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We have constructed an ultrafast laser spectroscopy facility from equipment purchased under this instrumentation award. When optimized, this system will produce pulses with energies up to 100 microjoules with a 100 fs duration over a range of wavelengths tunable from 400 to 2000 nm. The short pulse duration, high beam quality, and high peak power available from the laser provides state-of-the-art capabilities for research including ultrafast spectroscopy, characterization of nonlinear optical materials, control of self-action effects, and quantum control of wavepacket dynamics.

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Instrumentation for the Characterization and Spectroscopy of Nonlinear Optical Materials

Final Report

Defense University Research Instrumentation Program
AFOSR F49620-1-0127

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The Ultrafast Nonlinear Optics Facility

1. Introduction

We describe the state-of-the art experimental facilities for femtosecond nonlinear optics that we have developed under grant AFOSR F49620-1-0127. The instrumentation allows us to perform spectrally resolved and ultrafast time-resolved measurements of the nonlinear optical response of novel nonlinear optical materials, especially organic/polymeric materials, and to perform studies of fundamental optical processes for the development of novel nonlinear materials.

In particular, we have purchased and constructed a low-repetition rate, high pulse energy Ti:Sapphire regenerative amplifier, and pump laser, and an optical parametric generator. The capabilities of this system are the generation of 1 mJ pulses of less than 100 fs duration at 800 nm and pulses of 10 μJ and 150 fs tunable from 1.1 μm to 1.9 μm. Also, installed with this system is diagnostic equipment and instrumentation for experiments in nonlinear optics, such as visible and uv spectrometers, a CCD camera, a transient digitizer and an OMA, an autocorrelator, and various oscilloscopes and detection equipment.

This facility will continue to support DOD-funded research programs from 3 co-PIs with backgrounds in chemical engineering, optics, applied physics and physics who already have established successful joint research efforts, and have ample experience with nonlinear optical materials, processes and devices. The common theme of their research programs is the need for high peak power femtosecond laser pulses tunable from the visible to the mid-infrared, which this facility will provide. Examples of projects include the development of nonlinear optical materials (in particular organic polymers and semiconductor and other composites) and devices for the near and mid-infrared regions, novel ultrafast spectroscopic techniques for measuring weak and rapid nonlinear optical responses and applications of quantum optical phenomena to nonlinear optics, such as the enhancement of optical nonlinearities via quantum interference and the exploration of nonlinear optics at the one-photon level.

This facility has already increased the activities of the University of Rochester community that is active in nonlinear optical materials and processes research. Two graduate students are already pursuing their thesis research using the apparatus, and a postdoctoral research associate from Eastman Kodak company is using the facilities on a part-time basis for ongoing collaborative research.

2. Instrumentation for the Characterization and Spectroscopy of Nonlinear Optical Materials

The ultrafast laser system that has been installed under this grant has opened up new possibilities for exploring hithertofore inaccessible areas in physics and engineering. We are now poised to take advantage of these developments, and to extend our research capabilities into these areas. This multi-purpose instrument is being used for cutting-edge experiments in nonlinear optical materials characterization and nonlinear optical physics.

The components of each of the laser subsystems that have been installed in the facility are illustrated schematically in the following paragraphs and figures.

2.1 The ultrafast laser system

The essential features of the laser system are that it is capable of generating short pulses (about 100fs duration) with tunable mean wavelength (400nm to >5μm), with large energy (from 10 μJ to >1mJ per pulse, depending on the wavelength) at a moderate repetition rate (10Hz). The ability to generate several different wavelengths synchronously is a significant feature of our system. The main components of the system are diagrammed in Fig. 1. We purchased the unshaded items in this figure: a high power Nd:YAG laser that pumps a T:S regenerative amplifier and an optical parametric amplifier. These items are used in conjunction with equipment already available; an Argon-ion laser and a cw-modelocked Titanium-Sapphire (Ti:S) laser.

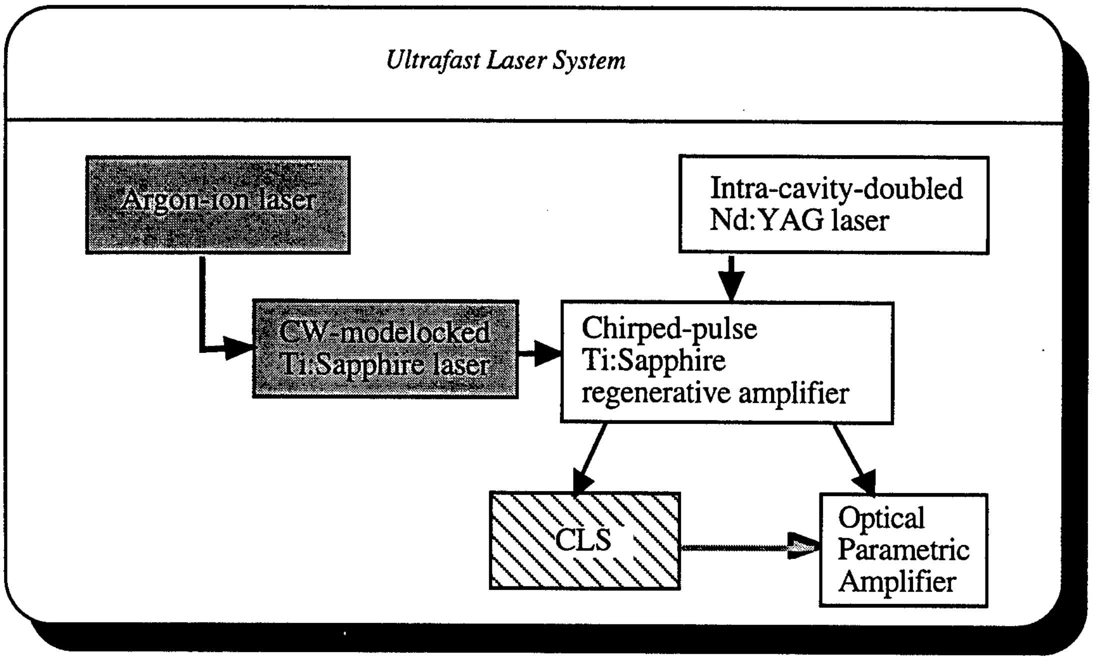

Fig. 1. The proposed high-power, ultrashort-pulse laser system. The shaded components (Argon-ion and Ti:Sapphire lasers) already exist in our laboratories. Unshaded components will be purchased, and a continuum light source (CLS) will be developed as part of this project.

The regenerative amplifier system is a Clark-MXR, Inc. model CPA-10-1 Titanium Sapphire chirped-pulse-amplifier (CPA). This is seeded by a Spctra-Physics Lasers, Inc. model Tsunami modelocked Ti:S oscillator, and is capable of delivering more than 1mJ of pulse energy in a pulse of 200fs duration at a repetition rate of 10Hz. The beam quality at the output of the system is close to the diffraction limit, primarily because of the use of a regenerative, as opposed to a multi-pass, amplifier system. This means that all of the energy is focussable into a very small spot (about 10μm diameter) so that extremely high peak intensities, on the order of 10^{16}W/cm^2, are obtainable. These powers are certainly adequate to perform the nonlinear optical spectroscopic experiments that are of interest to us, such as the characterization of the ultrafast nonlinear response of polymers.

The large pulse energy alone is a significant advance over the Ti:S oscillator alone (and indeed a synchronously pumped optical parametric oscillator OPO) using this as a pump source), and it is this feature which allows us to extent the useful wavelength range that the system can generate. An optical parametric amplifier allows the generation of radiation from the visible through the mid-infra-red (MIR) region of the electromagnetic spectrum, with all wavelengths emitted in pulses that have subpicosecond duration, with close to transform-limited bandwidth.

The pump source for the OPA is the 1mJ, 10Hz pulse train at 800nm generated by the Ti:S amplifier. The OPA typically produces pulses of 200fs duration with energies between 1 and 10μJ. Such large energies in a transform and diffraction-limited output pulse enables the study of the wavelength dependence of the efficiency of the generation of high-harmonics, and allow their feasibility as a probe of surfaces and structures to be evaluated. The OPA may also be operated at wavelengths as short as 400nm by pumping it with the frequency-doubled output of the Ti:S laser system. This further extends the useful range of wavelengths accessible to this system to the visible.

The numerous individual components will require stable surfaces so that they are reliably interconnected. For this reasons we purchased a 5 ft by 15 fr optical table. This provides sufficient surface area to contain the entire laser system. An adjacent 4 ft by 10 ft optical table provides ample space for several experimental setups. None of the DURIP budget was used for diagnostic equipment, since such capabilities are already available in our laboratories. We have at present several spectrometers and intensity autocorrelators to provide real-time measurement of the pulse spectrum and duration continually during the operation of the system. Our expertise in the design and construction of autocorrelators in the past leads us to believe that we can fabricate instruments with better performance than a commercial model at a fraction of the cost. Further, we can custom-design each of the correlators for a specific application, such as a particular wavelength range, or input power.

In summary, the new laser system provides several features that will dramatically further our research programs, which span the fundamental to the applied ends of the physics spectrum. We are constructing a state-of-the-art, high power short pulse laser system based on technology that has become available within the past two years. Further, we are also developing a high-power continuum source that will use this laser system as a pump, and will generate pulses of less than 6 fs duration.

2.2. Detection and experimentation apparatus

We are also developing several detection and experimentation capabilities as part of our facility.

One of the prime techniques involves z-scan measurements as a function of the laser wavelength and laser pulse duration. We expect that this approach will ultimately be limited by the inherent pulse broadening that occurs as a laser pulse propagates through a dispersive medium. We are also developing new techniques for the characterization of ultrafast photonic materials. One such approach is to use recently developed methods to measure the electric field of a laser pulse after propagating through a nonlinear optical material to determine how the combined action of nonlinearity and dispersion have influenced the temporal-spectral profile of the pulse. This method, when combined with spectral interferometry will enable measurements of very thin samples, or measurements far from resonance to be made.

One of the new techniques soon to be available in this facility is the capability to completely characterize the electric field of the femtosecond optical pulse that is incident and reflected or transmitted from the sample. A novel nonlinear optical interferometric method, SPDIER, and spectral interferometry (in this guise known as TADPOLE) will be used to measure the output field. The ability to measure the amplitude and phase of ultrashort pulses both before and after the sample means that the nonlinear response of a material can be determined with unprecedented accuracy. In fact, this technique can be extended to measure the quantum statistical properties of the pulses, thereby opening the door to characterizing extremely weakly nonlinear, or thin, samples.

3. List of purchased equipment

The following table lists all of the new equipment that has been purchased under this grant.

Ultrafast	Laser	System
Ti:Sapphire CPA/OPA	$110k	Clark-MXR, Inc.
Optical Table	$10k	TMC, Inc.
Argon plasma tube	$6k	Evergreen Laser
Intracavity doubled Nd:YAG	$28k	Continuum, Inc.

	Totals
Total cost of equipment purchased	$154k
Matching funds from University of Rochester	$25k
DoD funds	$129k

3. New Research Capabilities

The Ultrafast Nonlinear Optics facility now contains state-of-the-art apparatus for the characterization of novel nonlinear optical materials and processes, including organic polymers. Our approach is based on developing experimental techniques for measuring both the amplitude and phase of the nonlinear response function of materials with ultrafast temporal resolution and high sensitivity. Such capabilities are likely have significant impact on several areas of current interest to DoD including characterizing the high-speed photonic response and dispersion of organic polymers, and the creation of novel states of matter exhibiting enhanced optical properties.

The following specific capabilities are now available for future research projects:

• *Measurements of the nonlinear optical response of polymers and other organic materials.*

Professor Jenekhe spearheads our effort in designing new organic molecules and polymers for use in photonics. We are particularly interested in the use of rigid rod polymers, which possess great mechanical strength and hence a high threshold to laser damage, for these applications. We are also interested in the studies of blends and copolymers, and in developing an understanding of how the nonlinear properties of these systems can be enhanced, for example through the use of exciplexes. The new instrumentation allows us to to study these properties over broad spectral regions and with the use of ultrashort pulses.

• *Measurements of the nonlinear optical response of composite materials*

Recent work, in particular by members of our Rochester team, has demonstrated that composite optical materials can possess large optical nonlinearities which under proper circumstances can exceed those of the starting materials. Our work in this exciting field will continue and will be enhanced by the presence of the ultrafast laser facility. Specific questions that we are begining to address include the following: how can we optimize the enhancement of the nonlinear optical response of a composite material, what is the ultimate speed of a photonic material, and is there any modification of the time response that results from the use of a composite structure, and do composite materials possessing a fractal structure possess unusual photonic properties.

• *Measurements of the nonlinear optical response of semiconductor quantum confined structures.*

Our work in this area will deal both with doped glasses and with semiconductor MQW devices fabricated by our colleague G. Wicks. We are interested both in fundamental questions, such as the ultimate speed of semiconductor doped glasses and metallic colloids, and in issues such as the dynamical behavior of semiconductor optoelectronic devices. The proposed instrumentation will allow us to address these important issues.

• *Quantum coherence effects in novel nonlinear materials*

One of the important areas of recent research interest has been the development of materials which have large nonlinear susceptibilities. The availability of materials which possessed both a large magnitude and a fast responding nonlinearity would open up several new areas of application. There is a strong connection between the phenomenon of quantum-enhanced optical nonlinearities and the localization of the quantum state of the scatterer. If an internal degree of freedom of the scatterer that is coupled to the optical transition is highly localized (say, for example that the vibrational degree of freedom of a molecule is in a wave packet state) then the linear and nonlinear susceptibilities of the electronic transition are enhanced by an amount that is proportional to the degree of localization. Simple calculations show, for example, that if the vibrational mode of a dimer is in a quadrature squeezed state with position dispersion of about one half of the zero-phonon state, then the magnitude of the linear response function is enhanced by a factor of $e^4 = 55$. The proposed facility, will enable us to perform experiments which explore the limits of enhancement available by conducting a series of measurements on the nonlinear optical response of small molecules in non-stationary wave packet states.

• *Quantum optical effects in nonlinear optics,*

For example, single-photon nonlinear optics. Our work in this area is directed at the development of materials with sufficiently large nonlinearities that weak beams containing only a few photons can induce a nonlinear response, and at studying how quantum statistical effects can influence the characteristic of nonlinear optical devices and interactions. In this area, we shall examine how the nonlocal material response that is present in a semiconductor microstructure, for example, due to the exciton-polariton coupling, affects the passage of single, ultrashort photon wave packets through the material. Several of the new techniques we propose to develop, such as sub-shot-noise ultrafast detection, will be necessary for such experiments, and they could not be performed at present.

• *Ultrafast photodetection beyond the shot-noise limit.*

Many of the proposed experiments of novel material characterization require pulsed excitation and subsequent analysis of the scattered radiation. The ability to obtain information on the scattered field with high temporal resolution at or below the shot-noise limit will provide an environment where extremely weak or fragile samples can be tested. We have recently developed a technique that allows the complete quantum state of a light pulse to be constructed with femtosecond resolution using a spectrally-resolved DC-balanced homodyne detector. Its most innovative feature is that it can measured temporally multimode radiation, so that sub-shot-noise correlations between different time-slices of the pulse can be measured. This is in contrast to the conventional DC-balanced homodyne detection which can return the joint statistics of only one (or recently two) modes. Our apparatus will measure joint statistics of as many as 512 modes at once, depending on the photodetector array used and the resolution of the spectrometer. Such low-noise measurement techniques are vital in probing the quantum nature of relatively dilute samples.

4. Impact on Training of Future Scientists and Engineers

The Ultrafast Nonlinear Optics laser facility is expected to have a major impact on the research program and the education and training of students at the University of Rochester. These future scientists and engineers will be trained in areas of direct interest to DoD and thus will be available for future research and development in areas of importance to DoD and the Nation.

With three co-PIs, the laser system can be used much more efficiently than would be possible for a single researcher. An added benefit is the collaborative nature of this proposal. The very nature of the proposed work will lend itself to synergistic interactions among the researchers. Each is able to impart his expertise to the full range of experiments. Moreover, each investigator benefits from a wider range of experience and already joint research projects have their genesis in the activities associated with this facility.

The education of post-doctoral, graduate, undergraduate and secondary students will continue to be enhanced by the proposed laser system. The increased research activity associated with this laser system greatly expands the opportunities for the involvement of these students in the program. Presently, there are 4 post-doctoral research associates and 10 graduate students who will directly benefit from the new laser system. Many are all involved in research sponsored by DoD, including the Army and the Air Force, with additional support from other federal, state or industrial sources. In addition to this traditional form of student's involvement in research, the University of Rochester has been involved with innovative programs aimed at undergraduate and secondary students, such as the Advantage Scholars Internship Program, which allows students with merit scholarships an opportunity to work on projects during the summer, and the Ronald E. McNair Post-Baccalaureate Achievement Program, which seeks to increase the number of under-represented minorities entering Ph.D. programs. It is through the ongoing involvement of undergraduate and secondary students in our research programs that we are able to attract larger numbers of women and under-represented minorities into science and engineering. The new laser facility enhances that involvement.