coherence: the diffractive spread of the feedback beam establishes the spatial coherence of the radiation. For visible radiation the relative phase can now be kept constant for as long as 10^{12} cycles, equivalent to sustaining a musical note in phase for a third of a century.

The output of visible and near infrared lasers is routinely put into a single diffraction mode whose beam divergence and cross-sectional area are limited only by the fundamental wave nature of light. Pulsed lasers can now operate in a single spectral mode with a bandwidth which is set only by the finite duration of the pulse and the uncertainty principle $\Delta\tau\Delta\omega>1$.

NON-LINEAR OPTICS

The coherence and high field strength of laser radiation can cause non-linearities in the response of matter to radiation to become important on a macroscopic level. For example, the small non-linearity in the induced polarization of a KDP (potassium dihydrogen phosphate) crystal can be used to convert as much as 80% of the 1.06 micron infrared radiation from a Nd:YAG laser into .53 micron visible radiation. The ability to generate efficiently harmonics and subharmonics of light greatly extends the range of laser sources and also gives new insight into the structure of matter.

Mode locking techniques use a non-linear medium in a laser cavity for which higher intensities have lower loss. The result is a compression of the radiation into a single high intensity, short duration pulse which oscillates back and forth in the laser cavity. The output of such a laser is a series of pulses with a duration below 10^{-14}sec, any one of which can be selected using an optical switch. As described in Chapter 5, D.H. Auston has driven a non-linear crystal with a short pulse of visible light to generate radiation in the 2 to 5 micron region. The infrared output is the ultimate in a short pulse — a single cycle of the electromagnetic field!

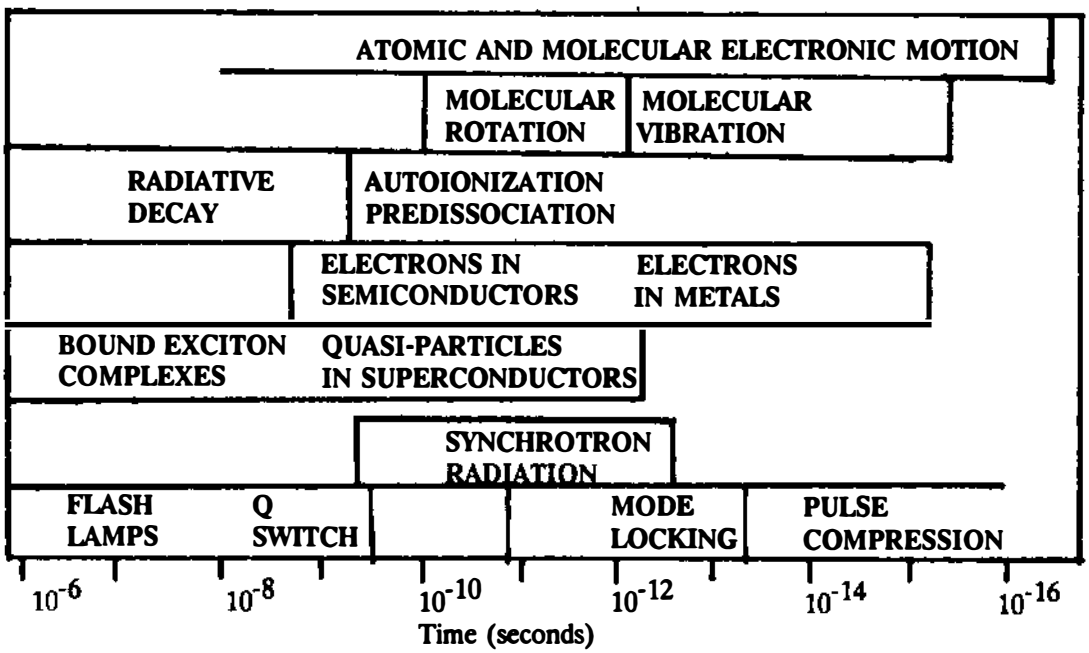

Figure 2.1 Time constants for some basic physical processes and new light sources.

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Figure 2.1 displays typical time constants for atomic and molecular processes. The minimum duration of the older light sources was longer than 10^{-8} seconds; the six order of magnitude decrease in pulse length has opened new areas of science. It is now possible to "freeze" the environment of atoms in solids and gases by exciting and probing the atoms in times shorter than the relaxation times of the environment. The short pulse sources have opened new areas of study in the dynamics of condensed systems, the reorientation of solvent molecules in liquids and the transfer of energy to electrons in various bands in semiconductors.

The ability to compress the modest energy of mode locked lasers into ultrashort pulses, combined with the ability to focus to diffraction limited spots, can be used to create fields of enormous intensity: the field strengths in the light beam can exceed the internal fields of atoms. These fields have led to startling experimental discoveries. For example, researchers have found evidence of efficient multielectron ionization of atoms corresponding to the simultaneous absorption of 99 photons by the atom, and also for efficient multiphoton absorption by electrons above the ionization limit. As L. Armstrong, Jr. describes in Chapter 4, these new experiments pose strong challenges to theorists, for they open a regime that could not be touched before the development of the current generation of light sources. The application of the short pulse source to the study of dynamics in condensed systems is just beginning. A limitation to this field is the lack of comparable sources in the ultraviolet and soft x-ray spectral regions.

Visible lasers today routinely operate in a single spatial mode and bandwidths of 10^6 Hz are common. Bandwidths below 10^2 Hz have been achieved. Focussed intensities of 10^{13} W cm^{-2} are common, and 10^{16} W cm^{-2} has been achieved as a routine value. These figures provide impressive evidence of the high control and flexibility of modern light sources in the optical regime.

Figure 2.2 shows the range of research that requires radiation in the vacuum ultraviolet (VUV; below 200 nm) or the extreme ultraviolet (XUV; below 100 nm). Lasers below 100 nm are not available, although several groups have recently demonstrated intense amplified spontaneous emission in that region. Lasers exist between 100 nm and 200 nm, but there are major difficulties associated with the lack of mirrors, windows and non-linear components that can stand the high intensities required. These problems have prevented the achievement of the high level of spatial and temporal control that is possible in the visible. Consequently, a variety of different approaches have been used to create sources below 200 nm.

XUV AND X-RAY SOURCES

Laser-Produced Plasmas

The simplest and most convenient laboratory source of XUV and soft x-ray radiation is a laser produced plasma. This source employs the light from a high energy, pulsed visible laser, focused to a spot with a diameter <100 microns, to create a hot ($kT \sim 500$ eV), dense plasma. The incident intensities are typically 10^{12}–10^{14}W cm^{-2}. When high-Z targets are used, intense continuum radiation is produced from 1 nm to ~100 nm (1.2 keV to 12 eV). Because the plasma is a thermal source the system is fundamentally simple in concept and execution. By using driving pulses from a good visible laser, much of the control inherent in the laser driver can be transferred to the short wavelength radiation generated by the plasma. Unlike electrically driven sparks, which are highly irreproducible in both size and duration, laser plasmas can be small (10 microns) and short in duration (<100 psec), with a pulse-to-pulse reproducibility that is as good as the driving laser. The output radiation is broad band and incoherent, but the near point source nature of the plasma results in a very high brightness output with a peak flux exceeding that of any other source of radiation between 1 nm and 100 nm. Such a source has the added advantage of being compact and relatively cheap.

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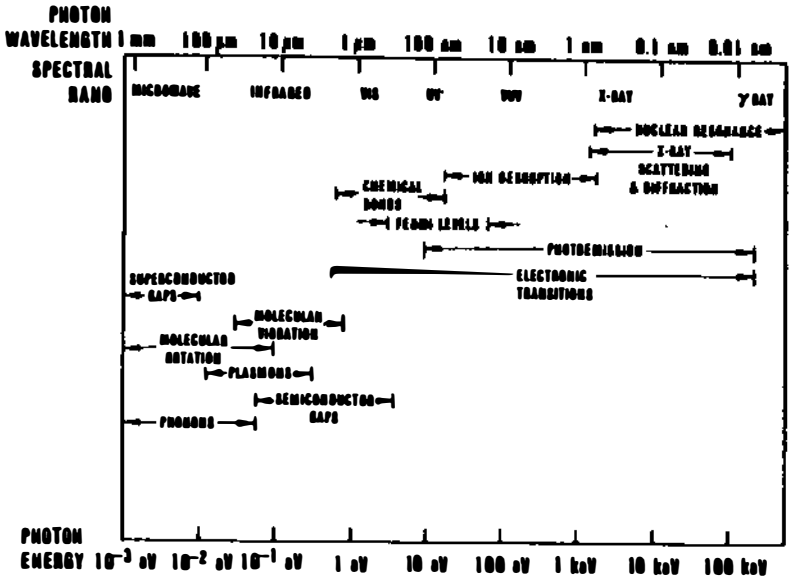

Figure 2.2. Energy range for basic processes.

Synchrotron Radiation Sources

The established tool for producing high brightness, partially coherent radiation below 100 nm is a synchrotron radiation source. The generation of radiation from high energy electrons that are deviated by bending magnets is well understood; these sources have made possible much of the XUV research in the past two decades. Efforts are now under way to make major advances in the capabilities of synchrotron radiation sources by using wiggler and undulator magnets, and to develop the related free electron laser.

An electron oscillating at frequency ν about a stationary center will produce radiation at a frequency ν and wavelength $\lambda_0 = c/\nu$. If the center of oscillation moves towards the observer with velocity v, then the radiation is Doppler shifted to a wavelength $\lambda = \lambda_0/2\gamma^2$ where $\gamma^2 = (1 - v^2/c^2)^{-1/2}$ and the electron energy is $E = \gamma m_0 c^2$. In the undulator source, the electrons circulating in the storage ring are made to pass through the field of an array of high field magnets of alternating polarity, the undulator magnet. The effect, as viewed by an observer watching the electron approach, is that of an oscillating electron approaching at a relativistic speed. If the length of the magnet period is λ_0, then the radiation observed in the laboratory will peak at $\lambda = \lambda_0/2\gamma^2$. For a 2 GeV electron ($\gamma$ = 2000) and a magnetic period of 1 cm, the radiation peaks at 1.25 nm (1 keV). The deviation of the electron at each magnet is small so that the radiation emitted along the beam adds coherently in the forward direction. The bandwidth of the radiation is $\Delta\lambda/\lambda = 1/N$, where N is the number of electrons in the beam.

Undulator radiation has been demonstrated, but it is not widely available and its properties have not been extensively evaluated experimentally. Existing current high brightness synchrotron radiation sources use wiggler magnets that produce large deviations of the electron beam. Wigglers have less severe experimental constraints than undulators because they do not maintain coherence in radiation from one period of the magnet to another.

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Undulator radiation will provide intense sources of XUV and soft x-ray radiation, but although they have a high degree of spatial coherence, their spectral bandwidth is wide and the temporal coherence is modest. Because an undulator in a high energy storage ring is such an intense radiator, the next obvious step is to surround it with the mirrors of an optical cavity. The result is the free electron laser. The effect of the cavity is to bunch the electrons so that there is not only coherence between the various periods of the magnet, but coherence in the radiation from separate electrons as well. The result is that the power scales as N^2n^2 where N is the number of magnetic periods and n is the number of electrons in the beam. Simultaneously the cavity introduces a memory in the radiation field which produces the long coherence lengths and narrow spectral bandwidths inherent in laser systems. Free electron lasers have been operated in the infrared spectral region where the anticipated major increase in source brightness should greatly expand the study of non-linear optical properties. The success of shorter wavelength free electron lasers, although a task of great technological difficulty and not at all assured, would provide a great advance in tunable sources.

XUV and Soft X-Ray Lasers

Fully coherent and fully controllable radiation below 100 nm will have to be derived from laser sources because the phase memory of a coherent system is inevitably connected with the sort of feedback mechanism provided by a laser cavity. At present the only coherent sources below 100 nm produce coherent XUV radiation by non-linear up conversion of visible radiation using gases and high intensity visible laser light. The radiation has the same temporal and spatial properties of the driving visible lasers, but at odd harmonics of the input frequency. (If several input beams are used, the outputs are at the sum and difference frequencies.) Non-linearities in rare gases have been used to generate radiation as short as 38 nm (32 eV) with conversion efficiencies up to 10^{-4}. Though these sources are useful, their intensities are modest and they provide only partial coverage of the XUV range.

Efforts are under way to achieve direct lasing action in the XUV and soft x-ray region. Formidable problems must be solved before a useful laser can be constructed below 100 nm, but the research has already demonstrated the ability to produce media with sufficient gain to give intense amplified spontaneous emission. These media could serve as the active elements within an x-ray laser cavity, or they could be combined as amplifiers for input beams produced by non-linear up conversion.

The difficulty in producing high gain media at short wavelengths stems from the fact that excited state lifetimes scale as λ^{-3}. At shorter wavelengths it is necessary to create the excited atoms in correspondingly shorter times in order to produce macroscopic gain before the system decays spontaneously. Thus the creation of a short wavelength laser depends on the ability to pump excited states rapidly. Most current efforts use short pulses of intense, long wavelength lasers to provide this pumping.

Demonstration of stimulated emission at 20.6 nm by researchers at Lawrence Livermore Laboratories has been accomplished using a sub-nanosecond kilojoule pulse of 1.06 micron laser radiation to vaporize a selenium foil target. Fast electrons in the expanding metal plasma collisionally excited neon-like selenium to create an inversion between excited states with a gain coefficient of 10 cm^{-1} over a length of 1 cm. The gain was sufficient to produce a collimated beam of stimulated radiation a factor of 10^3 more intense than the background emission.

Another approach, pursued at Princeton Plasma Physics Laboratories, employs a kilojoule CO_2 laser pulse at 10.6 micron of 10^{-7} sec duration to vaporize a carbon fiber. Recombination of ions and electrons during expansion of the plasma produces an inversion between excited states

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of the hydrogen-like carbon at 18.2 nm. The measured gain coefficient was 6 cm^{-1} over a 1 cm pulse. Again, a large enhancement of intensity was observed.

A third successful demonstration of gain below 100 nm has been carried out at the University of Illinois at Chicago. Krypton gas was irradiated by an intense, short pulse (5×10^{-12}sec) of 93 nm radiation. An inner shell excitation in krypton ($4s\,4p^6 5d$) was produced by four photon absorption. Intense, collimated light at 193 nm was observed, suggesting high gain on the $4s\,4p^6 4d \rightarrow 4s^2 4p^5 4d$ transition between excited states of krypton.

Research groups in the U.S. and abroad are attempting to observe stimulated emission below 100 nm, and it is likely that some of the proposed schemes will succeed. One goal is to produce shorter wavelength radiation, another is to achieve laser emission using smaller, more efficient pumping lasers. Work is under way on the development of optical components for cavities and beam optics to use with XUV and x-ray systems. Because the photon energies involved are larger than the work functions of all materials, the development of optical components demands major advances in material science and technology.

SUMMARY

The advances in modern light sources are sometimes not apparent to a casual observer because the total output of a modern source is often modest. The principal advance is that the radiation from the new sources is emitted in narrow spatial and temporal domains. This control is expressed in the concept of brightness: Figure 2.3 shows a comparison of the brightness of old and new radiation sources. Note that the scale of brightness covers a range of 10^{32}. It is evident that the advances have been tremendous.

Our control over radiation is greatest in the visible region where complete spatial and spectral coherence can often be achieved. The high intensities and short durations resulting from this coherence have opened new regimes of atomic and molecular physics. Many of the features of these sources are available in the infrared, although sources of higher intensity are still being sought to study non-linear processes.

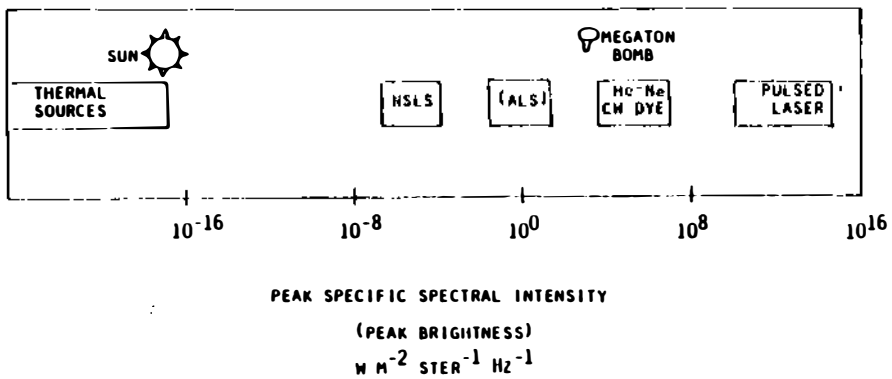

Figure 2.3 Brightness of some light sources. NSLS is the National Synchrotron Light Source; ALS is the proposed Advanced Light Source.

In the short wavelength XUV and soft x-ray regions, only a small fraction of the control possible at longer wavelengths is available. Because many of the problems on relaxation processes,

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material structure and non-linear interactions can only be addressed by using XUV and soft x-ray radiation, a major effort in optical science is under way to produce sources in this spectral region that are comparable to those that have been achieved in the visible spectral region.

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3. TRAPPED AND LASER-COOLED PARTICLES

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INTRODUCTION

Charged and neutral particles can now be trapped electromagnetically and cooled to the millikelvin regime with laser light. Trapped particles provide a unique medium for ultra-high precision spectroscopy; they make research possible on problems such as collisions at ultra-low energy and strongly coupled plasmas, and they open the way to new types of atomic clocks and optical frequency standards. This paper surveys the rapidly developing field of trapped and cooled particles, and some of the new scientific opportunities it provides.

THE EFFECTS OF THERMAL MOTION

Atoms and molecules are in constant thermal motion. At room temperature, the molecules of a gas such as air have an average speed of about 500 m/sec — more than the speed of sound — and the directions and magnitudes of their velocities are random. The spread of speeds is about as large as the average velocity itself. For scientists trying to make precise and well controlled measurements on atoms, this thermal motion can be disastrous.

The performance of atomic clocks, which are the best time keepers as well as the most accurate standards of frequency, is fundamentally limited by motional effects. These clocks make possible refined experiments on relativity, and they are essential for applications such as navigation on earth and in space, and encoding and decoding of secure communications. Experiments on collisions of atoms that seek to elucidate atomic interactions or chemical processes are often hindered by poor control of the velocities of the colliding atoms. The result is a serious impediment to precise tests of collision theory. Spectroscopy and experiments on the deflection of atomic beams are also severely affected by thermal motion.

As emphasized by R.N. Zare in Chapter 1, spectroscopic measurements are our primary source of information about the structure and dynamics of atoms, molecules and ions. Motion degrades spectroscopic measurements, for just as the apparent pitch of a train's whistle changes when the train moves toward or away from the listener, an atom's frequency is shifted by its motion. Random thermal motions produce a jumble of frequencies that limits the precision of measurement. Time dilation — the second order relativistic effect that makes moving clocks run slow — decreases all atomic frequencies by varying amounts. Motion also affects the precision of spectroscopic measurements by limiting the observation time: rapidly moving atoms spend only a short time in a measuring apparatus, and the shorter the time the less precise the measurement will be.

One approach to achieving a long observation time is to confine atoms in a small bottle. If the bottle is refrigerated, the atoms' thermal motion can be reduced. This procedure is useful in special situations, as in the hydrogen maser, but it is ultimately limited by the fact that collisions with the walls of the bottle perturb the atoms. Furthermore, cooling eventually causes the atoms to condense on the walls. The question, then, is how to confine atoms without a material bottle,

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and how to cool atoms without cold surfaces. The answers are electromagnetic traps and laser cooling.

CHARGED PARTICLE TRAPS

An electromagnetic particle trap is a configuration of electromagnetic fields that confines atoms without allowing them to hit surfaces. The electromagnetic "walls" of the trap may be static or oscillating electric or magnetic fields. (Light beams can also form a trap, as will be described below.)

The most widely used traps are for charged particles such as ions or electrons. The strong interaction between electric fields and charged particles makes it easy to trap ions using only modest electric potentials — as low as a few volts. A number of different ion traps have been used,[1] but only the Penning and Paul designs will be discussed here.[2]

Figure 3.1 shows the electrode configuration. In a Penning trap for positive ions the horizontal ring electrode is negatively charged and the cap electrodes, along the vertical axis, are positively charged. A positive ion moving vertically near the trap center is repelled from either cap, and forced back to the center. If the ion moves horizontally, it is attracted to the ring, away from the center, but the vertical magnetic field, B, prevents its escape by exerting a force perpendicular to the motion.

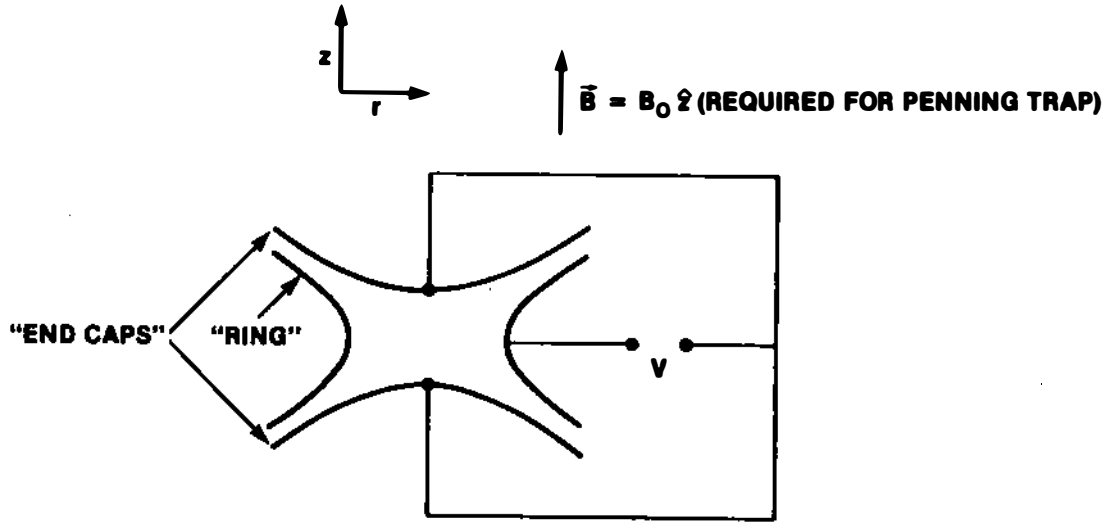

Figure 3.1. Electrode configuration for a Penning or rf (Paul) trap. The applied voltage is static for a Penning trap and oscillating for an rf trap. The trap is a figure of revolution about the z axis.

The Paul or rf trap is physically similar to the Penning trap, but lacks the magnetic field. An oscillating voltage is applied between the caps and ring and the ion oscillates under its influence. Sometimes the ion is pushed toward the center and sometimes away, but the average force is always toward the center of the trap. Ions can be trapped for long periods in either Penning or Paul traps: days or weeks are common, and a single electron was stored in a trap at the University of

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Washington for over nine months.[3]

The spectroscopy of trapped ions is nearly free of perturbations. Stark shifts due to the small electric field near the trap's center can be small and observation times can be long. Even single ions can be trapped and observed, eliminating the effects of collisions. However, when the ions are first trapped they are hot, and all the spectroscopic problems due to thermal motion occur. In such an isolated environment, how can kinetic energy be removed from the ions so as to cool them?

LASER COOLING

Laser cooling is a process for reducing the energy of ions or atoms by using radiation pressure. Figure 3.2 shows a moving atom irradiated by a laser beam directed against its motion. If the atom absorbs a photon from the laser beam, it also absorbs the momentum of the photon and recoils. This slows the atom by a few centimeters/second out of a typical thermal velocity of about 500 meters/second. The excited atom reradiates the photon and recoils once again, but successive photons are equally likely to be emitted in one direction as another and the average recoil due to radiation vanishes. In this way, after repeated absorptions and emissions, the atom can be brought nearly to rest.

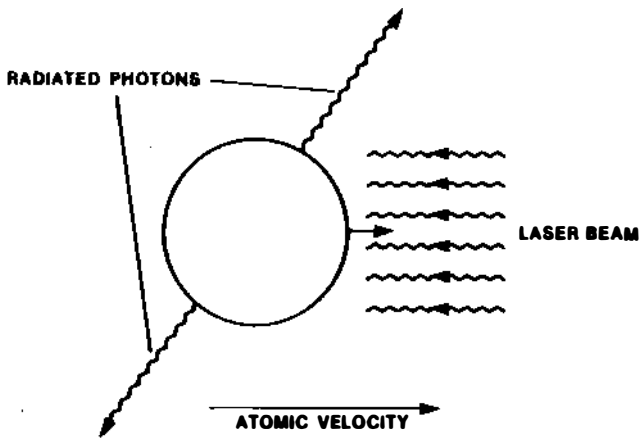

Figure 3.2. Laser cooling: momentum absorbed from the laser beam slows the atom. The momentum of the radiated photons averages to zero.

Atoms in a trap are as likely to be moving away from the laser as toward it, and in the former case the photon recoil would increase their velocity. However, atoms absorb laser light efficiently only if the frequency of the light lies within the natural linewidth of the transition, typically 10 MHz. The Doppler shift due to normal thermal motion of an atom is much larger than this. Thus, only atoms with a particular velocity will absorb light. If the laser frequency is tuned slightly below the resonant frequency for atoms at rest, only atoms moving toward the laser will absorb many photons.[4,5]

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Laser cooling of ions

Laser cooling was first demonstrated with trapped ions by groups at the National Bureau of Standards in Boulder[6] and at the University of Heidelburg in West Germany[7] in 1978.

Researchers at the University of Washington have observed ion temperatures that are no higher than 10 mK,[8] a thirty-thousand-fold reduction in energy from room temperature. Spectroscopy on such cold, trapped ions is virtually free of the motional effects that limit the resolution at ordinary temperatures. The ultimate low temperature for a typical ion is predicted to be a few hundred microkelvin.

In a recent experiment,[9] an oscillator was locked to a 303 MHz transition in laser-cooled, trapped $^{9}Be^{+}$. The error due to all known causes was estimated to be 9×10^{14}. This is about the same as the error in the U.S. cesium clock, the primary time standard for the nation and one of the most accurate clocks in existence. This truly remarkable accuracy in spite of the ion clock's low frequency (the Cs standard operates at about 9 GHz) is achieved because of the very long observation, 20 seconds, compared to 20 milliseconds for the Cs clock. The residual first order Doppler effects that limit the performance of the Cs standard are virtually absent because the average velocity of the trapped ions is zero.

Recently, an ion has been cooled and confined so tightly that it is restricted to a region of space smaller than a wavelength of light.[10] This is known as the Lamb-Dicke condition and results in the complete disappearance of the first order Doppler effect.

Laser cooling of neutral atoms

It is difficult to confine atoms because they are electrically neutral. The only forces that can be applied have to originate in the interaction between dipole or higher moments of the atoms and gradients in electric or magnetic fields. These forces are minute compared to the electric force that can be applied to ions. As a result, the only practical traps for ground state atoms require that their temperature be no more than a few degrees above absolute zero. The fundamental difficulty in trapping atoms is that atoms cannot be confined if they are hot, but they are difficult to cool if they are not confined. This dilemma has been solved by the development of efficient techniques for decelerating and cooling atomic beams.

To understand the problem of cooling an atomic beam, consider again the situation in Figure 3.2. The laser is directed against the atomic beam, so any photon absorption will slow the motion as desired. As an atom slows, however, its Doppler shift changes and it passes out of resonance with the laser. As this happens, the rate of deceleration decreases rapidly. (In a trap, this is of little consequence, since the particle's velocity constantly oscillates and the time available for cooling is very long.) To overcome this difficulty for free atoms, where the interaction time is limited, the changing Doppler shift must be compensated so as to keep the atom in resonance with the laser.

Two methods have been used to compensate the changing Doppler shift. The first is to change the frequency of the laser in time as the atoms decelerate.[11] The second is to change the resonance frequency of the atoms as a function of position, using an externally applied field.[12] Both of these methods have now been demonstrated[12–16] in experiments at the National Bureau of Standards in Gaithersburg and in Boulder.

The average velocity of an atomic beam can now be brought to zero and the spread of velocities substantially reduced. A fully state selected atomic beam with a velocity spread equal to 2% of its central velocity of 700 m/s, and a density per unit velocity 20 times higher than for a thermal beam, has been produced,[13] and samples of atoms with velocity spreads of only a few

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meters/second at an average velocity near zero have been created.[17]

A single laser beam cools only a single component of the motion of an atomic beam. With multiple laser beams, however, cooling can be accomplished in every direction. In an experiment at AT&T Bell Labs[18] atoms in a beam were first slowed in one dimension and then cooled three dimensionally with three perpendicular pairs of counterpropagating beams. The result was a sample of atoms with a measured temperature of 240 microkelvin. The cooling is so effective that any atomic motion is quickly damped: the atoms act as if they are immersed in a viscous fluid. In spite of the fact that there is actually no restoring force, atoms with typical instantaneous velocities of 0.5 meter/second require a tenth of a second to diffuse 4 mm.

Using three dimensional cooling as a "Maxwell demon" to collect, concentrate and cool atoms could lead to a simple, high density source of very cold atoms.[19]

TRAPPING NEUTRAL ATOMS

Electromagnetic traps are capable of confining neutral atoms provided that they are sufficiently cooled, and atom trapping has been achieved with a magnetostatic trap. In a recent experiment,[20] forces arising from the interaction of an atom's magnetic moment with an applied inhomogeneous field were used to confine atoms to a region of low magnetic field.

The magnetic trap is shown in Figure 3.3. A slow, laser-cooled atomic sodium beam is directed along the axis of a pair of coils with current in opposite directions. The field magnitude has a minimum at the center, forming a trap. Atoms are stopped in the center of the coils by a pulse of laser light and are trapped by the magnetic field. The velocity of the trapped atoms is no greater than 4 m/sec, and the average energy is about 10 millikelvin. The storage time, about one second, is limited only by collisions due to the imperfect vacuum.

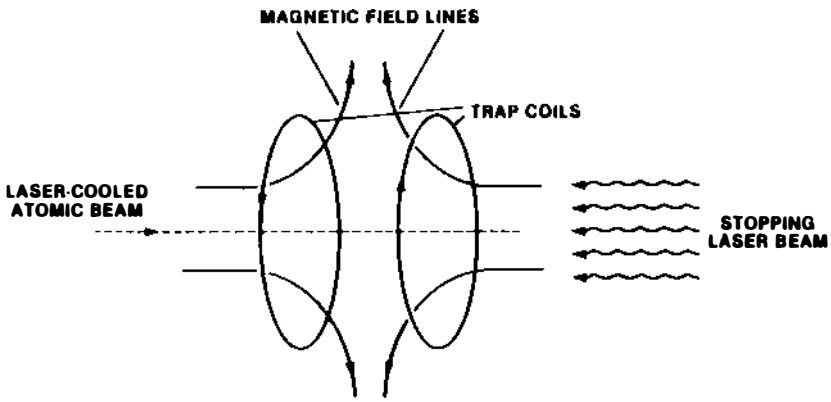

Figure 3.3. The first trap for neutral atoms. A laser-cooled atomic beam, incident from the left, is stopped by a pulse of laser light from the right. The stopped atoms are trapped near the magnetic field minimum formed at the center of the two opposed trap coils.

Other traps for neutral atoms are possible.[19,21] Of particular interest are laser traps that rely on radiation pressure and/or dipole forces to confine atoms. Dipole forces arise from the interaction between induced optical dipole moments and gradients of optical fields. Traps based on these forces are of particular interest because of theoretical controversy over their stability. The availability of cold atoms gives promise that laser traps will be demonstrated in the near future.

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Neutral particle traps share the disadvantage that they strongly perturb the atoms they contain, often making them unsuitable for high-resolution spectroscopy. Nevertheless, they are useful for collecting and refrigerating atoms. Proposals for ultra-refrigeration of neutral atoms in magnetic traps[22] indicate that temperatures in the microkelvin regime should be possible. This makes such traps potentially valuable as sources of extremely cold atoms for spectroscopic studies.

Most laser trapping experiments use expensive, delicate and cumbersome ion-laser-pumped dye-lasers. A recent and exciting development is the deceleration and stopping of a beam of cesium atoms using diode lasers.[23] Diode lasers are cheap, small, and reliable; this development greatly facilitates laser cooling techniques.

Compared to ion cooling and trapping, neutral atom cooling and trapping is still in an early stage. Development of atom-trapping techniques can be expected to complement the ion work. Neutrals can be trapped at higher densities than ions because of the absence of space charge. Neutrals can be used to study electric field effects, whereas ions are necessarily trapped in zero average electric field. The beam cooling techniques developed for neutral atoms will be useful where beams, as opposed to traps, are needed.

FUTURE DIRECTIONS

What are the future prospects and the expected benefits of research on laser cooled and trapped particles? Trapped ions have already demonstrated their potential as atomic clocks, giving accuracy comparable to the best existing beams clocks. A frequency standard that is accurate to 10^{-15} appears to be feasible. By extending the clock frequency into the optical spectrum, it may be possible to achieve an accuracy of 10^{-18}, equivalent to about one second in the age of the universe! An encouraging development in this direction is the recent success at NBS Boulder in cooling trapped mercury ions. Mercury is an excellent candidate for both microwave and optical frequency standards.

The coming generation of atomic clocks will have important practical and scientific applications. Field operational clocks for military global positioning systems need to be about as good as the existing primary standards:[24] this cannot be accomplished reliably without creating a better primary standard. Proposed systems for secure communications require greater stability than currently possible. Thus improvements to the 10^{-15} level that ion standards promise would be immediately useful for navigation and communication systems.

Scientific interest in better atomic clocks is high for they would allow better tests of general relativity and other gravitational theories. The gravitational red shift could be measured more accurately by comparing clock rates in different gravitational potentials. Spatial isotropy could be tested by measuring the clock rate as a function of orientation as has recently been done using a Be^{+} ion clock.[25] Along the same lines are tests for proposed spin-gravity interactions.

Other research with trapped and cooled particles includes the study of nuclear spatial distortion by external magnetic fields (nuclear diamagnetism), comparisons of the frequencies of distinct but presumably identical atoms as a test of quantum mechanical indistinguishability and absence of hidden variables, the search for atomic electric dipole moments as a test of time reversal invariance, measurements of g-factor ratios, tests of the influence of cavities on atomic frequencies, and the precise frequency measurements of the hydrogen spectrum as a test of quantum electrodynamics.

Another class of experiments centers on the physics of the trapping process. Atoms can be trapped at high densities and very low temperatures. This is particularly important in the search for Bose condensation.[26] Present indications are that the presence of walls limits the achievable

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density in systems of spin polarized hydrogen, and researchers are looking at atom traps as a possible solution.[27] Densely trapped ions may lead to strongly coupled non-neutral plasmas which can behave like liquids or solids.[28] On a more speculative note, atom traps could serve to store antihydrogen. Trapped ions can be used for mass spectroscopy, and trapped particles can be used to study the limits of cooling for comparison with a large body of theoretical work.

Laser-cooled monoenergetic atomic beams make it possible to study the quantum statistics of photon absorption by measuring the deflection of atoms due to photon momentum transfer. (Such quantum statistics can also be studied with single atoms or ions in traps[29] where there is the advantage of being able to look for correlation effects for long times.) Very slow beams can be used to test charge neutrality by searching for beam deflection under the influence of electric fields. Laser cooling can provide atomic beams of unprecedented quality for collision experiments and for atom-surface studies.

In summary, the opportunities for work in the laser cooling and trapping field are considerable. Ion trapping and cooling is a well demonstrated technique that is now ripe for a host of new applications and developments. Neutral trapping and cooling is in its early stages with many breakthroughs within reaching distance. Potential applications of these techniques range from navigation to chemistry and from general relativity to quantum statistics.

The author would like to thank D. Wineland, J. Hall and W. Nagourney for their contributions to this report.

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4. INTENSE ELECTROMAGNETIC FIELDS IN LASER-ATOMIC INTERACTIONS

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INTRODUCTION

Lasers are widely used to probe atomic and molecular structure and interactions but their intensity is generally so low that the light only weakly perturbs the structure of the atom under study. However, high intensity lasers have become available and their light strongly affects atomic structure. The study of atoms in very intense light has led to the discovery of unexpected and interesting new phenomena and opened a new area in the physics of light-matter interactions.

The electric fields produced by this new generation of lasers are gigantic: they can exceed the "natural" atomic unit of electric field 5×10^{9}V/cm^{2}, which is the electric field due to the proton at the position of the smallest Bohr orbit in hydrogen. Because this nuclear electric field determines the gross structure of atoms, it is not surprising that structure and interactions of an atom subjected to an atomic unit of external field are greatly altered. Such a field is produced by a laser with an intensity of about 10^{16} watts/cm^{2}, and today's lasers can produce intensities up to ten times that value.

This paper reviews some of the new atomic phenomena that have been observed using high intensity lasers. The discussion here will concentrate on multiphoton ionization processes. In an n-photon ionization process, an atom simultaneously absorbs n photons and is ionized. The number n is the smallest integer such that $n\,h\nu$ (ν is the frequency of the laser photon, and h is Planck's constant) exceeds the atom's ionization energy. This is usually the dominant process when atoms or molecules are subjected to a very intense laser light and it is important in problems that range from the very applied to the very basic.

"Strong field" effects are expected at the largest laser intensities that can be achieved in laboratories, 10^{16}-10^{17} watts/cm^{2}, but they have been discovered to begin at intensities that are one hundred to one thousand times smaller. This can be understood by a greatly simplified picture of multiphoton ionization.[1] In the low field limit, the rate (i.e. the probability per unit time) for n-photon ionization, W_n, can be written

$$W_n = \sigma_n F^n ,$$

where σ_n, the generalized absorption cross-section, is a parameter which contains all of the information relating to the specific atom being ionized, and F is the laser flux in photons per square centimeter per second. The rate for a 1-photon ionization, when that is energetically possible, is $W = \sigma_1 F$, where σ_1 is typically 10^{-17} cm^{2}. Roughly speaking, the rate for a two-photon ionization is then given by

$$W_2 = \sigma_1 F (\sigma_1 F t_0) ,$$

where t is a characteristic time which describes how soon the second photon must arrive after the absorption of the first photon if it, too, is to be absorbed. As an estimate of this time, one can take $t_0 = 1/\nu$. In a classical picture ν is the frequency at which the atom oscillates between the

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ground state and an excited state due to the driving of the photon field. A typical value of t is then 10^{-15} seconds. Continuing in this way, we can approximate the probability per unit time of an n photon process by

$$W_n = \sigma_1 F (\sigma_1 F t_0)^{n-1} . \tag{1}$$

This simple argument produces probabilities of roughly the correct magnitude. For our purposes, there are two important points to be taken from this result. The first is that at an intensity of 10^{14} watts/cm^2, which corresponds to a flux of roughly 10^{32} photons/cm^{2}sec,

$$\sigma_1 F t_0 = 1 ,$$

which, according to Eq. 1, implies that the multiphoton absorption rate, W_n, is independent of n, i.e., independent of the number of photons being absorbed. This rough equality of the probability for processes involving different numbers of photons is a key characteristic of "strong-field effects."

The second point follows from rewriting the generalized cross section as

$$\sigma_n = (\sigma_1 t_0^{(n-1)/n})^n . \tag{2}$$

When n is small the expression in parenthesis decreases rapidly with increasing n, but for large n (n greater than 10, say) it becomes almost independent of n. This implies that σ_n drops rapidly as n increases when n is small, but drops slowly for large values of n. This result tells us that if we extrapolate to large n the values of σ_n calculated for small n, we will obtain values that are much too small and will consequently underestimate the probability for processes involving many photons.

DESTRUCTION OF RESONANCE STRUCTURE

The probability for n-photon ionization increases by several orders of magnitude when the atom has an energy level that can be excited by absorbing an integral number of photons ($n-1$, say). This process is called multiphoton resonance. By changing the frequency of the laser light one can tune from the "off-resonance" situation described above, to the resonance, and off-resonance again. One can thus determine the photon frequency at which the resonance occurs, and consequently the energy of the excited state of the atom.

Lompre *et al.*[2] carried out such an experiment using krypton. For their laser, 12 photons are required to ionize krypton atoms, and there is an excited state of krypton such that there is an 11-photon resonance in the ionization. Figure 4.1 shows a sketch[1] of a typical resonance at a laser intensity of 10^{12} watts/cm^2. (The number of krypton ions detected is plotted as a function of frequency of the laser.) The resonance can be fit approximately by a Lorenzian curve having the form

$$N_{ions} = \frac{A}{(\omega - \omega_o + \alpha I)^2 + \beta I^2} . \tag{3}$$

In this expression, αI represents the relative AC-Stark shift of the ground and resonance excited states produced by the interaction of the atom with the laser field, and βI^2 represents the AC-Stark width of the resonant state. The laser is "in resonance" when N_{ions} is at a maximum as a function of the laser frequency ω, i.e. when the term in parenthesis in Eq. 3 vanishes. Figure 4.1 also shows the results for a similar scan of laser frequency when the power was increased to 10^{13} watts/cm^2:

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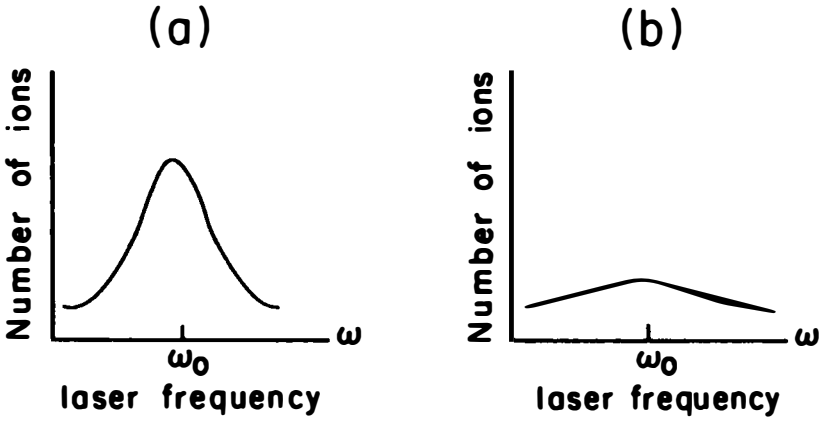

Figure 4.1. 11-photon resonance in 12-photon ionization of Kr.[1] (a) 10^{12} W/cm^2; (b) 10^{13} W/cm^2.

the resonance state of the atom has disappeared at the higher intensity!

One can understand the disappearance of resonance at higher intensity by simple arguments. Two effects contribute to this phenomenon. First, at high intensity, the Stark broadening term (βI^2) becomes very important, and the resonance becomes so broad that it scarcely shows a maximum. Second, in a real experiment with a pulsed laser, the intensity of the light is not constant. During the pulse the intensity grows from zero to its maximum value, then decreases to zero again. While this occurs, the Stark shift term in Eq. 3, (αI), varies in magnitude. Consequently, the laser, which has a fixed frequency, is in resonance only for a small fraction of the laser pulse. The first of these effects represents a real "destruction" of the excited, resonant states of the atom; the second is an experimental artifact, albeit an artifact which cannot at this time be avoided.

ABOVE THRESHOLD IONIZATION

Above threshold ionization (ATI) is the process in which an atom is ionized by absorbing more than the minimum number of photons for photoionization, *(n)*. The extra energy absorbed goes into the kinetic energy of the ejected photoelectrons; the signature of this process is a photoelectron energy spectrum which contains "clumps" of photoelectrons separated in energy by the energy of a photon.

In the absence of any real calculations, theorists expected that the excitation rate for the processes in which $n+s$ photons are absorbed could be described by

$$W_{n+s} = \sigma_{n+s} I^{n+s} , \tag{4}$$

where

$$\frac{\sigma_{n+s}}{\sigma_{n+s-1}} << 1 .$$

These expectations appeared to be borne out in results obtained by Agostini *et al.*[3] However, in a more recent experiment performed by Kruit *et al.*,[4] it was found that with increasing laser intensity, the peaks corresponding to higher order processes can dominate peaks due to lower order processes (Fig. 4.2). The lowest order peak, corresponding to $s=0$, actually disappears at high

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intensities!

Even more surprising were the values obtained for the effective orders of nonlinearity for the various peaks shown in Figure 4.2.

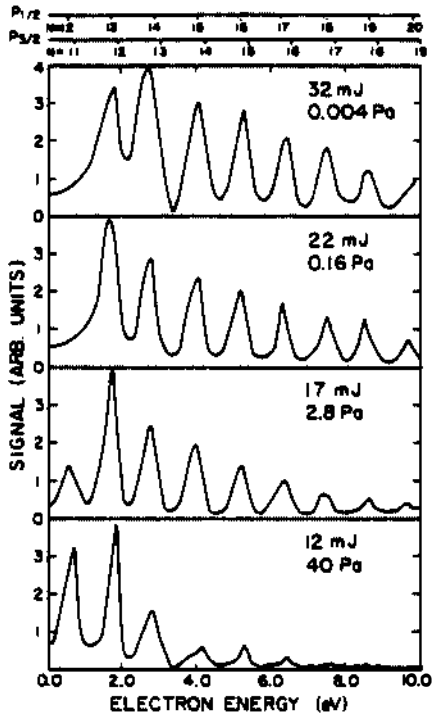

Figure 4.2. Photoelectron energy spectrum of Xe.[4]

The effective order of nonlinearity is the slope on a log-log plot of the size of the peak vs. the intensity of the laser. If (4) is correct, the number of ions in the $n+s$ peak is

$$N_{n+s} = W_{n+s}T = \sigma_{n+s} F^{n+s} T \ , \tag{5}$$

where T is the duration of the pulse. Using (5), one finds that the effective order of nonlinearity k_{n+s} of the $n+s$ peak is

$$k_{n+s} = n+s \ . \tag{6}$$

The results found by Kruit *et al.* are shown in Table 4.1. It is clear that Eq. 6 does not describe the experimental observations. In fact, with the exception of k_{11}, the measured values of k_{n+s} satisfy the mysterious inequality

$$n-1<k_{n+s}<n \ .$$

These results stimulated a great flurry of theoretical activity.[5] Most of the calculations led to the conclusion that these strange results could be explained if the laser intensity in the experiments was great enough for perturbation theory to break down (this is roughly equivalent to the statement that effects of all orders become equally probable). The techniques needed for non-perturbative

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Table 4.1. Measured effective orders of nonlinearity in Xe (n=11).

$n+s$	11	12	13	14	15	16	17	18	19
k	7.7	10.0	10.2	10.4	10.8	10.5	10.5	10.6	10.8

calculations were too complicated to be applied to real atomic wavefunctions and the complex structure of a real atom. Thus, all of these calculations used simplified model atoms, and it is not possible to claim that they actually reproduced the experimental results but only that they were consistent with these results.

Recently the assumptions made in these calculations have been called into question by new experimental results obtained by Mainfray,[6] which are shown in Fig. 4.3.

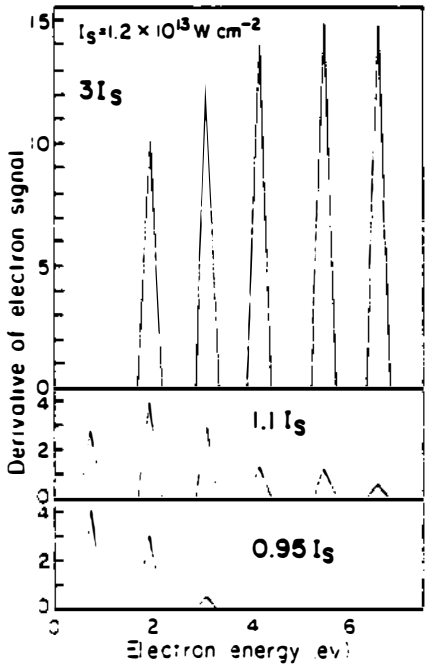

Figure 4.3. Photoelectron energy spectrum of Xe[6] I = 1.2 10^{13} W/cm^2.

These demonstrate the importance of what experimentalists call saturation. In this context, saturation refers to the depletion of atoms in the sample due to ionization: when saturation occurs, the observed ionization rate drops because all of the atoms in the sample have already been ionized. The results of Mainfray show that the photoelectron energy spectrum changes dramatically as the intensity passes from just below the saturation intensity to just above the saturation intensity. The theories described above do not recognize the saturation intensity as being critical in any way — the critical intensity according to these theories should be the intensity at which perturbation theory breaks down, an intensity which bears no direct relationship to saturation. A recent thermodynamic theory by Pan[7] does depend strongly on the saturation intensity, but at present one must view this problem as being unresolved.

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MULTIPLY CHARGED IONS

Recently, L'Huillier *et al.*[8] studied the formation of singly and doubly ionized xenon (Xe^+ and Xe^{++}) when neutral Xe was irradiated with a laser of wavelength 1064 nm and intensity in the range 10^{12}–10^{14}watts/cm^2. Their results are shown in Fig. 4.4.

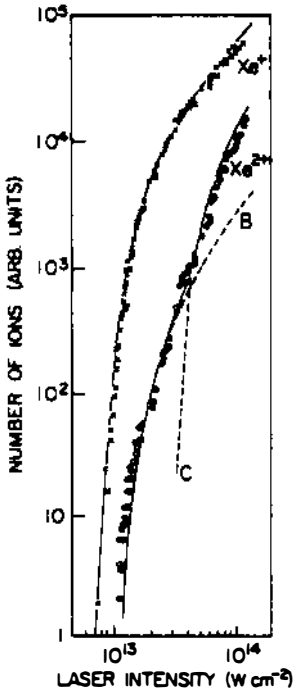

Figure 4.4. Ion formation in Xe as a function of intensity.[8]

They found that their results could not be described by equations which allow only for sequential ionization, i.e. Xe being ionized into Xe^+, which is then ionized into Xe^{++}. However, they were able to obtain excellent fits to the experimental data if they assumed the existence of a direct path from Xe to Xe^{++} which did not involve formation of Xe^+ as an intermediate step. In fact, they found that this direct process was much more likely than the sequential process at intensities below the saturation intensity. This is surprising since there is no obvious physical mechanism that predicts a large direct process of this type. Similar results have been obtained by this group for several other elements, so the phenomenon does not seem to be tied to any specific characteristics of Xe. Solid theoretical explanations for the importance of this direct path are so far lacking.

A series of interesting experiments on the formation of multiply charged ions has been carried out by Rhodes and coworkers[9] using much higher intensities — 10^{15}–10^{17}watts/cm^2. Results for Xe using a 193 nm wavelength laser are shown in Fig. 4.5. Rhodes and coworkers[10] also studied the photoelectron spectra produced during this high intensity pulse. The results, shown in Fig. 4.6, are similar to those obtained by Kruit *et al.*,[3] except that at higher intensities new lines, identified as being produced by Auger transitions, appear. This implies that at high intensities inner shell excitations are involved in the ionization process, at least in Xe, supporting the idea that some type of collective behavior of the atom is involved. The explanation proposed by Boyer and Rhodes[11] is that a collective motion of the electrons is driven by the intense electric fields of the laser, and that this ultimately results in the ejection of several electrons from the neutral atom.

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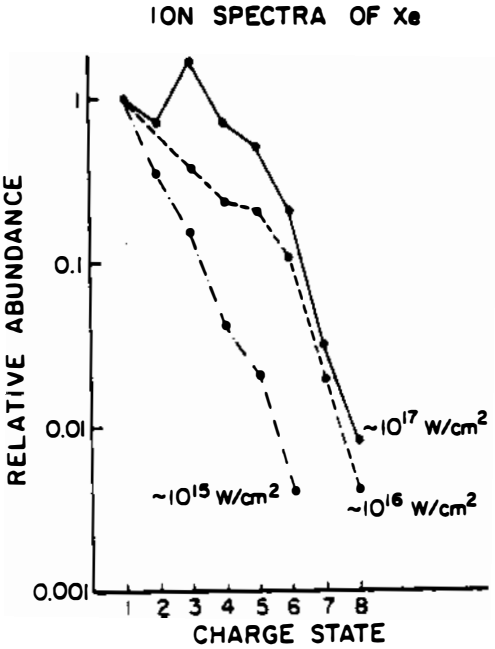

Figure 4.5. Ion formation in Xe as a function of intensity.[9]

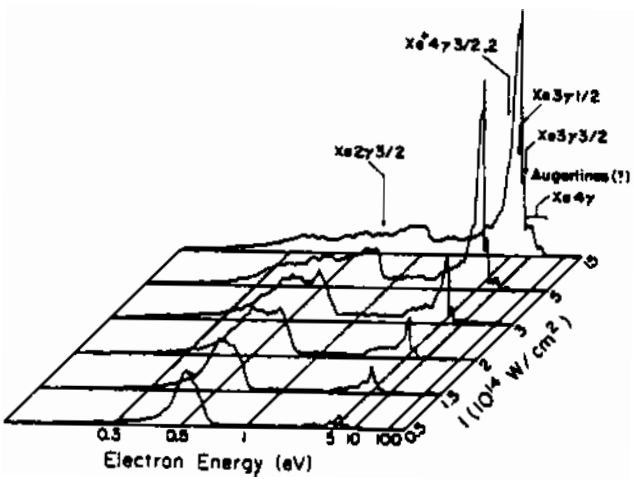

Figure 4.6. Photoelectron energy spectrum of Xe.[10]

On the other hand, Lambropoulos[12] has emphasized the key role that saturation and the shape of the laser pulse play these experiments. In particular, by estimating the time required to completely saturate the Xe to Xe^+ ionization at 10^{13} watts/cm^2 and the time to saturate the Xe^+ to Xe^{++} ionization at 10^2watts/cm^2, he argues that the Xe atom will never "see" the high intensities (10^{16}watts/cm^2) of Rhodes' experiments. Rather, there will be a sequence of ionization

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processes occurring during the rise of the laser pulse, so that only the higher states of ionization exist by the time the pulse reaches its peak intensity. Finally, he argues that the probabilities for sequential ionization (rather than some type of direct, collective ionization) have been underestimated.

A few calculations [13,14] have appeared that attempt to explain these observations qualitatively. Because of the complexity of the problem, these theories have been statistical in nature. Geltman [13] has emphasized that the results in Fig. 4.5 cannot be explained using lowest-order perturbation theory: suppose that the number of ions produced in an n-photon process has the perturbation form

$$P_n = \sigma_n F^n T \ . \tag{7}$$

Consider the ratio of the number of ions produced in an n-photon process to the number of ions produced in a 2-photon process, two photons are the minimum number needed to ionize xenon at this wavelength:

$$R_n = P_n/P_2 \ ,$$

and, from Equation (7)

$$\frac{R_n(I=10^{15})}{R_n(I=10^{16})} = \left(\frac{10^{15}}{10^{16}} \right)^{n-2} \ . \tag{8}$$

Taking the specific value of n =27 in Eq. 8, one finds that the predicted ratio is 10^{-25}, while Fig. 4.5 shows that the experimental ratio is closer to 10^{-1}. Geltman then went on to evaluate statistically the probability that s electrons would be ejected from a shell having n electrons if each electron had the probability p of being ejected. By treating p as a variable parameter, he was able to obtain a reasonable fit to Fig. 4.5.

A similar approach was used by Crance [14], who calculated the statistical probability of q photons being absorbed by one of n electrons if N photons are absorbed by the atom. All structure and details of the atom are ignored in this calculation, the only atomic parameters appearing being r_0, the atomic radius, and E_0, the ionization energy of the atom or ion. This calculation reproduced reasonably well the Xe^{++} production probability at three different wavelengths.

CONCLUSION

Many unexpected effects have been seen in experiments on multiphoton ionization using high intensity lasers. Some understanding of the processes involved is beginning to develop, but the subject is still in its infancy. Considerable theoretical and experimental work is needed for the behavior of atoms in intense light to be understood.

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5. LASER SCIENCE IN INDUSTRY

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INTRODUCTION

Basic research is crucial to modern industry. By expanding our knowledge of the physical and chemical properties of materials basic research lays the groundwork for new technologies. In the past decade, laser science has become an important tool for the study of materials and their interaction with light. Refinements in the precision, sensitivity, spectral resolution and range of laser measurement techniques have led to dramatic advances in areas such as communications, materials processing, remote sensing, and metrology. It is not possible to survey this topic comprehensively within the limited scope of this short review. Instead, I will attempt to illustrate the excitement and promising new directions this work is taking by briefly describing four recent examples of basic research in laser science that have been pursued in industrial (and other) laboratories. These are: (1) femtosecond optical spectroscopy, (2) quantum fluctuations of the vacuum electromagnetic field ("squeezed states"), (3) solitons in optical fibers, and (4) laser photofragmentation studies of atomic clusters. In each case, I hope to illustrate how this work is relevant to the industrial context in which it is done. My comments are not specific to a particular industry although they are clearly biased by my familiarity with the industrial environment in which I work.

FEMTOSECOND OPTICAL SPECTROSCOPY

In the past few years there has been a dramatic improvement in the science of ultrafast optics.[1] Two key developments are the refinement of mode-locked dye laser technology and the use of pulse compression. In the first case, the introduction of group velocity dispersion compensation into the passively mode-locked rhodamine 6G ring laser has produced pulses as short as 27 fs (1 fs = 10^{-15} s[2]). This was achieved by inserting four glass prisms into the laser as illustrated in Figure 5.1a. By adjusting the prisms it is possible to introduce negative group velocity dispersion into the resonator and thereby compensate for the pulse broadening arising from the dispersive effects of the flowing dye streams and optical mirrors. The pulse width of this laser, as measured by the second harmonic autocorrelation technique is shown in Figure 5.1b. This is the shortest pulse width yet produced by a laser. Even shorter pulse widths have been produced, however, by compressing these pulses.[3] This method uses the nonlinear technique known as self-phase modulation to impose a frequency chirp on the pulse. This spectral modification produces a carrier frequency which decreases from the rear to the front of the pulse. If this "chirped" pulse is then sent through a material or optical component having negative group velocity dispersion the low frequency components on the leading edge of the pulse travel slower than the high frequency components on the trailing edge, thereby producing a compressed pulse. A pair of diffraction gratings is often used for negative group velocity dispersion. This technique has produced pulses as short as 8 fs.[4] At a wavelength of 625 nm, the duration of one optical cycle is 2 fs. These pulses, which are only 4 optical cycles long, have a large spectral width of approximately 10^{14} Hz (=100 nm) and are readily broadened by the dispersion of optical components such as mirrors and lenses.

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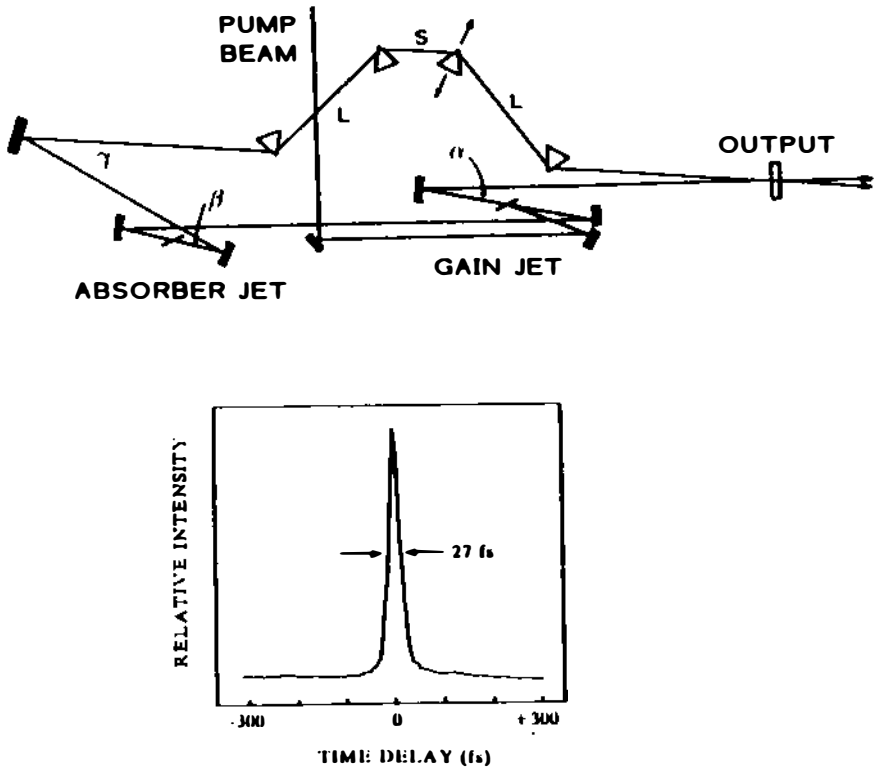

Figure 5.1 a (top) Schematic diagram of a colliding pulse mode-locked ring dye laser used to produce pulses of 27 fs duration.[2] The four prisms in the optical resonator are used to compensate the group velocity disperson due to other optical components. b (below) The optical pulses produced by the laser above as measured by second harmonic autocorrelation.

One application of femtosecond optics which illustrates its value to industry is the measurement of the electronic properties of high speed semiconductor devices and materials. This approach to high speed electronics has enabled measurements of devices and materials with speeds that greatly exceed the capabilities of conventional electronic measurement techniques.[5] Extension to femtosecond time scales has recently been achieved by the use of electro-optic materials. The nonlinear property of these materials known as optical rectification[6] permits the generation of extremely short electrical pulses from femtosecond optical pulses.[7] The bandwidth of these pulses extends from dc to the far-infrared and has a total range of as much as 5 THz. These pulses are a valuable source of radiation for both time-resolved and spectrally-resolved spectroscopy in a region of the electromagnetic spectrum where coherent sources are scarce.[8]

A recent application of these femtosecond electromagnetic pulses is the time-resolved excitation of coherent phonons in lithium tantalate.[9] This measurement shows clearly the influence of the ionic lattice vibrations on the electro-optic property of this material. As illustrated in Figure 5.2, individual cycles of the vibration of the ionic crystal lattice can be resolved. A speed limitation of approximately 150 fs was estimated for use of this material as an electro-optic modulator.

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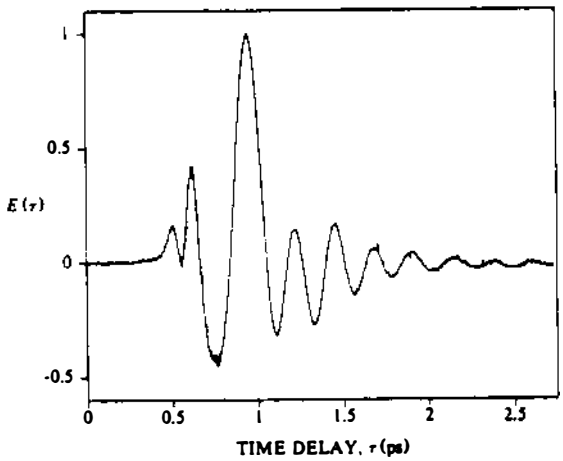

Figure 5.2. Ringing due to the ionic lattice vibrations of an electro-optic material (lithium tantalate) following impulsive excitation with a femtosecond optical pulse.[9] The coherent detection method used in this experiment has sufficient time resolution to resolve individual cycles of the lattice vibrations.

SOLITONS IN OPTICAL FIBERS

A soliton is a pulsed excitation of a material which propagates with either a stationary waveform or a waveform which periodically repeats itself with distance. The literature on solitons is extensive, with a wide variety of excitations in different material systems being reported. Optical solitons are most readily observed in single-mode optical fibers.[10] The physical basis for this type of soliton is a balance between negative group velocity dispersion and self-phase modulation. At wavelengths greater than approximately 1.3 μm the group velocity dispersion in single mode optical fibers is negative. This makes possible a pulse compression mechanism within the fiber similar to the method discussed in the previous section to produce short optical pulses by external pulse compression. The soliton effect is more complicated however, since the interaction between the self-phase modulation and negative group velocity dispersion occurs continuously over the length of the fiber, making possible stationary solitons. In addition to stationary solitons which retain their shape, higher order solitons also occur which repeat their waveform at periodic intervals along the fiber. These are illustrated in Figure 5.3.[11] These solitons were produced by injecting short optical pulses from a mode-locked color-center laser into a single-mode optical fiber. These results are in excellent agreement with theoretical models of optical solitons based on solutions of the nonlinear Schrodinger equation.[12]

Solitons have also been produced "inside" a laser resonator. This approach, known as a soliton laser,[13] has produced optical pulses as short as 130 fs at a wavelength of 1.5 μm.

Although the attenuation of optical pulses in glass fibers can be extremely low (less than 1 db/km), the resulting loss of amplitude over large distances can result in a non-stationary soliton arising from the decreasing amount of self-phase modulation due to the diminished pulse intensity. A method of compensating for this loss of amplitude has recently been demonstrated.[14] It uses Raman gain to amplify the pulses. This is accomplished by injecting a pump signal from a semiconductor at intervals along the fiber. The wavelength of the semiconductor laser is

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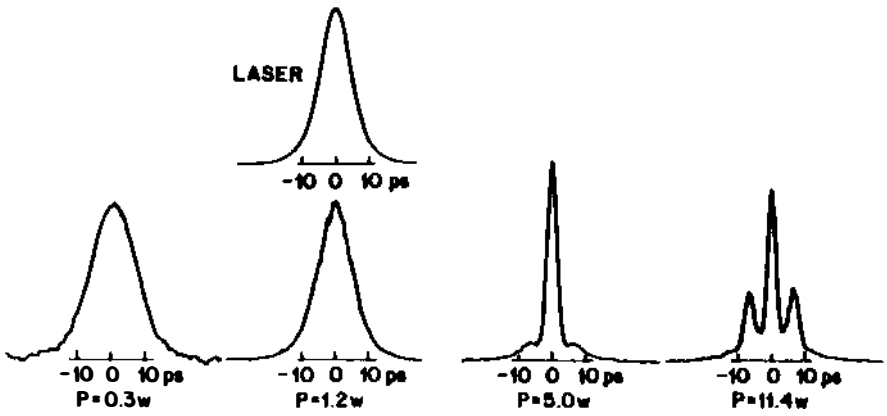

Figure 5.3. Experimental observation of solitons in single-mode optical fibers.[11] The top graph, labeled laser, shows the shape of the laser pulses launched into the fiber. The others show the shape of the pulses emerging from the fiber for various input powers. At low powers (0.3W) there is negligible change in shape. At higher powers, however, substantial pulse narrowing occurs due to soliton propagation. The length of the fiber was chosen to be one half the soliton period.

determined from the soliton wavelength by the stokes shift for Raman scattering in glass. This technique makes possible the use of solitons for optical communications. This unique property of propagating extremely short stationary pulses over long distances provides a promising method of using high bit rate transmission with single fibers.

QUANTUM FLUCTUATIONS OF THE VACUUM ELECTROMAGNETIC FIELD

A rigorous understanding of the sources and limitations of noise in optical signals is essential for achieving the optimum sensitivity both for experiments in basic research and for practical optical systems. In the latter case, noise levels determine the bit error rate in optical communications links, and minimum noise levels are important to obtain the maximum possible distance between repeaters.

In a semiclassical description, the noise arising from random excitations of a detector having quantized electron levels by a classical electromagnetic field is interpreted as a basic noise limit and is commonly referred to as "shot noise." In a fully quantum mechanical model, however, this noise arises from vacuum fluctuations of the electromagnetic field and satisfies a minimum uncertainty relationship of the form:

$$\Delta X_1 \cdot \Delta X_2 = \frac{1}{4}, \tag{1}$$

where X_1 and X_2 are the quantum mechanical operators describing the in-phase and quadrature components of the electric field:

$$E(t) = E_0\{X_1 \cos \omega t + X_2 \sin \omega t\}. \tag{2}$$

Coherent fields such as single mode laser light have equal variances for each of the two quadrature components (i.e., $\Delta X_1 = \Delta X_2 = 1/2$ in Eq. (1)). Squeezed states are a set of specially prepared

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states of the electromagnetic field in which the variance of one component is substantially less than the other.[15] In an equivalent representation, the uncertainty in the phase of an electromagnetic field may be made arbitrarily small at the expense of an arbitrarily large uncertainty in the amplitude (and vice versa). These squeezed states make possible the detection of optical signals below the semiclassical shot noise limit. As such, they are a unique manifestation of the quantum character of the electromagnetic field.

A number of experiments have recently been undertaken to prepare squeezed states and to measure their unique noise properties. An experiment by Slusher *et al.*[16] uses nondegenerate four-wave mixing to produce squeezed states in an optical cavity. The amplification of noise in one quadrature component of the field and de-amplification in the other due to optical parametric gain in sodium vapor is measured with a phase sensitive homodyne detector. They have recently observed a reduction in the noise level of the de-amplified quadrature component below the vacuum level. Although the reduction was relatively small (approximately 7%) it clearly demonstrates the attainment of "squeezing" below the vacuum noise level. A detailed theoretical description of their experiment which accounts for the subtle role of spontaneous emission gives good agreement with observation. In a related experiment, evidence for squeezing near the vacuum level has also been reported due to nondegenerate four-wave mixing in an optical fiber by Levenson and co-workers.[17]

LASER PHOTOFRAGMENTATION OF SILICON CLUSTERS

A thorough knowledge of the structure of semiconductor materials is essential to understand their physical properties and to exploit their unique capabilities for new electronics and optical technology. Much is now known about the bulk properties of the more commonly used semiconductors such as silicon, germanium and gallium arsenide. The surfaces of these materials are now under active investigation and a great deal of new information has been obtained with the aid of different surface analytic tools such as the recently invented scanning tunneling microscope. An exciting new frontier of investigation of the structure of semiconductors is the study of clusters consisting of small (e.g. 1 to 100) numbers of atoms. The symmetries and properties of these clusters are expected to be substantially different than bulk and 2-D surfaces and could have an important bearing on our understanding of how materials grow by such techniques as molecular beam epitaxy and chemical vapor deposition.

Smalley and co-workers[18] have demonstrated an effective technique for producing clusters by the vaporization of solid targets using pulsed lasers. The clusters produced by this method are usually neutral and have a wide distribution of numbers of atoms. Subsequent photoionization by a second pulsed UV laser permits them to be mass selected in a time of flight spectrometer so that clusters having the same numbers of atoms can be isolated and studied.

To obtain a better understanding of the stability and a clue to the structure of these clusters, Bloomfield, Freeman, and Brown[19] have developed a photofragmentation technique to study the distribution of numbers of atoms in the clusters. After selecting clusters of a specific size, they fragment them into smaller clusters using a pulsed laser. They find that certain sized clusters are easier to fragment and that the fragmentation products have a distribution of numbers of atoms that favor certain sizes. For example, when a Si_{12} cluster is fragmented, Si_6 and Si_{10} are much more likely to be produced than other sized clusters (Figure 5.4). These "magic numbers" are thought to be closely related to the stability of the bonding of the clusters. Similar results have also been obtained for clusters of carbon atoms. In a recent measurement, Smalley and co-workers[20] have shown a dramatic propensity for occurrence of the C_{60} cluster. They have hypothesized a highly stable and symmetric structure for C_{60} which has the appearance of the surface of a soccer ball.

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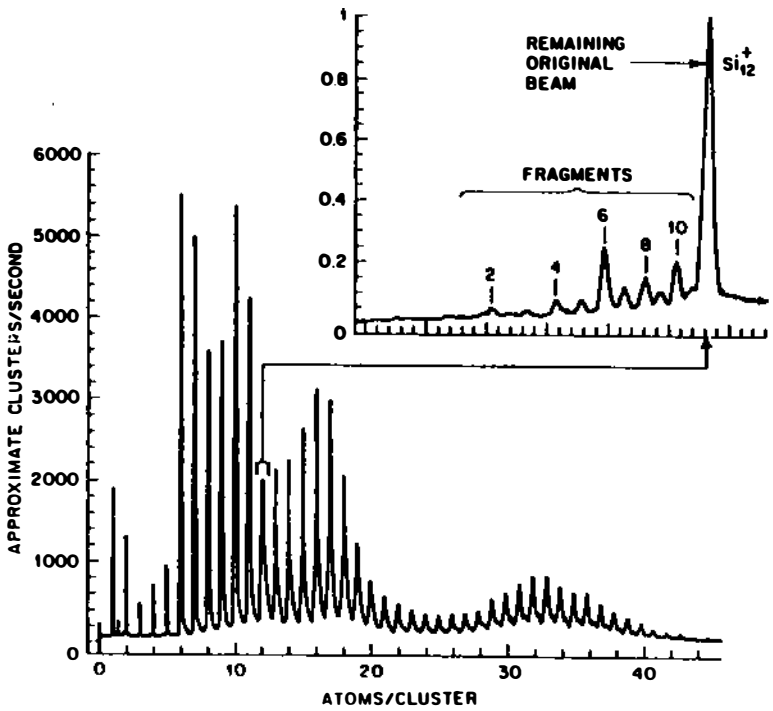

Figure 5.4 A spectrum of the distribution of silicon cluster sizes.[18] An example of the preselection of cluster size is shown in the inset where a single ion mass, Si_{12}^+, is isolated and fragmented with 266 nm radiation to produce the fragmentation spectrum shown.

DISCUSSION

The examples described in this short review were chosen to illustrate the excitement and vitality of basic research currently pursued in industry in the broad field of laser science. The work is not unique to industry and each example discussed here is also being pursued with equal vigor in many university and government research laboratories. What makes the industrial environment unique is the relevance of this work to the potential long term impact on technology. This not only provides the underlying motivation for pursuing this work but also provides the technical resources, materials, and tools for insuring the successful attainment of each particular research goal.

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6. NATIONAL PROGRAMS AT THE LASER-ATOMIC FRONTIER

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INTRODUCTION

In the 25 years since the laser was invented, lasers have been broadly applied to national programs such as energy, defense, the environment and health. The rapid development of lasers with tailored spectral, temporal, energetic and spatial properties has in many cases been directly motivated by these programs. A few examples of national programs whose success depends on advances at the laser-atomic frontier are indicated in Figure 6.1.

- ***ENERGY AND ENVIRONMENT***
  - NUCLEAR ENERGY: ISOTOPE SEPARATION; MAGNETIC AND INERTIAL CONFINEMENT FUSION
  - FOSSIL ENERGY: CATALYSIS, COMBUSTION, SYNTHETIC FUELS
  - MATERIALS PROCESSING
  - METEOROLOGY AND POLLUTION TRANSPORT
  - SPACE ENVIRONMENT
- ***NATIONAL SECURITY***
  - COMMUNICATION, NAVIGATION, IDENTIFICATION AND RANGING
  - ISOTOPE SEPARATION
  - INERTIAL CONFINEMENT FUSION
  - DIRECTED ENERGY WEAPONS
  - CHEMICAL AND BIOLOGICAL DEFENSE
- ***BIOLOGY AND MEDICINE***
  - DETECTION AND DIAGNOSIS
  - BIOCHEMICAL KINETICS
  - THERAPY
  - SURGERY
- ***EDUCATION AND TRAINING***

Figure 6.1. Some national programs at the laser-atomic frontier.

Each of these programs is motivated by a problem whose solution is urgent to the nation. The research spans the spectrum from fundamental to applied science. For the most part, the work is funded by mission agencies. Sometimes the research is carried out by small groups, but large teams utilizing major national facilities often play the major role. In each of these program areas there have been important spinoffs to other fields of science and technology in the form of new knowledge, new instrumentation and new techniques.

Applications of lasers in these national programs often capitalize on their unique abilities to detect and measure small amounts of species that can range from simple atoms to complex

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macromolecules, to selectively promote chemical and physical transformations by laser-tuning to absorption features of the species of interest, or to project large amounts of energy in a highly directional fashion, sometimes over long distances and in short time periods. Success in these programs frequently requires advances in the creation of lasers as well as in the understanding of the interaction of laser light with materials. Each of these programs involves frontier research on fundamental processes. This paper will describe several examples of this research that were selected to emphasize the unique opportunities afforded to our national programs by lasers, including issues concerning laser development, the fundamental interactions between the laser light and materials, and the promises and prospects for future advances.

ENERGY PROGRAMS

We describe here a few of the many notable contributions to our energy programs from developments at the laser-atomic frontier.

Laser-based methods are now being developed to enrich uranium isotopes for nuclear reactors. In the past the U.S. has achieved isotope separation using gaseous diffusion. However, because of substantial projected reductions in capital and operating costs, the U.S. Department of Energy (DOE) has recently selected the Atomic Vapor Laser Isotope Separation (AVLIS) process developed at the Lawrence Livermore National Laboratory for further demonstration and potential deployment. The AVLIS process is based on the ability to tune a laser to the 235 isotope of uranium, allowing this species to be selectively ionized and electromagnetically collected. The technology is being developed in a full-scale demonstration facility at Livermore.

Lasers are being increasingly applied to measurement and process control in synthetic fuel process development. In the early days of development of coal gasification processes, the gasifiers were poorly instrumented — sometimes it was not even possible to know if the coal was flowing within the system. Scientists at the Los Alamos National Laboratory have installed several types of laser spectroscopic diagnostics in a gasifier at the Morgantown Energy Technology Center (METC) that provides real-time indications of species concentrations and conditions in an operational gasifier. One of the diagnostic approaches used at METC is coherent anti-stokes raman scattering (CARS). By making these measurements as a function of process conditions, it has become possible to characterize and optimize the process. Once this phase is completed, it is planned to apply these diagnostics process control.

Several national laboratories are collaborating with the U.S. steel industry to help improve the productivity and economic competitiveness of steel processes. A collaboration between the American Iron and Steel Institute and scientists at Los Alamos has as its principal goal the development of *in situ* diagnostics to measure the composition of molten steel. Among the techniques they are employing is laser induced breakdown spectroscopy (LIBS), which has been shown to have detection limits that compare favorably with those obtained using traditional methods.

DEFENSE PROGRAMS

One of the earliest recognized potential uses of lasers is to drive thermonuclear fusion by employing inertial confinement. Some of the new and promising research areas at the laser-atomic frontier that benefit from Inertial Confinement Fusion (ICF) research are outlined in Figure 6.2.

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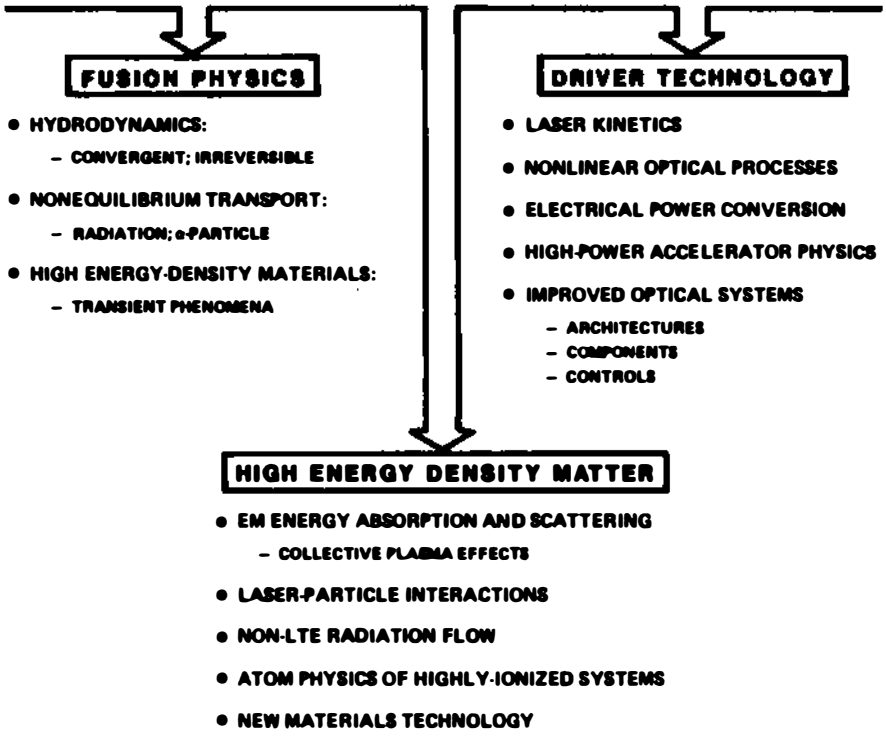

Figure 6.2. Avenues of research in inertial confinement fusion.

Current major ICF issues include:

- Extend current understanding to longer plasma scale-lengths
- Determine requirements imposed by drive symmetry
- Characterize behavior of hydrodynamic instabilities
- Achieve laser-driven compression to higher densities (200 g/cc).

There has been interest in the potential use of lasers as weapons. This interest has recently intensified owing to the role laser systems could play in the Strategic Defense Initiative (SDI). In a recent, full-scale test of the Navy's mid-infrared advance chemical laser against the second stages of a Titan-1 missile on the ground, the second stage of the missile exploded after it was irradiated by this 3.8 micron DF chemical laser for several seconds.

Lasers are among several types of defensive weapons that are being evaluated in the SDI research program. The strengths and weaknesses of lasers as potential weapons are outlined in Figure 6.3. One of the issues that limits the type of applications of lasers is that the Earth's atmosphere is a strong absorber of electromagnetic radiation at certain wavelengths. A "back of the envelope" calculation [1] shows that a power of 100 MW and a brightness of roughly 9×10^{20} W/sr will be required for effectiveness against ICBMs. Other laser development goals are compared with current performance in Figure 6.4 which is taken from ref. 1.

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Weapon	Type	Wavelength, micrometers	Excitation mode	Power	SDI application	Strong point	Weak point
Laser	Chemical HF Iodine	 2.7 1.3	Chemical reaction	Chemical	Ground-based or space-based	Speed of light; needs only small power source	Needs large optics; hard to manufacture; easy to damage
	Free-electron	Tunable from 0.1-20	Electron beam excites electrons	Electric	Ground-based	Speed of light; penetrates atmosphere	Laser destroys optical coatings
	Excimer KrF XeCl XeF	 0.25 0.31 0.35	Electron beam excites gas	Electric	Ground-based	Speed of light	Inefficient power user
	X-ray	—	Lasing rods	Nuclear bomb	Pop-up	Speed of light	Must be launched

Figure 6.3. Strengths and weakness of laser weapons.

	Goal	Present
Laser brightness	10^{21} watts/steradian	10^{17} W/sr
Particle energy:	250 megaelectronvolts	50 MeV
Space power:	100–300 megawatts	25 kW
Efficiency:	600 kilojoules per kilogram	NA
Cost:	$1 per optical joule	$50/J

Figure 6.4. Goals and present status of lasers for use as weapons.

Lasers also have potential application in chemical and biological defense, where they could provide sensitive, remote detection of these agents before they could become a threat to troops or the population. One approach involves several forms of Raman scattering. In experiments to date [2], simulants have been employed rather than actual chemical agents. The results of these experiments give promise for the creation of sensitive remote spectroscopic techniques for detection of chemical and biological agents.

BIOLOGY AND MEDICINE

Numerous advances in health and medicine have been made possible by successes at the laser-atomic frontier. A promising new development is the possibility for rapid detection of bacterial and viral disease. Present methods of identifying viruses and bacteria take from 2-3 days to several weeks. Many diseases are, however, life-threatening and a physician must make an immediate decision regarding whether to administer a broad spectrum drug or delay treatment. Within this scattergun treatment approach, broad spectrum antibiotics are expensive and sometimes ineffective; sometimes the treatment chosen may not be the appropriate one. Costs mount as patients spend more time in hospitals awaiting diagnosis. In addition, many diseases are highly communicable, especially among newborns, and delay in treatment may present grave risk to others.

The technique of multiparameter light scattering (MLS) holds promise for the rapid (minutes to hours), less expensive (factor of 10-20) identification of bacterial and viral disease. The

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scientific feasibility of bacterial and viral detection by MLS was established by Los Alamos scientists in experiments carried out under funding from the NIH. Development of prototype instrumentation for bacterial and viral detection is under way in a collaboration between Los Alamos and a company called Mesa Diagnostics in what represents the most substantial transfer to date of technology developed at a national laboratory to the private sector. The issues remaining to be addressed are outlined in Figure 6.5.

- ***DEMONSTRATION OF UNIQUE SIGNATURES FOR PURE VIRAL CULTURES AND BACTERIA***
  - EFFECT OF MEDIUM
  - TIME DEPENDENCE
  - COMPENSATION FOR BIREFRINGENCE OF DISPOSABLE CUVETTE
- ***DEVELOP PROTOCOLS FOR CLINICAL SAMPLE PREPARATION***
- ***AVAILABILITY OF SMALL, INTENSE LIGHT SOURCE***
  - 100 mW AT 420 nm, cw
  - DESIRABLE BUT NOT ESSENTIAL

Figure 6.5. Issues involved in the application of light scattering to the detection and diagnosis of bacterial and viral diseases.

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1. M.A. Fischetti, IEEE Spectrum, September, 1985, pp. 47-54.
2. A. Hartford, Jr., Los Alamos National Laboratory, private communication, September, 1985.

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7. LASER-ATOMIC RESEARCH IN UNIVERSITIES

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Laser-atomic research is pursued in each of the three major arenas for U.S. science: federal laboratories, industrial laboratories, and universities. The universities, however, have a special role: in addition to carrying forward a major component of the basic research, they bear the responsibility for training the next generation of professional scientists. This paper will survey the current situation and future prospects for laser-atomic research in the universities.

One cannot predict with certainty which topics in laser-atomic research will be most fruitful in the coming years but a look at past developments provides some perspective on what the future can hold. These developments include

clusters	ultraintense fields
laser cooling	optical frequency standards
state-to-state collisions	exotic atom spectroscopy
supersensitive detection	laser-generated plasmas
Rydberg atoms	cavity electrodynamics
molecular ion spectroscopy	four-wave mixing
optical bistability	radiative level shifts
squeezed states	relativistic beam spectroscopy
femtosecond spectroscopy	optical chaos
atom trapping	single particle spectroscopy
coherent XUV generation	inner shell spectroscopy
x-ray lasers	phase conjugation

The topics on this list have one feature in common: none of them was anticipated at the time of the last physics survey (the "Bromley report") in the early 1970's. Either they were unknown, or their potential significance could not have been appreciated. When one considers the abundance of these developments together with the advances described in this workshop, it is impossible not to be optimistic about the scientific discoveries and technological developments to be expected in the coming decades. If the U.S. is to share in these, however, the university groups must have the resources for cutting edge research and for training enthusiastic young scientists at the frontiers of knowledge.

The universities are facing increasing difficulties in maintaining the quality of their research programs. There is growing evidence that they may not be able to attract capable young scientists to their faculties when the number of positions increases sharply in the next decade. These concerns apply broadly to physics in the universities, but, as will be discussed below, they are particularly acute in laser-atomic physics. The following general summary, which draws on material provided by the American Institute of Physics Manpower Statistics Division, provides a background for understanding the particular concerns of the future of laser-atomic research in the universities.

The recent history of federal support for physics research in the U.S. is dominated by two crises: the Sputnik crisis in 1958, and the Vietnam crisis around 1970. U.S. science underwent

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massive buildup following the U.S.S.R.'s launching of Sputnik in 1958. Between 1960 and 1970 the number of Ph.D.'s granted annually in the U.S. soared from 500 to over 1500 (see Figure 7.1),

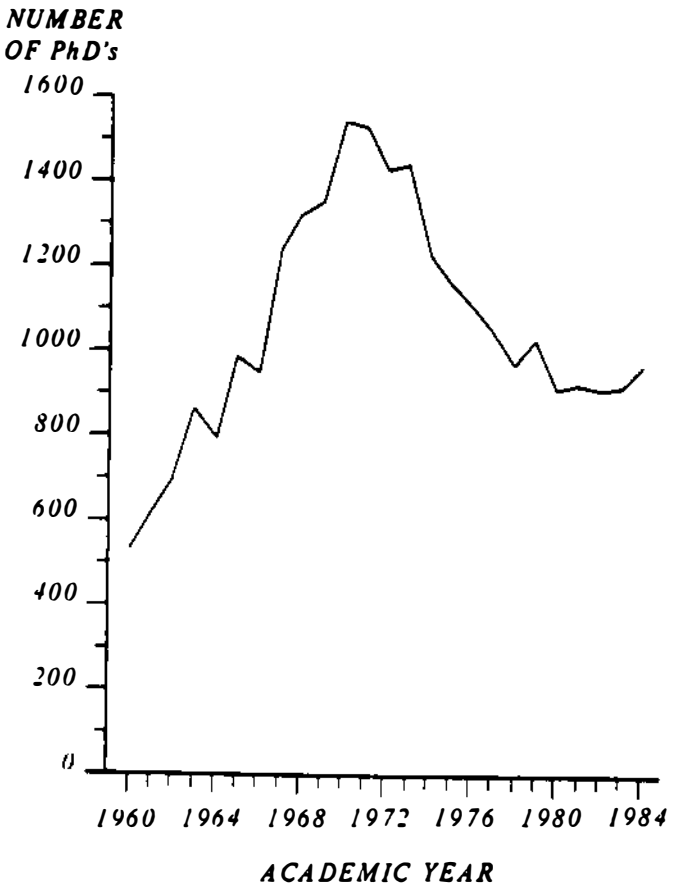

Figure 7.1. Annual production of physics Ph.D.'s in the U.S. (Source: American Institute of Physics Manpower Statistics Division.)

and the physics faculties doubled in size. Around 1970, however, the national priorities abruptly changed due to the pressures of the Vietnam War. Support for science switched from an expanding to a contracting mode. Ph.D. production fell to about 950 per year, where it has remained roughly constant, and faculty hiring diminished to a trickle.

Since the early 1970's, physics faculties in the U.S. have been aging due to the lack of junior appointments. The median age has increased steadily; today it is almost fifty. As Figure 7.2 shows, the median age is expected to drop precipitously in the next decade when the faculty members appointed in the 1960's start to retire and young scientists are appointed to replace them.

For the next few years there is expected to be a rough overall balance between supply and demand for Ph.D. physicists, though only if industrial needs do not increase substantially and the nation does not embark on any large physics-intensive federal programs. (The demand in optics is actually much greater than the current Ph.D. production rate of about fifty per year, but this is met

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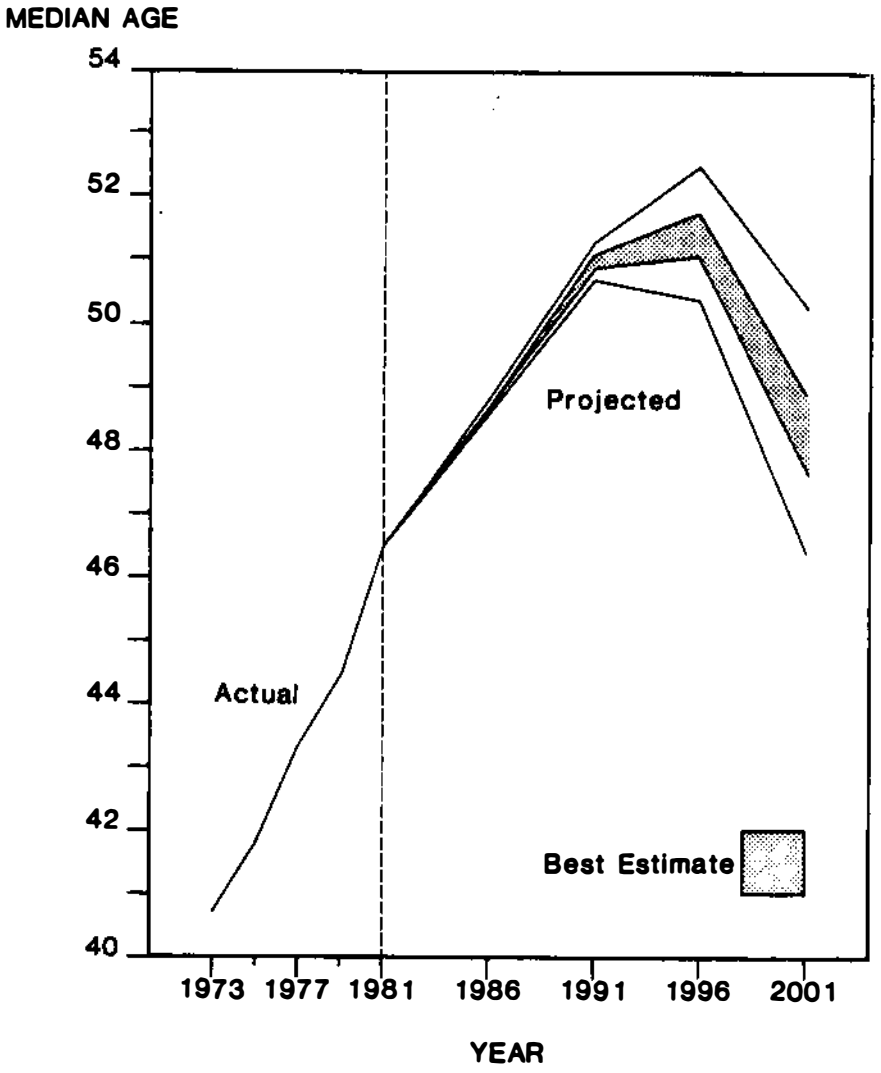

Figure 7.2. Median age of university physics professoriat: actual and projected, 1973-2001. (Source: American Institute of Physics Manpower Statistics Division, AIP and NRC unpublished data.)

by an influx from other branches of physics. Optics is now the fourth largest area for young Ph.D. physicists.) As Figure 7.3 indicates, however, the demand is expected to exceed the supply in the next decade. The increased demand is due primarily to the need for young faculty at universities and colleges.

A qualitative change has occurred in the structure of the physics graduate student population. The fraction of foreign students has steadily increased. Today it stands at 40%; by the end of the 1990's it may exceed 50%. In the past, approximately half of the graduate students from abroad have made careers in the U.S. Any change in this career pattern would seriously affect the U.S.

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supply-demand balance. Furthermore, slightly more than 50% of the physics postdoctoral researchers in the U.S. are foreign. Because postdoctoral research is a normal step in preparing for an academic career, one can anticipate that our universities will draw increasingly on foreign scientists to fill faculty positions. It is evident that the U.S. has become highly dependent on foreign talent to meet its scientific needs.

Laser-atomic physics reflects these demographic changes. The number of Ph.D.'s produced annually in the U.S. in atomic, molecular and optical physics (AMO physics) reached a peak of 182 in 1970; today it stands at about 130 per year. (This discussion applies to AMO physics rather than to laser-atomic research alone, since the latter is not categorized separately in the manpower and funding studies.) As a fraction of all physics Ph.D.'s, AMO physics climbed from 13% in the period 1960-65 to 18% in the period 1980-84.

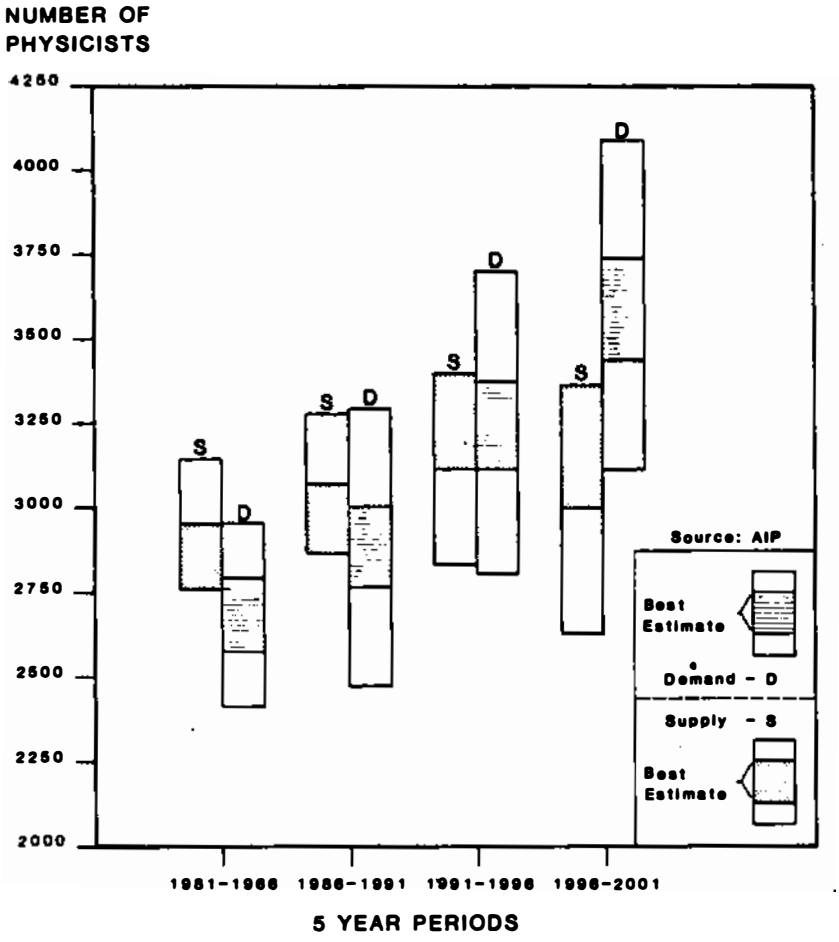

Figure 7.3. Projected supply and demand of physicists, 1981-2001. (Source: American Institute of Physics Manpower Statistics Division.)

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The funding history for AMO physics since 1973 is shown in Figure 7.4.

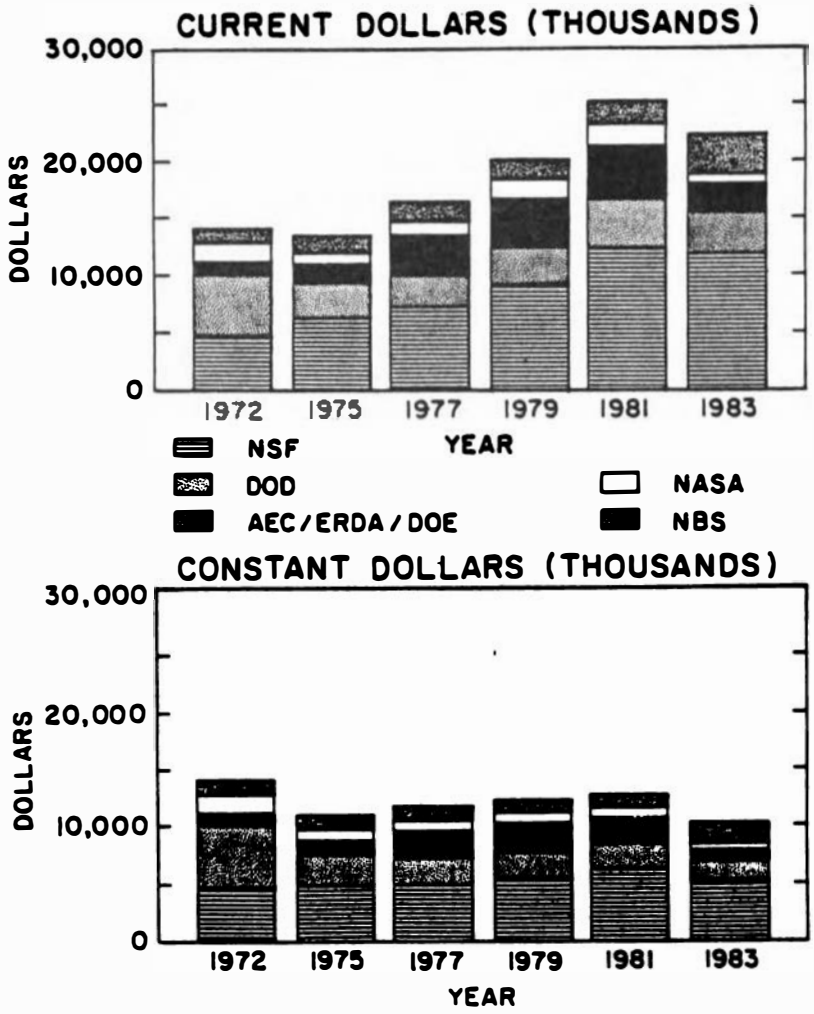

Figure 7.4. Funding of basic atomic and molecular physics by the federal government in universities and allied institutes, 1972-1983. (Source: the Interagency Atomic and Molecular Administrator's Group of the NSF Physics Division. These figures include federal funding of atomic and molecular physics at universities, university-related private research institutes, and a small fraction of the basic atomic and molecular science at federally funded research and development centers. A portion of optical physics is included.)

There has been a significant loss of support in constant dollars. The real loss is even larger than shown due to the increasing costs of equipment. In any case, the current grants are generally too small for effective research. In 1984 the average AMO research grant was only $85,000/year, whereas the cost for running an active university group is estimated to be typically $200K-$400K/year.

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The effects of the erosion of support for AMO physics are widespread. Most conspicuous is the critical shortage of lasers and other instrumentation for laser-atomic research. Today's lasers far exceed the lasers of the early 1970's with respect to power, spectral range, stability and tunability. A state-of-the-art tunable dye laser system can cost $150,000; many experiments need more than one of these devices. Most university groups in the U.S. lack up-to-date lasers. In contrast, in the mid-1970's the Deutsche Forschungsgemeninschaft initiated a program to equip West German universities with modern lasers. There have been periods when U.S. manufacturers of scientific lasers shipped more than half of their production to Germany. Laser-atomic research is now flourishing in West German universities, whereas U.S. university groups are finding it more and more difficult to carry forward competitive research.

Other consequences of the erosion of support for laser-atomic research in universities include a shortage of support for graduate students and postdoctoral researchers and widespread deterioration of shops and special services. Startup funds for young scientists are generally lacking. Most seriously, the research budgets are too small to allow flexibility in pursuing new scientific opportunities.

Although similar problems have occurred in other areas of university-based physics, AMO research was particularly hard hit by the loss of DoD support. Prior to 1970, the majority of the research was supported by the DoD. The field was effectively disenfranchised when the DoD withdrew. These problems are documented in the physics survey, *Physics through the 1990s* (National Academy Press, Washington, DC, 1986). It should be a matter of national concern that a growing shortage exists for qualified applicants for university positions in AMO physics. The shortage is due in part to industrial competition. Laser-atomic research is actively pursued in industrial laboratories that pay substantially higher wages than universities — approximately half again as much. Although salary is an important consideration in a career choice, university positions have traditionally been coveted by many of our most able young scientists. This situation is changing rapidly.

In universities across the nation, unfilled faculty positions exist in laser-atomic physics. Since we are already experiencing a shortage of faculty candidates in this area, when the demand for faculty candidates increases in the next decade one can anticipate a widespread shortage. The nation faces the prospect of being incapable of training new laser-atomic scientists.

The visible loss of attractiveness of university careers for able young scientists is the legacy of a decade and a half of undersupport for university-based research in the U.S. Today, young scientists contemplating university positions face the prospect of careers spent struggling to carry forward research with inadequate resources. One can hardly blame them for turning away from academia.

If the U.S. is to remain competitive in laser-atomic research, the university groups must have adequate support. The additional funds required, based on estimates in the forthcoming physics survey, amount to an increment of approximately $10 million a year for four years. Considering the scientific opportunities, and the broad range of federal and industrial programs that need the research, the investment is modest.

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