

REPORT DOCUMENTATION PAGE

AFRL-SR-BL-TR-00-

0255

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1. AGENCY USE ONLY (Leave blank)		2. REPORT DATE	3. REPORT TYPE AND PERIODICITY Final - 01 Mar 98 - 28 Feb 99
4. TITLE AND SUBTITLE DURIP-97 Optical Source For Organic and Polymeric Nonlinear Device and Material Testing			5. FUNDING NUMBERS F49620-97-1-0114
6. AUTHOR(S) Dr Eric W. Van Stryland / Dr David Hagan			
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) CREOL University of Central Florida Orlando FL 32826			8. PERFORMING ORGANIZATION REPORT NUMBER
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) AFOSR/NL 801 N. Randolph St., Rm 732 Arlington VA 22203-1977			10. SPONSORING/MONITORING AGENCY REPORT NUMBER
11. SUPPLEMENTARY NOTES			
12a. DISTRIBUTION AVAILABILITY STATEMENT Approved for Public Release: Distribution Unlimited			12b. DISTRIBUTION CODE
13. ABSTRACT (Maximum 200 words) We used the DURIP funds as requested to purchase equipment to build a state-of-the-art nonlinear spectroscopy facility at CREOL. This facility, based on a CLARK-MXR CPA-2000 (recently replaced by the company with a CPA-2001 at no charge) has already produced a wealth of nonlinear spectroscopic data. Armed with these data we have published several articles and given many talks on the subject. The facility works well and has been used to determine the nonlinear optical properties of organic materials and semiconductors. The system is now primarily used to measurements of the spectrum of nonlinear absorption and dispersion of nonlinear refraction in organic materials. Such spectra are crucial for determining the applicability and the range of application of organic materials for sensor protection and optical switching. These measurements are now allowing physical models to be properly tested. For example, a 5-level model for reverse saturable absorbers has been shown to work for a large class of organic dyes. We also added an optical parametric generator/amplifier to the system to allow tuning of the excitation wavelength. The parametric oscillator also enables us to absolutely calibrate the system by performing Z-scans at specific wavelengths. This is an important advance in our characterization capability for organic materials.			
14. SUBJECT TERMS Spectroscopy, Nonlinear, Z-scans		20000718 061	
17. SECURITY CLASSIFICATION OF REPORT UNCLASS		15. NUMBER OF PAGES 4	
18. SECURITY CLASSIFICATION OF THIS PAGE UNCLASS		16. PRICE CODE	
19. SECURITY CLASSIFICATION OF ABSTRACT UNCLASS		20. LIMITATION OF ABSTRACT	

Final Technical Report  
DEPARTMENT OF DEFENSE UNIVERSITY  
RESEARCH INSTRUMENTATION PROGRAM

Optical Source for Organic and Polymeric Nonlinear  
Optical Device and Material Testing

submitted by

CREOL  
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We used the DURIP funds as requested to purchase equipment to build a state-of-the-art nonlinear spectroscopy facility at CREOL. This facility, based on a CLARK-MXR CPA-2000 (recently replaced by the company with a CPA-2001 at no charge) has already produced a wealth of nonlinear spectroscopic data. Armed with these data we have published several articles and given many talks on the subject. The facility works well and has been used to determine the nonlinear optical properties of organic materials and semiconductors. The system is now primarily used for measurements of the spectrum of nonlinear absorption and dispersion of nonlinear refraction in organic materials. Such spectra are crucial for determining the applicability and the range of application of organic materials for sensor protection and optical switching. These measurements are now allowing physical models to be properly tested. For example, a 5-level model for reverse saturable absorbers has been shown to work for a large class of organic dyes. We also added an optical parametric generator/amplifier to the system to allow tuning of the excitation wavelength. The parametric oscillator also enables us to absolutely calibrate the system by performing Z-scans at specific wavelengths. This is an important advance in our characterization capability for organic materials.

We are using the experimental instrumentation to measure the nondegenerate nonlinear absorption spectra of materials by exciting the material with a femtosecond excitation beam at any wavelength from 500 nm to 1.6  $\mu\text{m}$  and then probing the excited material with femtosecond time delay accuracy with a femtosecond continuum. This is the basis for the Femtosecond Continuum Nonlinear Spectrometer developed at CREOL. This sophisticated optical system produces an ultrashort "white light" probe pulse including wavelengths from greater than 2  $\mu\text{m}$  to  $\approx 250$  nm.

Our spectral characterization facilities now allow us to monitor wavelengths from 1.7 mm to 200 nm (dual InGaAs linear array with spectrometer from 1.7 mm to 850 nm and two CCD arrays with spectrometer from 850 nm to 200 nm).

Our contribution to the government using this equipment is:

- a) supplying accurate measurements of the nonlinear absorption spectrum of materials (specifically organics such as phthalocyanine dyes);
- b) time resolving the nonlinear absorption spectrum. This will allow separation of two-photon and excited-state nonlinearities.
- c) calculation of the dispersion of the nonlinear refraction (including temporal response);
- d) direct measurement of the degenerate nonlinear refraction and absorption at any desired wavelength in the visible and near-IR;
- e) incorporation of these results into a framework of fundamental understanding;
- f) utilizing these results to effect improvements in NLO materials being developed by the government (both in-house and by its contractors).

Publications and presentation resulting from the addition of this equipment are:

"Introduction to Ultrafast and Cumulative Nonlinear Absorption and Refraction", E. W. Van Stryland, in *Beam shaping and control with nonlinear optics*, Eds. F. Kajzar and R. Reinisch, Plenum, p. 39, New York 1998.

"Z-scan Technique for Materials Characterization", Eric Van Stryland and M. Sheik-Bahae, in *Materials Characterization and Optical Probe Techniques*, Critical Reviews of Optical Science and Technology, Vol CR69, 501-524, SPIE 1997.

"Purely Refractive Transient Energy Transfer Via Stimulated Rayleigh Wing Scattering", A. Dogariu, T. Xia, D. Hagan, A. Said, E. Van Stryland, and N. Bloembergen, *J. Opt. Soc. Am. B*, Vol. 14, No. 4, 796 (1997).

"Reverse saturable absorption in polymethine dyes", S. Khodja, D. J. Hagan, J. Lim, O. V. Przhonska, S. Yang, J. Buckley and E. W. Van Stryland, *Proc. SPIE-3146* (1997).

"Introduction to Ultrafast and Cumulative Nonlinear Absorption and Refraction", E. W. Van Stryland, NATO Summer School on Beam shaping and control with nonlinear optics, Cargese Corsica, France, Aug. 4-15, 1997.

" $\chi^{(3)}$ 's: Their Characterization and Understanding", E. W. Van Stryland, Nonlinear Optics '98, Materials, Fundamentals and Applications Topical Meeting, Kauai, HI, Aug. 1998.

"Picosecond Visible CLBO Optical Parametric Oscillator", Scott C. Buchter, JinHong Lim, Sean Ross, Hans P. Jenssen, E.W. Van Stryland, Arlete Cassanho, Greg Mizell, Advanced Solid State Lasers meeting, Coeur d'alene, Idaho, Feb. 2-4, 1998.

"Excited-State Absorption Spectroscopy of Metallo-Porphyrin Dyes with Both Q and B-Band Excitation", T. Sean Ross, David J. Hagan, Arthur Dogariu, Paul M. Buck and Eric W. Van Stryland, CLEO 98, San Fransisco, 1998.

"Spectral Measurements of Nonlinear Absorption in Polymethine and Squarylium Dyes", J.H. Lim, O. Przhonska, T. S. Ross, D. Hagan, and E. Van Stryland, CLEO 98 San Fransisco, 1998.

"Measuring Nonlinear Spectra with a Visible, Picosecond Parametric Oscillator", T.S. Ross and E. Van Stryland, Annual meeting of the Optical Society of America, Long Beach, CA, 1997.

"Photophysics and Nonlinear Absorption of Dye-Doped Polymeric Media", J. Lim, O. Przhonska, D. Hagan, S. Khodja, E. Van Stryland, Annual meeting of the Optical Society of America, Long Beach, CA, 1997.

"Laser Beam Propagation through Nonlinear Media", D. Kovsh, S. Yang, S. Khodja, D. Hagan, E. Van Stryland, Annual meeting of the Optical Society of America, Long Beach, CA, 1997.

"Nonlinear Light Absorption of Polymethine Dyes in Liquid and Solid Media", Olga V. Przhonska, Mikhail V. Bondar, Yuriy L. Slominsky, Jinhong Lim, David J. Hagan, and Eric W. Van Stryland, JOSA B, 15, 802-809 (1998).

"Nonlinear spectrometry of chromophores for optical limiting", D. J. Hagan, E. Miesak, R. Negres, S. Ross, J. Lim and E.W. Van Stryland, SPIE, Proc. SPIE-3472, 80-90 (1998).

"Synthesis & Characterization of New Two-Photon Absorbing Polymers", K. Belfield, B. Reinhardt, L. Brott, S. Clarson, O. Najjar, S. Pius, E. Van Stryland, and R. Negres, Polymer Preprints, 40, 127-128 (1999).

"Two-Photon Absorption vs Reverse Saturable Absorption and RSA in Polymethine Dyes", E. Van Stryland, US/UK Optical Limiter Modeling Workshop, Air Force Research Laboratory, WPAFB, OH, May 18-21, 1998.

"Host Dependent Excited State Lifetime in Polymethine Guest-Host", Jin Hon Lim Olga Przhonska, D. Hagan, E. Van Stryland, Annual meeting of the Optical Society of America, Baltimore, 1998.

"Femtosecond white light continuum for nonlinear index dispersion measurements", Raluca A. Negres, Ed Miesak, David Hagan, Eric W. Van Stryland, Annual meeting of the Optical Society of America, Baltimore, 1998.

"Microfabrication via Two-Photon Photoinitiated Polymerization", Kevin D. Belfield, X. Ren, D. Hagan, E. Van Stryland, V. Dubikovskiy and E. Miesak, American Chemical Society, Fal meeting Ploymer Materials Science and Engineering Division, New Orleans, 1999.

“Ultrafast Nonlinear Optical Spectrometer for Material Characterization”, R. Negres, E. Van Stryland, D. Hagan, K. Belfield, and B. Reinhardt, American Physical Society Centennial meeting, Atlanta, Georgia, 1999.

“Polymethine and squarylium molecules with large excited-state absorption” Jin Hong Lim, Olga V. Przhonska, Salah Khodja, Sidney Yang, T.S. Ross, David J. Hagan, Eric W. Van Stryland, Mikhail V. Bondar and Yuriy L. Slominsky. Chemical Physics, (Accepted, in Press, 1999)

In addition the following Ph.D. students used some of the equipment listed in the pursuit of their degrees. They have now graduated.

Arthur Dogariu, “Spectral and Temporal Response of optical Nonlinearities”, Physics, 1997

T. Sean Ross, “A Picosecond Visible Optical Parametric Oscillator as a Tool for Nonlinear Spectroscopy”, Electrical and Computer Engineering, 1998

JinHong Lim, “Nonlinearities of Polymethine and Squarylium Molecules for Optical Limiting”, Physics, 1998

Edward Miesak, “Femtosecond Tunable Light Source”, Electrical and Computer Engineering, 1999

The following describes the major equipment purchased with this DURIP funding:

Many optical components and optical hardware were also purchased which are all parts of the continuum spectroscopy system we assembled.

Clark MXR, Model CPA 2000 Ti:Sapphire Regenerative Amplifier System (this was recently replaced with a Model 2001 at no cost)	\$186,000
Optical Crystals	\$3,357
Inrad, Optical hardware	\$6,044
Optical Parametric Oscillator Components (Primarily Quantronix)	\$41,643
Dual diode array based Detection system (primarily Princeton Instruments)	\$17,726

The combination of these components along with existing equipment resulted in our establishment of a state-of-the-art nonlinear spectroscopy facility. As far as we are aware there is no other comparable facility.