5.0 Optics and Quantum Electronics

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5.1 The Nonlinear Waveguide Interferometer

National Science Foundation (Grant EET 87-00474)
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James G. Fujimoto, Hermann A. Haus, Michael J. LaGasse, Shigeru Oho, John Moores, Masataka Shirasaki, Dilys L. Wong

All-optical switching can be accomplished with femtosecond pulses. Devices that utilize optical control signals may be classified as resistive or reactive. Resistive modulators utilize the change of absorption, reactive the change of index as a function

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of an optical control signal. The reactive modulators have the advantage that their insertion loss can be made small in principle, whereas the absorption of resistive modulators cannot be fully eliminated in the "on state." The phase modulation produced by index changes can be transformed interferometrically into amplitude modulation.

Our previous work with nonlinear optical waveguide interferometer switches using the bulk nonlinearity of GaAs[1] has revealed that, not only the required peak intensities tend to damage the waveguides, but that the thermal changes of the waveguide prevent high throughput rates.[2] For this reason, we investigated optical fiber interferometers as all-optical switches. The single fiber interferometer proposed by M. Shirasaki[3] uses two polarizations for the two "arms" of the interferometer. The pulse to be switched is separated into two mutually orthogonally polarized and time delayed pulses. If one of the two pulses is phase shifted by a cotraveling pulse, polarization rotation ensues and switching action can be accomplished. The advantage of this scheme is that long-term (nanosecond) changes of the fiber index do not affect the interferometer operation. It is also immune to heating effects. Preliminary experiments were reported at CLEO.[4] In more recent experiments,[5] switching of subpicosecond pulses was demonstrated in a 3 m length fiber interferometer. Also, in a 400m interferometer using 100 ps pulses from a modelocked Nd:YAG laser, switching was accomplished with 2 W peak power. The interferometer had excellent stability.

5.2 Picosecond Optical Signal Sampling

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Hermann A. Haus, Lynne Molter-Orr, Weiping Huang

The work on waveguide couplers and interferometers as basic components for optical switches concentrated on the theory of coupled waveguides and their dispersion characteristics, on the fabrication of such waveguides in GaAs, and on the experimental verification of the theoretical predictions.

In work reported in the last progress report,[6] Dagli described an equivalent circuit method for the analysis of rib waveguides, based on a mode matching technique. The method uses both guided and radiation modes and is very computation intensive. Also, it was limited to a scalar analysis and did not yield modal birefringence. We have developed a variational method for the evaluation of the dispersion characteristic. The program is "user friendly" and is implemented on a PC. Its accuracy as compared with Dagli's results and other published work is excellent. Birefringence effects are included as corrections to the results of the scalar analysis. The program was made available to the Celanese Research Division in Summit, New Jersey.

The fabrication of two- and three-guide couplers in GaAs carried out by L. Molter-Orr with rib waveguides, in cooperation with Dr. J. Donnelly of the MIT Lincoln Laboratory, tested the idea of the waveguide lens described previously.[7] The mode confinement in rib waveguides is weaker than in channel waveguides and thus enhances effects associated with the non-orthogonality of modes pointed out by Hardy and Streifer.[8] In our own work[9] we had shown that the crosstalk produced by the effect in a two-guide coupler switch could be essentially eliminated by proper detuning and length adjustment of the coupler. This prediction was confirmed with the rib-waveguide couplers tested by L. Molter-Orr.[10]

In the coming year, the switching techniques developed in connection with the single fiber (two polarizations) fiber interferometer described in the preceding section will be applied to optical waveguides, both as a means of measuring the nonlinear optical parameters and as a means of all-optical switching.

5.3 Solitons

Joint Services Electronics Program (Contract DAAL03-86-K-0002)
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Hermann A. Haus, Erich P. Ippen, Janice M. Huxley, Y. Lai, Ling-Yi Liu, John Moores

The soliton laser first realized by Mollenauer is the only means to date for the production of femtosecond pulses in the wavelength regime above 1.5 microns.[11] Direct saturable absorber modelocking of the *F*-center laser, or the metal ion laser, the only sources of high power within this frequency regime, is not possible because of the long relaxation times of these laser media. In another section of this report, the operation of such a laser is described. As part of the general theory of solitons, the operation of the two-cavity soliton laser is the subject of investigation. The aim is to determine the limit on pulse energy and pulse width imposed by the system parameters. A paper detailing the theory of the two-cavity soliton laser is currently in preparation.

Solitons are also candidates for high rate all-optical switching. The fact that a soliton can propagate undistorted along a dispersive fiber, the nonlinearity of the fiber balancing the spreading of the pulse by group velocity dispersion, suggests that the all-optical nonlinear interferometer switch described above could use solitons for the switching operation. We have started a theoretical investigation, both analytic and numerical, of the interaction of two orthogonally polarized "solitons." This entails the solution of two coupled nonlinear Schroedinger equations. It turns out that this problem is not "integrable" in general. For the values of the parameters describing an isotropic fiber, "solitons" do not fully recover after a collision; the pulses are solitary waves and not solitons. The perturbation from "soliton" behavior is small, if the collision is weak (the pulses pass each other rapidly), and the mutually induced phase shift is much smaller than π. Unfortunately, this is not adequate for switching operation of the interferometer. In future work we intend to pinpoint operating conditions for which switching may be feasible.

In work sponsored by Draper Laboratory, we initiated experimental and theoretical studies of soliton formation in an anomalously dispersive fiber using a Raman pump pulse at 1.3 microns from a modelocked Nd:YAG laser. The ultimate goal is to explore the use of solitons in a laser gyro using counterpropagating solitons. Since quantum noise is the ultimate source of noise in such a gyro, it is necessary to investigate the soliton quantization problem. This study has just begun.

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5.4 Strained Layer Superlattices on <111> Oriented Substrates for Optical Devices

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Joint Services Electronics Program (Contract DAAL03-86-K-0002)

Kimberley Elcess, Stuart D. Brorson, Richard Singer, Clifton G. Fonstad, Jr., and Hermann A. Haus, in collaboration with Chrishen Mailhoit and Daryl Smith

When sufficiently thin layers of semiconductors possessing different lattice constants are grown on top of each other, the lattice mismatch can be accommodated by straining the individual layers. That such "strained layer super-lattices" can be grown without defects was first pointed out by Osbourn.[1] Strained layer superlattices are a topic of considerable current interest since they promise increased flexibility in fabricating superlattices tailored to particular purposes.

Recent theoretical work predicts that the strain in In_xGa_{1-x} As/GaAs superlattices fabricated on <111> GaAs induces a polarization in each layer via the piezoelectric effect.[2] The built-in polarization fields are oriented along the growth axis and alternate in sign between the $In_xGa_{1-x}As$ well and the GaAs barrier. This affects the electronic properties of the bound quantum well states, and hence the optical properties of the structure.[3] In particular, superlattices possessing such built-in fields display a linear electrooptic effect, instead of the weaker quadratic effect present in conventional (non-polarized) quantum wells. This renders these structures ideal for optical device applications.

Molecular beam epitaxial growth on <111> substrates has proven to be different in significant ways from <100> growth, specifically in terms of the oxide desorption and growth initiation procedures which must be followed, the growth temperatures, and the sensitivity of the growth process to the arsenic flux.[4] The critical temperatures, fluxes, and flux ratios have now been determined and high quality layers can now be grown reproducibly.

We used the growth procedures we have developed to grow what are, to our knowledge, the first <111> oriented strained layer superlattices with built-in polarization fields. We have constructed a system for measuring near-infrared linear absorption in the quantum well samples, and are focusing on identifying features in the absorption data. Future work will center around measuring the dependence of the absorption edge on an externally applied DC electric field. From this one will be able to verify the existence of a built-in polarization field and deduce its actual magnitude. Further research will include fabricating slab waveguides which exploit the linear electro-optic effect to make optical phase modulators.

Finally, we are collaborting with Dr. D. Smith and colleagues at Los Alamos and with Prof. B.A. Weinstein and colleagues at the State University of New York at Buffalo on various characterization studies of these superlattices including Rutherford backscattering and ion channeling, low temperature optical absorption and photoluminescence, and transport.

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5.5 Diffraction Coupled Diode Laser Arrays

Joint Services Electronics Program (Contract DAAL03-86-K-0002)

Elias Towe, Robert E. Hillman, Clifton G. Fonstad, Jr.

We have continued development of our novel phase-locked laser diode array, the mixed mode phase locked (M^2PL) laser array. This array achieves phase-locking and operation in the fundamental, single-lobed super-mode by diffraction in a region introduced into the middle of the array. On either side of the mode-mixing region the modes are guided in ridge waveguides which terminate at the laser facets, but within the mode-mixing region the radiation is not confined laterally. Thus a small fraction of the radiation in each element of the array is coupled in this region to the adjacent elements. If the length of the mode-mixing region is properly designed, coupling will occur in phase, and the radiation in the adjacent guides will be locked in phase and single-lobed emission will result.

The M^2PL laser concept was very successfully demonstrated for the first time last year.[1] The structure has been shown to routinely produce stable, single-lobed far-field patterns, and in this respect appears to be unique amongst the multitude of array structures which have been proposed. Moreover, it is one of the simplest to produce.

Work in the past year has focused largely on theoretical analysis of the M^2PL structure. More detailed modeling of the optimum mode-mixing region length has been performed. Calculation of the strength of the coupling across the mode-mixing region for various device geometries, and estimates of the loss introduced by the use of the mode-mixing region, have also been completed. These predictions have been confirmed experimentally by measurements made viewing the M^2PL lasers as passive waveguide structures.[2]

Very recently, we have begun a collaboration with an industrial laboratory interested in applying the M^2PL concept to high power arrays. A mask set having a variety of mode mixing region lengths corresponding to phase shifts between adjacent guides ranging from under 2π to over 3π has been designed. Thus, in addition to independently confirming the importance of the M^2PL concept, we expect these studies to provide verification for the theoretical analyses already completed, and data for use in more refined and extensive modeling, and optimization.

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5.6 Multiple Quantum Well Semiconductor Waveguide Optical Devices

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Kristen Anderson, Richard Singer, Mary Phillips, James Vlcek, Clifton G. Fonstad, Jr., Hermann A. Haus

Multiple quantum well semiconductor optical modulators and switches in integrated waveguide structures have important application in all-optical processing systems. Research in this area is focused on theoretical optimal device design and novel material processing techniques.

In semiconductor materials, thermal effects due to nonradiative carrier recombination have a significant effect on optical device performance,[1] and are a limiting factor in many device designs. The effects are particularly crucial when a multiple quantum well material is excited with photons of energy corresponding to the exciton absorption peak where the optical nonlinearity is large. One way to benefit from the large optical nonlinearity, and reduce thermal effects, is to use short device active lengths, restricting the size of the quantum well region within the waveguide.

Device performance and thermal effects in both loss modulation devices, and nonlinear Mach-Zender interferometers were examined theoretically. The results indicated the feasibility of devices with small active regions. Loss modulation devices with active regions 5 μm long are expected to exhibit contrast ratios of as much as 6 dB using conventional diode laser light sources. Interferometric configurations with insertion losses of $<$ 2.5 dB are predicted in which carrier induced heating effects are negligible.

A new material processing technique, disordering of GaAs-GaAlAs multiple quantum well materials via thermal annealing, is currently under investigation as a possible method to achieve small active lengths. Quantum wells under a silicon oxide capping layer are unstable and will disorder when annealed at temperatures above 825°C. This causes a shift to higher energy of the material band edge. With a silicon nitride capping layer, however, quantum wells subject to the same annealing conditions remain stable. In a multiple quantum well sample grown by molecular beam epitaxy, using the different dielectric capping layers, we have successfully demonstrated patterned disordered regions. These results were verified by transmission measurements and scanning electron microscopy. Buried waveguide structures are also currently being fabricated using this technique.

The ultimate goal is to use this disordering technique to compositionally mix, or homogenize, the GaAs-GaAlAs MQW's everywhere we wish to eliminate them. Subsequently shallow rib waveguides will be defined to produce the desired integrated optical circuitry.

Optical absorption measurements were performed on the disordered material. The measured band edge corresponds to that of an $Al_xGa_{1-x}As$ alloy with a fractional aluminum content, x, which would be expected if the original multiple quantum well layer were completely disordered. Future studies of linear and nonlinear absorption and refractive index as well as carrier relaxation phenomena will provide further comparison of the disordered material to an epitaxially grown alloy. These studies well lead to specific device designs and eventual fabrication of nonlinear modulators and switches in integrated optical waveguide geometries.

In addition to working with the well established GaAs-GaAlAs system, we are also studying the InGaAs-InAlAs system lattice-matched to InP. This system is compatible with operation in the 1.3-1.5 μm wavelength region commonly used in single-mode optical fiber telecommunications systems. Furthermore, the energy band discontinuities in this system are larger than they are in the GaAs-GaAlAs system, and larger non-linear optical effects are anticipated. The work in the InGaAs-InAlAs system is less advanced than that on the GaAs-GaAlAs system, and efforts are being directed first at simply producing and characterizing multiple quantum well structures, prior to addressing the issue of incorporating them into guided wave optical devices.

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5.7 Femtosecond Laser Systems and Pulse Generation

Joint Services Electronics Program (DAAL03-86-K-0002)
National Science Foundation (ECS 85-52701)

James G. Fujimoto, Erich P. Ippen, Robert W. Schoenlein, Michael J. Lagasse

One of the key issues in the study of ultrafast phenomena is the availability of laser sources with femtosecond pulse durations at suitable wavelengths. Our research on femtosecond laser generation techniques has centered on the development of high repetition rate femtosecond amplification techniques as well as the development of tunable femtosecond laser sources. These sources provide the basis for a wide range of experimental investigations of ultrafast phenomena.

We have recently completed the construction of a high repetition rate femtosecond laser amplifier which can generate high intensity pulses with 50 fs duration at 620 nm. The laser source is a colliding pulse modelocked ring dye laser which uses internal prisms for the compensation of dispersion.[1] This laser is amplified with a copper vapor laser pumped dye jet amplifier operating at an 8 kHz repetition rate.[2] Using a six pass configuration, a gain of 10^4 is obtained to generate pulse energies of a few microjoules. The high repetition rate of the laser source is a key factor in improving time resolved measurements since it permits the use of signal averaging and lock-in detection techniques to enhance sensitivity. In addition, the high peak intensities allow the direct observation of nonlinear optical processes.

The high peak intensities produced by this laser are also important since they permit the application of nonlinear optical techniques for generating other wavelengths or performing optical pulse compression. By focusing intense pulses into a medium with an intensity dependent index of refraction, nonlinear self phase modulation can be used to generate additional frequencies. A broadband femtosecond continuum can be produced which contains wavelengths ranging from 400 nm to 900 nm. This source is especially useful as a broadband probe of transient absorption or reflectivity line shape. We have applied our high repetition rate amplifier to perform pump and continuum probe measurements of a variety of femtosecond processes including non-equilibrium electron heating in metals, carrier relaxation in GaAs and AlGaAs, and transient excited state dynamics in polydiacetylene.

One of our principal objectives in continuing our development of high repetition rate sources is to extend the temporal resolution and bandwidth of femtosecond continuum pump and probe measurements. Previously, temporal resolutions for broadband continuum probe measurments were limited to ~ 100 fs. Because of the extremely large bandwidths and short pulse durations involved, one of the key problems in femtosecond measurement is controlling dispersion produced by the frequency generation processes and propagation through the optical components which are present in the experimental apparatus. Group velocity dispersion causes different frequency pulses to propagate with a varying time of flight through an optical system and thereby produce pulse broadening. The use of diffraction grating pairs or prism pairs has been demonstrated for the compensation of linear group velocity dispersion. However, for our pulse durations and experimental bandwidths, higher order dispersion was also found to play a critical role in limiting experimental resolution. We have been investigating dispersion compensation techniques in an effort to perform broadband measurements over several tens of nanometers with resolutions on the 10-20 fs time scale.

Complementing our high repetition rate femtosecond amplifier we have recently completed the construction of a tunable femtosecond laser. The approach that we are using is similar to that developed by Kafka and Baer,[3] and combines pulse compression techniques with synchronous modelocking. The laser system is based on a conventional modelocked Nd:YAG laser which produces 90 ps pulse durations at 1.06 μm Pulses from this laser are compressed using an optical fiber and diffraction grating pair to 5 ps. After frequency doubling, this laser is used to pump a synchronously modelocked, cavity dumped dye laser which generates 500 fs pulse durations. Finally, a second optical fiber and prism pair pulse compressor is used to obtain pulse durations as short as 55 fs.

Since this laser system uses pulse compression techniques and synchronous modelocking, the wavelength of the output pulse can be easily tuned. Using different laser dyes, we have generated femtosecond pulses in the wavelength range from 580 nm to 950 nm with typical pulse durations of 75 fs. The tunability and short pulse duration of this laser make it especially promising for femtosecond studies of GaAs and AlGaAs materials and devices. The laser can be tuned both above and below the bandgap of these semiconductors, thereby permitting the investigation of excited carrier relaxation processes as well as dynamic processes in guided wave devices.

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5.8 Femtosecond Carrier Dynamics in GaAs

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Erich P. Ippen, James G. Fujimoto, Wei-Zhu Lin, Beat Zysset, Robert W. Schoenlein, Michael J. Lagasse

The investigation of transient carrier dynamics in GaAs and AlGaAs semiconductors is relevant to electronic and optoelectronic devices which depend on high speed transient carrier phenomena. Our research program focuses on the study of carrier dynamics using optical techniques which can provide measurements on the time scale of the fundamental scattering processes. Using pump and probe absorption saturation spectroscopy, we have investigated carrier scattering, energy relaxation, and intervalley scattering on a femtosecond time scale.

Pump probe absorption saturation measurements have been performed to measure the initial scattering time of carriers as they relax from their initial optically excited states. Using pulse durations of 35 fs generated by a CPM laser at a fixed wavelength of 620 nm, we have previously measured[1] initial scattering times as rapid as 13-30 fs. Because the laser source for these measurements was not tunable, studies were performed by varying material composition[2] of AlGaAs.

In order to investigate the energy relaxation of excited carriers, femtosecond pump and continuum probe measurements were performed using our recently developed high repetition rate amplifier system. By generating optically excited carriers at the pump pulse wavelength and probing absorption saturation over a range of wavelengths using a broadband continuum, the energy relaxation dynamics of the excited carriers could be observed. Our investigations provide evidence for transient nonthermal carrier distributions which occur on a femtosecond time scale.[3,4] Probing absorption saturation using transitions from the split-off band permits an investigation of the electron distribution as distinct from the hole distribution. This suggests possible methods for studying carrier-carrier scattering and separating electron-hole scattering contributions. Systematic measurements performed using different material compositions of AlGaAs permit an exploration of the effects of generating carriers with different excess energy. Intervalley scattering was found to contribute significantly to the initial energy relaxation of carriers which are excited above the X and L valleys.

Our recently developed tunable femtosecond laser system provides a unique opportunity to perform complementary investigations of transient carrier dynamics. One process of special importance to electronic device operation is intervalley scattering. Scattering of energetic carriers from the Γ valley to the L and X satellite valleys is one of the dominant mechanisms which limit carrier transport and give rise to negative differential resistivity effects.[5]

Using our tunable femtosecond laser, we have begun an investigation of intervalley scattering and energy relaxation in GaAs and AlGaAs. A tunable femtosecond source is a particularly important tool for investigating carrier dynamics since the excess energy of the excited carriers can be continuously varied by adjusting the laser wavelength. Preliminary measurements[6] show dramatic changes in carrier scattering times when the excess energy of the carriers are increased to permit $\Gamma - L$ and $\Gamma - X$ intervalley scattering. While intervalley scattering has recently been investigated using a number of different techniques, including transient conductivity and luminescence measurements, femtosecond pump probe probe measurements represent one of the few approaches which has sufficient temporal resolution to directly measure the initial carrier scattering times.

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5.9 Femtosecond Spectroscopy of Electronic and Optoelectronic Materials

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US Air Force - Office of Scientic Research (Contract AFOSR-85-0213)

James G. Fujimoto, Erich P. Ippen, Wei-Zhu Lin, Paolo Mataloni, Robert W. Schoenlein, Janice M. Huxley, Stuart D. Brorson

Femtosecond optical studies of electronic and optoelectronic materials are of importance since they provide an approach for directly investigating the transient dynamics associated with these materials. These studies are relevant to both increasing the understanding of fundamental physical processes as well as to applications of these materials in high speed electronic and optical signal processing.

In addition to studies in semiconductors, another topic of special interest is transient electronic processes in metals. Metals and semiconductors are the principal constituents of devices. In contrast to semiconductors, relatively few investigations have been performed in metals. Because of the high densities inherent in metals, nonequilibrium processes occur on an extremely rapid time scale and changes in optical properties are usually quite small. Thus, transient spectroscopy in metals has been made possible only through the recent development of new femtosecond laser technology which provides high temporal resolution and sensitivity.

Using femtosecond laser excitation, it is possible to generate and measure nonequilibrium electron heating phenomena. If the incident laser pulse duration is less than the electron-phonon energy transfer time, a transient nonequilibrium between the electrons and lattice may be produced. Since the heat capacity of the electron gas is much less than that of the lattice, electron temperatures in excess of the lattice temperature can be generated. We have investigated nonequilibrium electron energy relaxation and temperature dynamics in metals. Using noble metals, changes in electron temperature may be investigated optically by monitoring optical transitions from the *d* bands to the Fermi level. By performing pump and continuum probe transient reflectivity lineshape measurements on a femtosecond time scale, nonequilibrium electron heating was demonstrated.[1] Electron energy relaxation to the lattice was observed on a 1-2 ps time scale.

Since measurements can be performed on a time scale less than the electron lattice energy transfer time, it is possible to directly measure nonequilibrium properties of the electron gas. We have performed a study of transient energy transport in thin metal films using pump probe time-of-flight measurements.[2] A femtosecond pulse is used to heat the electron distribution on one side of a thin film while changes in electron temperature on the opposite side of the film are monitored with a probe pulse. Subpicosecond energy transport times are observed in 500 Å - 3000 Å thick gold films. This corresponds to an effective transport velocity of $\sim 10^8$ cm/sec, almost two orders of magnitude faster than equilibrium lattice heat diffusion. This experiment is the first observation of nonequilibrium energy transport in metals.

As an extension of these studies, we are currently working in collaboration with researchers at the General Motors Research Laboratories to investigate the transient dynamics of image potential states in metals.[3] When an electron is photoexcited from a bulk metal, it can relax into a state where it is bound to the surface in an image potential. These states are not surface states associated with the bulk. The electron wavefunction is maximized outside rather than inside the metal. We are currently developing techniques for transient photoemission spectroscopy in an effort to measure the transient dynamics of the image potential state. Femotsecond spectroscopy can be performed by populating the optical states using a short laser pulse, then subsequently photoemitting from these states using a second pulse. The photoemission energy spectrum may be obtained with a conventional energy analyzer to determine the transient photoemission spectrum from the metal. These experiments are relevant for the measurement of fundamental physical processes and also represent a step toward combining femtosecond optical techniques with surface science diagnostics.

The investigation of nonequilibrium electronic effects in metals can provide new information on the fundamental physical processes in condensed matter. An increased understanding of these processes is important for problems such as the generation of plasmas and x-ray and UV laser sources which involve the interaction of high intensity laser pulses with metals. In addition, nonequilibrium electronic processes and transport are relevant for future high speed semiconductor devices where nonequilibrium processes can occur at semiconductor-metal interfaces.

In addition to semiconductors and metals, we are also investigating transient processes in polydiacetylenes. Polydiacetylenes are a class of organic polymers which are of technological importance because they exhibit large third order optical nonlinearities.[4] Thus they are an attractive candidate for all optical signal processing applications. The enhanced nonlinear optical response of these materials is the result of their unique chemical structure which is characterized by long carbon chains with bond superalternation and delocalized electronic states. Polydiacetylenes are also a model system for investigating one dimensional electron behavior.

We have investigated the transient excited state dynamics of polydiacetylene Langmuir-Blodgett films and crystals of PTS using pump probe absorption saturation spectroscopy.[5] Measurements performed at 620 nm just above the absorption edge indicate a recovery response consisting of a picosecond time scale process combined with an initial ultrafast relaxation occurring on a 100 fs time scale. Further studies of excited state dynamics were performed using continuum probe spectroscopy. The initial absorption saturation recovery was correlated with the onset of an induced absorption at a lower energy which is produced by relaxation of the excited state. Transient absorption saturation hole burning was observed near the excitation energy.

In addition to studies of absorption saturation, we have also investigated effects in thin PTS crystals. This system presents a natural Fabry-Perot cavity so that measurements of transmission versus wavelength provides information on both absorption and index. Pump and continuum probe measurements which investigate the spectrum at discrete time intervals after the occurrence of a pump pulse provide can permit the measurements of both the nonlinear absorption saturation corresponding to energy relaxation of the electronic excitations as well as the nonlinear index changes associated with excited state relaxation. At present there are a number of competing theories which explain the electronic nature of the excited state and the origin of the enhanced nonlinear effects observed in the polydiacetyelene system. Further studies of excited state transient dynamics could provide important information which could distinguish between different theoretical pictures and also describe the rate limiting processes in potential signal processing applicatons.

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5.10 Ultrashort Pulse Laser Medicine

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Working in collaboration with researchers at the Massachusetts Eye and Ear Infirmary and the Wellman Laboratories of the Massachusetts General Hospital, we are investigating the application of short and ultrashort pulses lasers and time resolved spectroscopic techniques to problems in laser medicine. In contrast to more established applications of cw lasers to surgery which relay primarily on thermal effects to acheive the desired therapeutic response, the use of high intensity ultrashort pulsed lasers is promising since it permits the possibility of nonlinear laser tissue interactions. In addition, the application of measurement techniques traditionally used in time resolved spectroscopy to studies of transient processes in laser tissue interaction and medical diagnostics can yield new and significant information which will allow the optimization of desired laser tissue interaction processes.

One area of special interest is the application of high intensity short pulsed lasers for ophthalmic surgery. In particular, the *Q*-switched and mode-locked Nd:YAG lasers have recently emerged as important tools for ophthalmic surgery.[1,2] High intensity laser pulses in the nanosecond or picosecond regime can be used to produce optical breakdown which is used for surgical incision of transparent structures in the anterior eye. We have developed and applied techniques of time resolved spectroscopy to investigate the transient processes of plasma absorption, acoustic wave generation, and cavitation which are associated with laser induced breakdown.[3] Our ultimate objective is to characterize physical processes, determine correlations with physical effects in ophthalmic surgery, and finally to optimize the laser surgical process.

Our initial investigations were performed using a nanosecond laser, since this is the most widely used system clinically. During the past year we have extended our investigations using pulse durations of 30 ps generated by a modelocked Nd:YAG laser.[4] The rationale for performing measurements with shorter pulses is twofold. By developing and applying time resolved techniques such as pump probe studies, we can perform measurements of physical processes with a temporal resolution limited only by the pulse duration. In addition, since peak intensities scale up as laser pulse duration is increased, the desired nonlinear laser-tissue interactions can be generated at lower energy densities. We have performed pump and probe optical measurements to characterize the spatial and temporal evolution of the plasma, acoustic wave, and cavitation in optical breakdown. Comparison of nanosecond and picosecond lasers show that the transverse dimensions of the physical processes can be reduced by using shorter pulse laser sources, thereby producing an increased localization of surgical effects.

In order to establish a correlation of the physical phenomena in laser induced breakdown to a clinically observable effect in a biological system, the cornea endothelium was used as a model system. Studies were performed with enucleated bovine corneas. The corneal endothelium is a monolayer of cells on the posterior surface of the cornea which can provide a sensitive indicator of laser surgical effects.[5] Laser induced breakdown was generated both directly on the endothelium and at varying distances from the endothelium immersed in saline solution. Ultrastructural changes, including removal of cells and production of an incision were characterized using light and scanning electron microscopy. The localization of surgical lesions was observed to vary roughly as $(\text{energy})^{1/3}$ for both nanosecond and picosecond laser pulses. This implies that shorter pulse durations with reduced energy can be used surgically to significantly enhance controlability over currently utilixed nanosecond laser sources.

The limiting case of short pulse laser surgery was investigated using high intensity femotsecond laser pulses with pulse durations of 70 fs and peak exposure intensities of greater than 10^{12}w/cm^2 . Because of the high peak intensities but low pulse energies, strongly nonlinear effects could be observed. Laser ablation of the cornea could be performed using pulses at 620 nm wavelength where the cornea is nominally transparent.[6] Ultrastructural studies including light and transmission electron microscopy were performed to characterize the degree of collateral damage. These findings show a surprisingly small degree of thermal denaturation and a localization of the damage on a 10 μm scale. This high degree of incision control approaches results which can be obtained with some types of ultraviolet ablation.[7] Our study represents one of the first investigations using a high intensity visible laser for corneal ablation. Preliminary findings indicate that in contrast to UV ablation, nonlinear absorption plays a key role in energy deposition processes. The use of femtosecond lasers may thus provide a technique for the microsurgcial treatment of internal intraocular structures where UV ablation is not feasible.

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5.11 Short Wavelength Lasers

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A major objective of our research is to develop a small scale (table top) extreme ultraviolet laser (EUV) which operates in the 200-300 Å spectral regime. The purpose of this research effort is to explore the new short wavelength laser technology and its associated scientific applications. Serious exploitation of x-ray laser applications will require an economical, reliable and high-repetition-rate x-ray laser system. As a result, our initial efforts (and efforts of our colleagues at a number of laboratories) are primarily aimed at developing such a short wavelength laser source.

The scientific applications of x-ray lasers probably include:

1. Phase sensitive soft x-ray surface reflection probing.
2. Soft x-ray/optical nonlinear studies, and mixing to provide bright and tunable soft x-ray sources.
3. Short pulsed holographic imaging of microscopic surfaces, including biological surfaces.
4. Source of accurate wavelength standards in the soft x-ray regime.
5. Source for the creation of microscopic plasmas.

Undoubtedly many additional applications will become apparent in time. Our hope is that our efforts will help this technology to mature to the point of being of significant use to the scientific and industrial communities.

Our research group is also interested in a number of atomic physics and quantum electronics projects which we will report on in the next RLE Progress report.

5.11.1 Monopole Collisional Excitation Scheme

Electron collisional excitation as a mechanism to produce population inversions at EUV and soft x-ray wavelengths has played a key role in the successful demonstration of short wavelength amplification in recent years. Amplification has been observed in a number of highly stripped neon-like ions,[1–3] and more recently in nickel-like ions.[4] Searches for amplification in Nd-like ions are currently underway at a number of laboratories. We plan to pursue the electron collisional excitation scheme experimentally at lower Z in coming years at MIT.

The monopole excitation scheme is based on the occurrence of very large electron collisional excitation cross sections for the excitation of a highly stripped ion from a closed shell $^1S_0$ ground state to a singly excited state of the same parity and total angular momentum. For example, in neon-like ions, transitions of the type $1s^22s^2p^6\ {}^1S_0 - 1s^22s^2p^53p\,{}^1S_0$ can lead to strong population inversions on 3p-3s laser lines.[5] In nickel-like ions, the dominant electron collisional excitation drives the $1s^22s^2p^63s^2p^6d^{10}\ {}^1S_0 - 1s^22s^2p^63s^2p^6d^94d\,{}^1S_0$ transition which leads to gain on 4d - 4p transitions.[6]

The upper laser state in these schemes cannot radiatively back down to the ground state due to the parity selection rule, and as a result, inter-shell population inversions readily develop. The lower laser state is a short-lived J = 1 state, which radiatively decays rapidly back to the ground state.

The observation of significant amplification in nickel-like yterbium has been reported down to 50.2 Å,[4] which for the time being appears to be the shortest wavelength at which such amplification has been demonstrated in a laboratory. One of our goals is to study the 4d-4p laser lines in low Z nickel-like ions up to molybdenum (the strongest of which in molybdenum is computed to lie near 195 Å).

We have been involved during the past years in the simulation and analysis of collisional excitation schemes in Ne-like, Ni-like, and Nd – like[7] systems. The design of our low Z lasers is being carried out with the physics simulation codes LASNEX, XRASER and YODA, which have previously been used to design[8–12] nearly all of the laboratory x-ray laser experiments to date that have been done at Livermore.

5.11.2 Concept of a Benchtop Short Wavelength Laser Facility

One of the advantages of using the monopole collisional excitation scheme in the proposed wavelength regime is that the required intensity of the pump laser scales very rapidly with increasing wavelength of the output beam, ($\sim \lambda^{-6}$). Thus, by exploiting the isoelectronic transitions in lower Z ions ($Z = 38$-45), it is possible to not only reach the desired wavelength regime but also to achieve the necessary pump intensity on an academic laboratory scale. Currently no cheap optical pumping source is readily available; one of the objectives of this research is to develop such a system.

Once a population inversion has been generated, it is desirable to use a cavity to provide feedback and collimation of the short wavelength light. By allowing the signal to travel through an inverted plasma several times, the required pump energy is further reduced by approximately an order of magnitude. Because the EUV radiation is highly energetic, traditional cavity designs used in the infra-red and optical regimes will not sufficiently contain the radiation. Therefore, a second objective of this research is the development of a practical resonant cavity for the EUV regime.

Finally, to be considered a useful scientific device, a practical EUV laser must have at least a moderate repetition rate. Currently, there exists no short wavelength laser in the world that can generate pulses of EUV light at a repetition rate that is even close to rates normally associated with optical systems. Therefore, the entire system will be constructed to operate at rates on the order of 1 pulse every few seconds. Specifically, the third major goal of this work, is to develop a method of replenishing the target material that is used to make the EUV plasma at a rate consistent with this design criterion.

Some of these objectives are discussed in more detail in the following sections.

5.11.3 The Optical Pumping System

The pump laser system which will be used to create the laser plasma will consist of a conventional modelocked oscillator and preamplifier system which is commercially available, followed by a thin slab Nd-glass power amplifier.[13,14] (See figure 5.1). Our goal is to obtain approximately 50 Joules out of two power amplifiers in series.

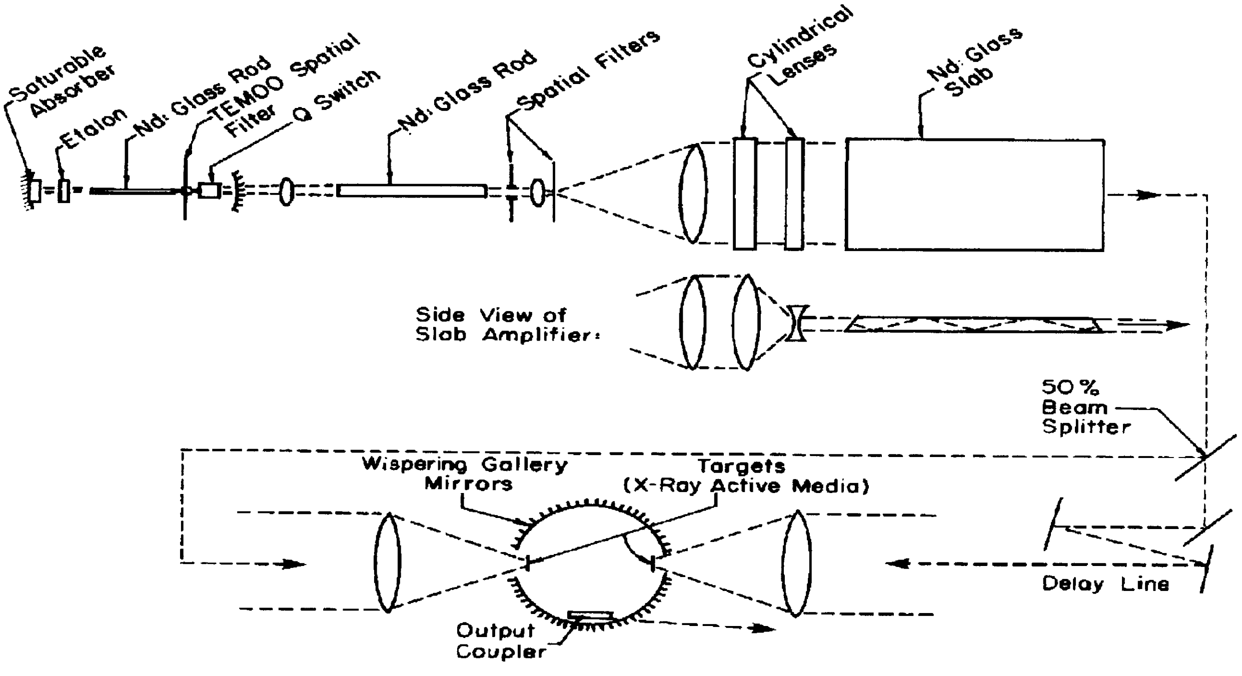

Figure 5.1 Schematic of pump system for a small scale short wavelength laser system.

Glass rod amplifiers are commonly used in high-power laser facilities, and we could in principle construct our power amplifiers using rods instead of slabs. Since glass is a poor thermal conductor, high-power glass laser amplifiers tend to cool rather slowly. The characteristic thermal relaxation time of a phosphate glass rod is approximately

$$\tau = 45\mathrm{R(cm)}^2\mathrm{sec.} \tag{1}$$

In order to stay below the damage threshold for the glass, we would require a rod of radius 2.5 cm, and would be faced with a corresponding cooling time of close to 250 seconds between shots.

The primary advantage of a thin slab amplifier over a rod amplifier is in the relative cooling rates of the two geometries. The double requirements of moderately high optical laser energy high repetition rate places severe constraints on the design of the pump laser, and these considerations have lead us to consider the more unconventional approach of thin slab power amplifiers. For example, a slab amplifier of thickness 0.4 cm which is cooled from one side would have a thermal relaxation time of 17 seconds. Cooling from both sides reduces this number to 4 seconds.

5.11.4 Whisper-Gallery Mode Mirrors in the Soft X-Ray Regime

A single pass amplification scheme for the EUV portion of the system for applications is impractical (although most soft x-ray laser work done to date has been on single-or double-pass systems), as it would require roughly an order of magnitude increase in the required pump energy. Normal incidence reflection is most often obtained using lossy multilayer mirrors in the EUV and soft x-ray regimes, with observed reflection coefficients approaching 50% per bounce. The extension of this technique to the construction of stable short wavelength laser cavities has not yet been explored.

An alternate approach using successive glancing angle reflection has lead to higher observed reflection coefficients for 180 degrees turning (the equivalent of a normal incident reflection for a multilayer mirror) from 100-140 eV.[15] This approach should scale favorable to the somewhat longer wavelengths of interest to us.

We have therefore examined the construction of such a cavity by exploiting whisper gallery modes (multiple glancing angle reflection modes), as shown in figure 5.2. Originally analyzed by Rayleigh in connection with acoustic waves, these modes can guide waves that are in grazing incidence to the mirrors. Stable cavities are easily constructed by arranging for the shape of the mirror to cause refocusing of the expanding beam which emerges from the plasma amplifier.

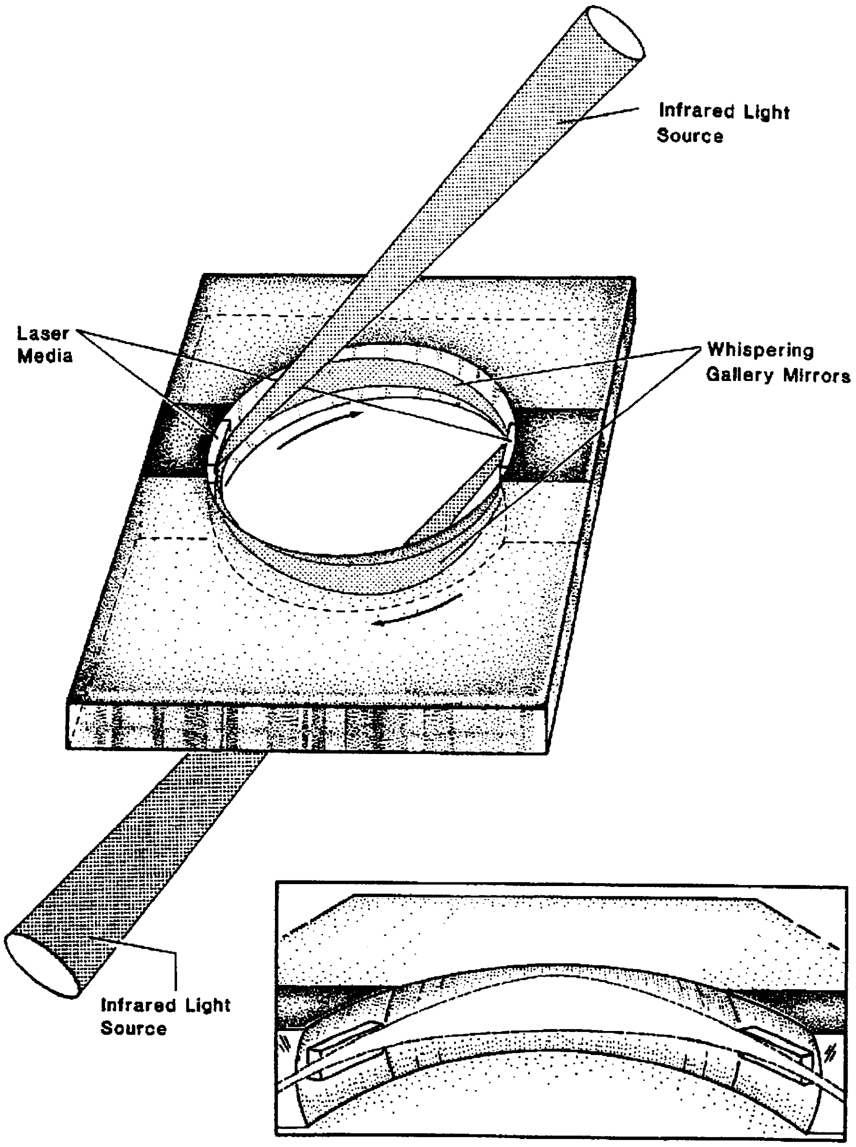

Figure 5.2 Illustration of concept of whispering gallery mirror for a short wavelength laser cavity.

As envisioned, the EUV resonant cavity will be spherical. Arranging two inverted plasmas antipodally will therefore guarantee that radiation in the cavity will always pass through the amplifying media, since any great circle on a sphere that passes through one antipode will necessarily pass through the other. Laser amplification is therefore possible twice for every round trip in the cavity.

Moreover, it can be shown that the radius of the cavity does not affect the efficiency of the whisper modes to first order. Thus the cavity can be constructed to a size convenient to the EUV pumping scheme. By subdividing the optical pulse to a train of 15 pulses, the inverted population of the plasma can be refreshed for each pass of the beam through the plasma amplifier. If the subpulses come 1 ns apart, a spherical cavity 10 cm in diameter will allow a complete round trip between plasma reheating.

5.11.5 A Fast High Resolution Soft X-Ray Detector

In conjunction with the development of the x-ray laser, research efforts have been made in our group developing schemes for time-resolved and time-gated x-ray detectors which are both efficient and have excellent spatial resolution.[16] Applications for these soft x-ray detectors include time-gated spectrometers and cameras for the soft x-ray laser beam, diagnostics for the plasma amplifiers, and imaging instruments for microscopic and holographic applications.

The proposed schemes are based on the use of optical nonlinearities of multiple quantum well (MQW) structures to obtain potentially fast (10 psec) and efficient x-ray detection with submicron spatial resolution. Specifically, two related schemes have been devised that are based on: 1) the scattering; and 2) the reflection of a pulsed optical probe beam by carriers produced by the absorption of a single energetic soft x-ray photon.

The mechanism of the detection relies on the strong carrier density dependence of the (complex) index of refraction of the exciton absorption lines in MQW GaAs/AlGaAs layers. In optical experiments, the carriers are usually generated by the absorption of an optical laser above the bandgap. In our schemes, we propose to create carriers directly by soft x-ray absorption.

Figure 5.3 shows the setup for the reflection scheme where the probe beam is incident from the back and the soft x-rays are incident on the front of the MQW structure. The pixels give the necessary spatial and temporal resolution. The pixels are essentially Fabry-Perot cavities whose refractive index can be altered by the carriers generated by the x-ray. The optical signal is reflected from individual pixels where soft x-ray absorption has occurred and is then imaged onto a CCD.

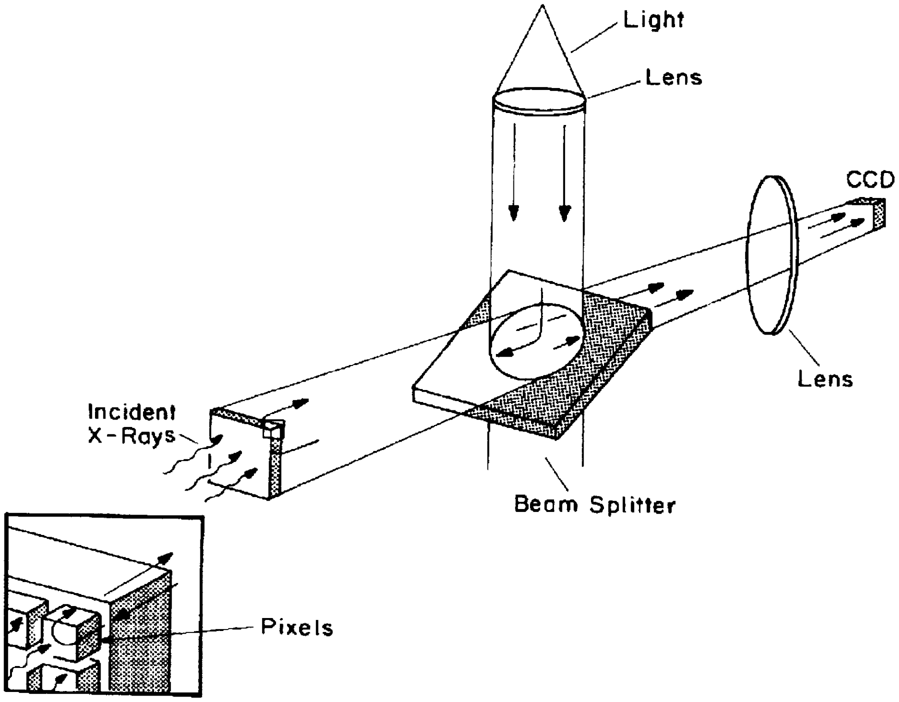

Figure 5.3 Microetalon approach to sort x-ray detection.

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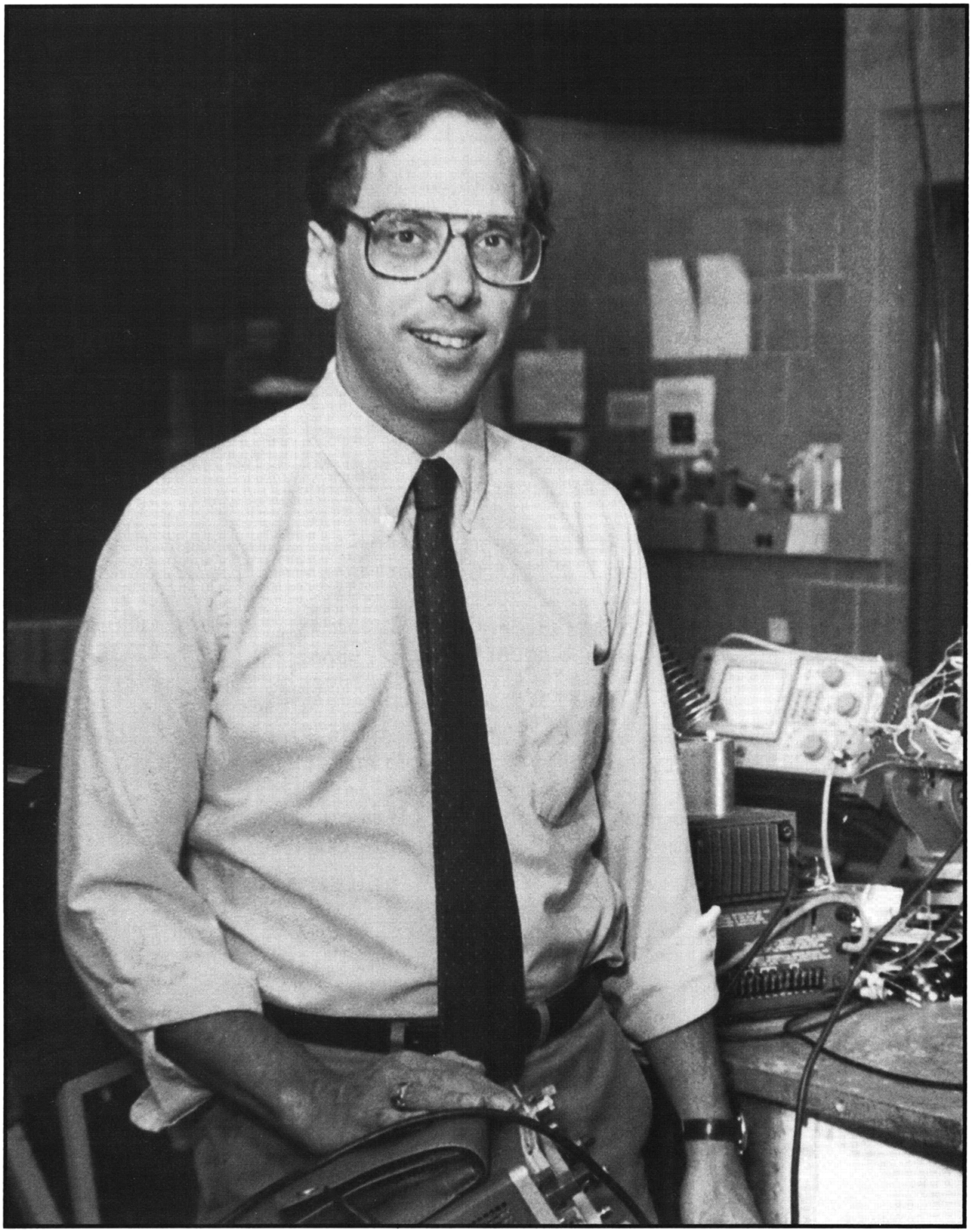

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