CREOL

(Center for Research in Electro-Optics and Lasers)

FINAL REPORT

OPTICAL POWER LIMITER
DAAK20-83-C-0164
October 21, 1988

DARPA/CNVEO

UNIVERSITY OF CENTRAL FLORIDA
Orlando, Florida
32816

CENTER FOR RESEARCH IN ELECTRO OPTICS AND LASERS

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I. ABSTRACT

We have performed a fundamental study of optical/materials interactions which can be used for passive control of the spatial energy (or power) distribution of optical beams. The study included materials for use in various spectral bands within the spectral range of the UV to 11 microns and the temporal range from cw to picoseconds. We concentrated on materials and concepts with broadband response.

Materials studied under DARPA/CNVEO program include aromatic liquids, liquid crystals, semiconductors, and liquid suspensions. We have demonstrated optical limiting using nonlinear absorption in liquid crystals, optical self-action (self-focusing) in liquids and solids, and multiphoton absorption plus sub-sequent self-defocusing in semiconductors. We have theoretically modeled the nonlinearities in these materials and understand the limiting mechanisms although in the organic materials the magnitudes of the nonlinearities are not well understood. In the semiconductors we have developed a predictive capability that allows us to calculate the nonlinearity knowing only a few fundamental material parameters. In addition, we have developed a preliminary model that explains the very low limiting thresholds observed in a liquid suspension of carbon particles as due to nonlinear scattering from rapidly expanding microplasmas initiated by thermionic emission.

The dynamic ranges of these devices is determined by the ratio of the energy or power at which the device itself is damaged irreversibly to the limiting energy or power. We have demonstrated dynamic ranges of greater than 10^3 in liquids and we have observed even larger dynamic ranges in bulk ZnSe. However in liquids the dynamic range of the device is determined by optical damage to the container walls (usually a cuvette), since liquids are self-healing and this dynamic range should be easily extended. A monolithic semiconductor limiting device with a measured dynamic range of > 10^4 was recently fabricated and tested using psec 0.53μm pulses. This device had a limiting energy of 10^{-8} Joules which corresponds to a limiting power for psec pulses of ~300 Watts. Using a similar ZnSe based limiter we have

obtained limiting at ~80 Watts although the dynamic range was reduced. Measurements of output spatial profiles and temporal profiles as recorded by a 2 psec resolution streak camera verify that no "hot" spots occur at the output in space or time for either semiconductor limiter even for high input powers.

Great care has been taken over the past several years to develop well characterized laser systems for use in nonlinear optics research. In particular, the laser facilities including picosecond and nanosecond laser systems have been meticulously tuned to operate in the Gaussian TEM_{OO} mode such that the free space propagation is well known. This allows us, along with our careful temporal pulsewidth measurements, to accurately measure deviations from free space linear propagation in these materials and thereby, determine their nonlinear optical responses.

The work emphasized materials with broadband capability. That being the case, materials exhibiting two-photon absorption (2PA) or nonresonant self-action and/or scattering were stressed. For example, ZnSe exhibits nearly constant 2PA from 900 to 450 nm, and the suspension of carbon particles should have an even braoder bandwidth.

In what follows we list publications and presetations that have resulted from this reseach and then summarize the basic results with reference to the appropriate publications.

II. PUBLICATIONS AND PRESENTATIONS

The results of research sponsored under this program have been presented in 48 presentations at national and international conferences, and have been published in 41 scientific articles as summarized below. Reprints of publications are included at the end of this report.

PUBLICATIONS

Invited

1. "Picosecond Damage Studies at 0.5 and 1 μm," M.J. Soileau, William E. Williams, E.W. Van Stryland, Thomas F. Boggess, and Arthur L. Smirl, Opt. Eng. 22, 424 (1983).

2. "Laser Light Induced Bulk Damage to Optics," M.J. Soileau, E.W. Van Stryland and William E. Williams, critical review in Radiation Effects in Optical Materials, Paul W. Levy, ed., SPIE 541, 110-122 (1985).

3. "Passive Optical Limiting with Picosecond Response", M.J. Soileau, S. Guha, and E.W. Van Stryland, in Ultrashort Pulse Spectroscopy and Applications, M.J. Soileau, ed., SPIE 533, 144-148 (1985).

4. "Passive Optical Limiting", M.J. Soileau, E.W. Van Stryland, and Shekhar Guha, SPIE 540, 520-527 (1985).

5. "Two-Photon Absorption, Nonlinear Refraction and Optical Limiting in Semiconductors", E. W. Van Stryland, H. Vanherzeele, M.A. Woodall, M.J. Soileau, Arthur L. Smirl, Shekhar Guha, Thomas F. Boggess, Opt. Eng. 24, 613 (1985).

6. "Nonlinear Absorption and Associated Refraction in Semiconductors", E.W. Van Stryland, H. Vanherzeele, Shekhar Guha, M.A. Woodall, and M.J. Soileau, Proceedings of the IVth International Symposium on Ultrafast Phenomena in Spectroscopy, Teubner-Texte zur Physik, Ed. Edgar Klose, and Bernd Wilhelmi, GDR (1985).

7. "Nonlinear Optical Properties of Liquid Crystals in the Isotropic Phase", M.J. Soileau, E.W. Van Stryland, Shekhar Guha, E.J. Sharp, G.L. Wood, and J.L.W. Pohlmann, Mol. Cryst. Liq. Cryst. 143, 139, (1987).

8. "Nonlinear Beam Propagation in Semiconductors", E.W. Van Stryland, D.J. Hagan, M.J. Soileau, Y.Y. Wu, Proceedings of the International Conference "Lasers 87", (1987).

9. "Influence of Self-Focusing on Bulk Laser Induced Damage", M.J. Soileau, E.W. Van Stryland, SPIE proceedings of the O-E LASE conference, to be published 1988.

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10. "Optical Power Limiter with Picosecond Response Time," M.J. Soileau, William E. Williams, and E.W. Van Stryland, IEEE J. Quantum Electron. QE-19, 731 (1983).

11. "Optical Limiting by Two-Photon Absorption and Nonlinear Refraction in GaAs," T.F. Boggess, Arthur L. Smirl, Steven C. Moss, Ian W. Boyd, and E.W. Van Stryland, IEEE J. Quantum Electron. QE-21, 488 (1985).

12. "Studies of the Nonlinear Switching Properties of Liquid Crystals with Picosecond Pulses," M.J. Soileau, Shekhar Guha, William E. Williams, Eric W. Van Stryland, Juergen L.W. Pohlmann, E.J. Sharp, and Gary Wood, Mol. Cryst. Liq. Cryst. 127, 321 (1985).

13. "Optical Switching and n_2 Measurements in CS_2," William E. Williams, M.J. Soileau, and Eric W. Van Stryland, Opt. Commun. 50, 256 (1984).

14. "Energy Band-Gap Dependence of Two-Photon Absorption," Eric W. Van Stryland, M.A. Woodall, H. Vanherzeele, and M.J. Soileau, Opt. Lett. 10, 490 (1985).

15. "Self-Defocusing in CdSe Induced by Charge Carriers Created by Two-Photon Absorption," Shekhar Guha, Eric W. Van Stryland, and M. J. Soileau, Opt. Lett. 10, 285 (1985).

16. "Verification of the Scaling Rule for Two-Photon Absorption in Semiconductors," Eric W. Van Stryland, Shekhar Guha, H. Vanherzeele, M.A. Woodall, M.J. Soileau, and Brian S. Wherrett, Optica Acta 33, 381-386 (1986).

17. "Self-Focusing in CS_2 at 10.6 μm," M. Mohebi, P.F. Aiello, G. Reali, M.J. Soileau, and Eric W. Van Stryland, Opt. Lett. 10, 396 (1985).

18. "Nonlinear Optical Properties of Bi-Phenyl Compounds," M.J. Soileau, J. Tyminsky, N. Mansour, E. Canto, E.W. Van Stryland, S. Guha, E.J. Sharp, G.L. Wood, and J.L.W. Pohlmann, accepted for publication in Mol. Cryst. Liq. Cryst., 1986.

19. "Time Resolved Self-Defocusing in $Cd_{23}Hg_{77}Te$ and InSb", D. Craig, A. Miller, and M.J. Soileau, Opt. Lett. 11, 794 (1986).

20. "Two-Photon Absorption Induced Transmission Changes in ZnSe Interference Filters," E.W. Van Stryland, Steven A. Miller, B.S. Wherrett, and M.A. Woodall, JOSA B5, 1289 (1988).

21. "Self-Protecting Semiconductor Optical Limiters," D.J. Hagan, E.W. Van Stryland, M.J. Soileau, and Y.Y. Wu, Opt. Lett. 13, 315 (1988).

22. "Optical Limiting with Semiconductors," E.W. Van Stryland, Y.Y. Wu, D.J. Hagan, M.J. Soileau, and Kamjou Mansour, JOSAB Sept. (1988).

23. "The Effect of Linear Absorption on Self-Focusing", Mehrdad Mohebi, M.J. Soileau, and E.W. Van Stryland, Opt. Lett. 13, 758 (1988)

24. "Resolution of Discrepancies in Measurements of n_2 in CS_2 at 10 μm", Mehrdad Mohebi, M.J. Soileau, and E.W. Van Stryland, Opt. Lett. 13, 649 (1988).

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25. "The Effects of Self-Focusing on Laser-Induced Breakdown," William E. Williams, M.J. Soileau, and Eric W. Van Stryland, in Laser Induced Damage in Optical Materials, (1983).

26. "Simple Direct Measurements of n_2," William E. Williams, M.J. Soileau, and Eric W. Van Stryland, in Laser Induced Damage in Optical Materials, (1983).

27. "Nonlinear Absorption and Nonlinear Refraction Studies in MEBBA," M.J. Soileau, William E. Williams, E.W. Van Stryland, Shekhar Guha, H. Vanherzeele, J.L.W. Pohlmann, E.J. Sharp, and G.L. Wood, in Ultrafast Phenomena IV, eds. D. Auston, K.B. Eisenthal, Springer-Verlag, New York, 205 (1984).

28. "Self-Focusing in Damage Experiments Revisited," M.J. Soileau, William E. Williams, and Eric W. Van Stryland, in Laser Induced Damage in Optical Materials, NBS special publication 727, 394 (1984).

29. "Picosecond Damage in Y_2O_3 Stabilized Cubic Zirconia," N. Mansour, M.J. Soileau, and Eric W. Van Stryland, in Laser Induced Damage in Optical Materials, NBS special publication 727, 31 (1984).

30. "Two-Photon Absorption, Nonlinear Refraction, and Optical Limiting in Semiconductors," H. Vanherzeele, M.A. Woodall, Eric W. Van Stryland, M.J. Soileau, A.L. Smirl , S. Guha, N. Mansour, and M. Mohebi, in Laser Induced Damage in Optical Materials. NBS special publication 727, 404 (1984).

31. "Nonlinear Refractive Index of CS_2 at 10.6 μm," M. Mohebi, G.C. Reali, M.J. Soileau, and E.W. Van Stryland, SPIE 540, 528 (1985).

32. "Two-Photon Absorption, Nonlinear Refraction and Optical Limiting in Semiconductors", E.W. Van Stryland, Shekhar Guha, H. Vanherzeele, Mehrdad Mohebi, M.A. Woodall, M.J. Soileau, A.L. Smirl and T.F. Boggess, proceedings of the Fall meeting of the Materials Research Society, in Nonlinear Optical Materials, ed. D.A.B. Miller, 69, (1985).

33. "Measurement of n_2 in Liquids at 1.06 μm and 0.53 μm," Shekhar Guha, M.J. Soileau, and E.W. Van Stryland, Proceedings of the Southwest Conference on Optics, Albuquerque, NM SPIE 540, 533 (1985).

34. "Two-Photon Absorption and Nonlinear Refraction in Isotropic Liquid Crystals," M.J. Soileau, Eric W. Van Stryland, and Shekhar Guha, Proceedings of Conference 682, SPIE 30th Annual International Technical Symposium on Optical and Optoelectronic Applied Sciences and Engineering, (1986).

35. "Nonlinear Absorption Initiated Avalanche Breakdown in a Wide Gap Dielectric ZrO_2", Nastaran Mansour, Edesly Canto, E. Miesak, M.J. Soileau and Eric W. Van Stryland, Proceedings of the 1986 Boulder Damage Conference, Boulder, Colorado (1986).

36. "Nonlinear Transmission of Semiconductor Thin Films," Eric W. Van Stryland, S.A. Miller, and B.S. Wherrett, in Modern Optical Characterization Techniques for Semiconductors, O.J. Glembocki, Fred H. Pollak, J.J. Song, ed., SPIE 794, (1987).

37. "Temporal and Spatial Distribution of Light from a Monolithic Limiter", M.J. Soileau, D.J. Hagan, Y.Y. Wu, and E.W. Van Stryland, proceedings of the Joint U.S.-U.K. Conference on Sensor Protection, Malvern, England, 1987.

38. "Repetition Rate Dependence of the Nonlinear Response of Suspended Particles", M.J. Soileau, K. Mansour, E.W. Van Stryland, E. Canto, and D. J. Hagan, proceedings of the Joint U.S.-U.K. Conference on Sensor Protection, Malvern, England, 1987.

39. "Optical Damage Protector Based on Self-Defocusing in Semiconductors", E.W. Van Stryland, M.J. Soileau, D.J. Hagan and Y.Y. Wu, Proceedings of SPIE, O-E LASE 88, to be published, Los Angeles (1988).

40. "Observation of Two-Photon Absorption Prior to Laser-Induced Damage in ZrO_2", N. Mansour, K. Mansour, M.J. Soileau, E.W. Van Stryland, Proceedings of the 1987 Boulder Damage Conference, Boulder, Colorado (1987).

41. "Nonlinear Optics for Sensor Protection and Switching", E.W. Van Stryland, Proceedings of Southcon/88, Orlando, Fl., March (1988).

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1. "Picosecond Laser-Induced Damage", M.J. Soileau, Eric W. Van Stryland, Thomas F. Boggess, and Arthur L. Smirl, Lasers '83, San Francisco, CA., December 12-16, 1983.

2. "Laser Light Induced Bulk Damage to Optics", M.J. Soileau, E.W. Van Stryland and William E. Williams, Critical Review Paper, Southwest Conference on Optics, Albuquerque, NM., 1985.

3. "Passive Optical Switches with Picosecond Response", M.J. Soileau, S. Guha and E.W. Van Stryland, SPIE 1985 Los Angeles Technical Symposium on Optical and Electro-Optical Engineering, Los Angeles, CA., 1985.

4. "Passive Switching of Picosecond Light Pulses", M.J. Soileau, E.W. Van Stryland and S. Guha, Southwest Conference on Optics, Albuquerque, NM., 1985.

5. "Nonlinear Absorption and Associated Refraction in Semiconductors", E.W. Van Stryland, H. Vanherzeele, S. Guha, M.A. Woodall and M.J. Soileau, IV International Symposium, Ultrafast Phenomena in Spectroscopy, UPS 85, Reinhardsbrünn, GDR, October 23-26, 1985.

6. "Nonlinear Optical Properties of Liquid Crystals in the Isotropic Phase", M.J. Soileau, E.W. Van Stryland, S. Guha, E.J. Sharp, G.L. Wood and J.L.W. Pohlmann, Optical Properties of Liquid Crystals Conference, Naples, Italy, July 15-18, 1986.

7. "Nonlinear Beam Propagation in Semiconductors", E.W. Van Stryland, D.J. Hagan, M.J. Soileau and Y.Y. Wu, Lasers '87, Lake Tahoe, Nevada, Dec. 7-11, 1987.

8. "Influence of Self-Focusing on Bulk Laser Induced Damage", M.J. Soileau, and E.W. Van Stryland, SPIE, O-E LASE '88 Symposium on Lasers and Optics, Los Angeles, CA, 1988.

9. "Nonlinear Optical Properties of Modified Liquids", M.J. Soileau, E.W. Van Stryland, D.J. Hagan, and Kamjou Mansour, International Conference on Nonlinear Optics, Ashford Castle, Ireland, May 3-6, 1988.

10. "Role of Defects and Impurities in Bulk Laser-Induced Damage", M.J. Soileau, Nastaran Mansour, and E.W. Van Stryland, Annual Meeting of the Optical Society of America, Santa Clara, Ca., 1988.

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11. "Simple Direct Measurements of n_2," William E. Williams, M.J. Soileau, and Eric W. Van Stryland, 1983 Boulder Damage Conference, Boulder, CO, 1983.

12. "The Effect of Self-Focusing on Laser-Induced Breakdown," William E. Williams, M.J. Soileau, and Eric W. Van Stryland, 1983 Boulder Damage Conference, Boulder, CO, 1983.

13. "Measurements of the Nonlinear Refractive Index in Low Index Materials at 0.53 and 1.06 Microns," William E. Williams, M.J. Soileau, and Eric W. Van Stryland, 4th Conference on Lasers and Electro-Optics (CLEO), Anaheim, CA, 1984.

14. "Nonlinear Absorption and Nonlinear Refraction Studies in MEBBA," M.J. Soileau, William E. Williams, Eric W. Van Stryland, H. Vanherzeele, J.L.W. Pohlmann, E.J. Sharp, and G.L. Wood, Topical Meeting on Ultrafast Phenomena, Monterey, CA, 1984.

15. "Studies of Nonlinear Optical Properties in Liquid Crystals with Picosecond Pulses," M.J. Soileau, William E. Williams, E.W. Van Stryland, H. Vanherzeele, Shekhar Guha, J.L.W. Pohlmann, E.J. Sharp, and G.L. Wood, Tenth International Liquid Crystal Conference, York, U.K., 1984.

16. "Self-Focusing in Damage Experiments Revisited," M.J. Soileau, William E. Williams, and Eric W. Van Stryland, 1984 Boulder Damage Conference, Boulder, CO, 1984.

17. "Picosecond Damage in Y_2O_3 Stabilized Cubic Zirconia," N. Mansour, M.J. Soileau, and Eric W. Van Stryland, 1984 Boulder Damage Conference, Boulder, CO, 1984.

18. "Energy Gap Dependence of Two-Photon Absorption in Semiconductors," H. Vanherzeele, M.A. Woodall, Eric W. Van Stryland, M.J. Soileau, A.L. Smirl, Shekhar Guha, N. Mansour, and M. Mohebi, 1984 Boulder Damage Conference, Boulder, CO, 1984.

19. "Photoacoustic Measurement of Onset of Self-Focusing in Liquid CS_2 at 10.6 μm," M. Mohebi, P. Aiello, G.C. Reali, M.J. Soileau, and E.W. Van Stryland, Southwest Conference on Optics, Albuquerque, NM, 1985.

20. "Measurement of n_2 in Liquids at 1.06 μm and 0.53 μm," Shekhar Guha, M.J. Soileau, and E.W. Van Stryland, Southwest Conference on Optics, Albuquerque, NM, 1985.

21. "Two-Photon Absorption and Nonlinear Refraction in Semiconductors," Eric W. Van Stryland, S. Guha, H. Vanherzeele, M.A. Woodall, and M.J. Soileau, 7th National Quantum Electronics Conference, Malvern, England, 1985.

22. "Two-Photon Absorption, Nonlinear Refraction and Optical Limiting in Semiconductors," Eric W. Van Stryland, S. Guha, H. Vanherzeele, Mehrdad Mohebi, M.A. Woodall, M.J. Soileau, Arthur L. Smirl, and Thomas F. Boggess, 1985 Fall Meeting of the Materials Research Society, Boston, MA, 1985.

23. "Dynamic Nonlinear Properties of Dielectric Interference Filters," Y.T. Chow, B.T. McGuckin, B.S. Wherrett, and E. W. Van Stryland, The XIIIth International Quantum electronics Conference, San Francisco, CA, 1986.

24. "Nonlinear Optical Properties of BiPhenyl Compounds", M.J. Soileau, J. Tyminski, N. Mansour, E. Canto, E.W. Van Stryland, S. Guha, E.J. Sharp, G.L. Wood and J.L.W. Pohlmann, International Liquid Crystal Conference, San Francisco, CA.,1986.

25. "Self Protecting Solid State Optical Limiter," M.J. Soileau, D.J. Hagan, and E.W. Van Stryland, Conference on Lasers and Electro-Optics (CLEO), San Francisco, CA, 1986.

26. "Nonlinear Absorption Initiated Avalanche Breakdown in a Wide Gap Dielectric ZrO_2", Nastaran Mansour, Edesly Canto, E. Miesak, M.J. Soileau and Eric W. Van Stryland, 1986 Boulder Damage Conference, Boulder, CO, 1986.

27. "Fabry Perot Enhancement of Nonlinear Absorption in Optical Coatings," E.W. Van Stryland, Steve Miller and B.S. Wherrett, Conference on Lasers and Electro-Optics (CLEO), Baltimore, MD., 1987.

28. "Picosecond-Induced Dynamic Responses of Optically Bistable Interference Filters," Y.T. Chow, B.T. McGuckin, S.D. Smith, B.S. Wherrett, and Eric W. Van Stryland, Conference on Lasers and Electro-Optics (CLEO), Baltimore, Md., 1987.

29. "Characterization of Semiconductor Optical Power Limiting Devices", D.J. Hagan, M.J. Soileau, Y.Y. Wu, and E.W. Van Stryland, Conference on Lasers and Electro-Optics (CLEO), Baltimore, Md., 1987.

30. "Nonlinear Absorption-Induced Transmission Changes in ZnSe Interference Filters," D.J. Hagan, Y.Y. Wu, M.J. Soileau, E.W. Van Stryland, Y.T. Chow, and B.S. Wherrett, XV International Conference on Quantum Electronics (IQEC), Baltimore, Md., 1987.

31. "Temporal and Spatial Distribution of Light from a Monolithic Limiter", M.J. Soileau, D.J. Hagan, Y.Y. Wu, and E.W. Van Stryland, Joint U.S.-U.K. Conference on Sensor Protection, Malvern, England, 1987.

32. "Repetition Rate Dependence of the Nonlinear Response of Suspended Particles", M.J. Soileau, K. Mansour, E.W. Van Stryland, E. Canto, and D.J. Hagan, Joint U.S.-U.K. Conference on Sensor Protection, Malvern, England, 1987.

33. "Nonlinear Transmission of Semiconductor Thin Films", E.W. Van Stryland, Steven A. Miller and B.S. Wherrett, SPIE Symposium on Characterization of Very High Speed Semiconductor Devices and Integrated Circuits: Critical Review of Technology, Bay Point, FL, Mar 23 - 27, 1987.

34. "Optical Damage Protector Based on Self-Defocusing in Semiconductors", E.W. Van Stryland, M.J. Soileau, D.J. Hagan, and Y.Y. Wu, SPIE O-E/LASE '88 Symposium on Lasers and Optics, Los Angeles, CA, 1988.

35. "Picosecond Degenerate Four-Wave Mixing Studies in ZnSe", D.J. Hagan, E. Canto, E. Miesak, M.J. Soileau, and E.W. Van Stryland, Conference on Lasers and Electro-Optics (CLEO), CA., 1988.

36. "Two-Photon Absorption and Free-Carrier Absorption in Semiconductor Doped Glasses", E. Canto, E. Miesak, D.J. Hagan, M.J. Soileau, and E.W. Van Stryland, Conference on Lasers and Electro-Optics (CLEO), CA., 1988.

37. "Analysis of Semiconductor Based Optical Limiters", E.W. Van Stryland, Y.Y. Wu, D.J. Hagan, and M.J. Soileau, International Quantum Electronics Conference (IQEC), Tokyo, Japan, 1988.

38. "Optical Limiting with Semiconductors", E.W. Van Stryland, Y.Y. Wu, D.J. Hagan, M.J. Soileau, and K. Mansour, Topical Meeting on Laser Materials and Laser Spectroscopy, Shanghai, China, July 25-27, 1988.

Contributed

39. "The Role of Self-Focusing in Laser-Induced Breakdown of Selected Materials", William E. Williams, M.J. Soileau and E.W. Van Stryland, 1983 Annual Meeting of the Optical Society of America, New Orleans, LA., 1983.

40. "Pulsewidth Modulation Nonlinear Spectroscopy", E.W. Van Stryland and Milton A. Woodall, 1983 Annual Meeting of the Optical Society of America, New Orleans, LA., 1983.

41. "10.6 μm Optical Limiter", M.J. Soileau, E.W. Van Stryland, Mehrdad Mohebi and Nastaran Mansour, 1983 Annual Meeting of the Optical Society of America, New Orleans, LA., 1983.

42. "Spatial Beam Propagation of Picosecond Pulses Through Optical Limiting Media", Shekhar Guha, E.W. Van Stryland and M.J. Soileau, 1984 Annual Meeting of the Optical Society of America, San Diego, CA., 1984.

43. "Energy Gap Dependence of Two-Photon Absorption in Semiconductors", H. Vanherzeele, M.A. Woodall, E.W. Van Stryland, M.J. Soileau, Shekhar Guha, N. Mansour and M. Mohebi, 1984 Annual Meeting of the Optical Society of America, San Diego, CA., 1984.

44. "Irradiance Modulation Spectroscopy", Steven A. Miller, M.A. Woodall and E.W. Van Stryland, 1984 Annual Meeting of the Optical Society of America, San Diego, CA., 1984.

45. "Power Limiting and Self-Focusing in Nonlinearly Absorbing Liquid Crystalline Media", S. Guha, M. Mohebi, E.W. Van Stryland and M.J. Soileau, 1985 Annual Meeting of the Optical Society of America, Washington, DC, 1985.

46. "Picosecond Study of Charge Carrier Induced Nonlinear Refraction in Intermediate Band Gap Semiconductors", S. Guha, E.W. Van Stryland and M.J. Soileau, 1985 Annual Meeting of the Optical Society of America, Washington, DC, 1985.

47. "The Observation of Nonlinear Transmission Prior to Laser Damage in ZrO_2", N. Mansour, M.J. Soileau and E.W. Van Stryland, 1986 Annual Meeting of the Optical Society of America, Seattle, Washington, 1986.

48. "Semiconductor Optical Limiters with Large Dynamic Range", David J. Hagan, M.J. Soileau, Yuan-Yen Wu, and E.W. Van Stryland, 1986 Annual Meeting of the Optical Society of America, Seattle, Washington, 1986.

49. "Two-Photon Absorption and Nonlinear Refraction in Isotropic Liquid Crystals", M.J. Soileau, E.W. Van Stryland and Shekhar Guha, Conference 682, SPIE Optoelectronic Applied Sciences and Engineering, San Diego, Ca., August 1986.

50. "Dispersion of Nonlinear Index of Refraction of CS_2", M.Mohebi, M.J. Soileau, and E.W. Van Stryland, 1986 Annual Meeting of the Optical Society of America, Seattle, Washington, 1986.

51. "Color Center Formation by Two-Photon Absorption at 532 nm in Cubic Zirconia," N. Mansour, K. Mansour, M.S. Soileau and E.W. Van Stryland, Annual meeting of the Optical Society of America, Rochester, N.Y. 1987.

52. "Temporal and Spatial Response of Semiconductor Power Limiters", E.W. Van Stryland, M.J. Soileau, Y.Y. Wu, K. Mansour, and D.J. Hagan, Annual Meeting of the Optical Society of America, Rochester, N.Y., 1987.

53. "Nonlinear Optics for Sensor Protection and Switching", E.W. Van Stryland, Southcon 88, Orlando, Fl., March 1988.

54. "Nonlinear Transmission of Carbon Black in Suspension", Kamjou Mansour, E.W. Van Stryland, and M.J. Soileau, Annual Meeting of the Optical Society of America, Santa Clara, Ca., 1988.

55. "High Order Optical Nonlinearities in Semiconductors", E. Canto, J. Young, E. Miesak, E.W. Van Stryland, and D.J. Hagan, Annual Meeting of the Optical Society of America, Santa Clara, Ca., 1988.

56. "Thermal Lensing in CS_2 at 10 μm", Monsoor Sheik-Bahae, A. Said, E.W. Van Stryland, and M.J. Soileau, Annual Meeting of the Optical Society of America, Santa Clara, Ca., 1988.

Local Presentations at Optical Society Meetings

1. "Optical Limiting", D.J. Hagan, E.W. Van Stryland, Y.Y. Wu, and M.J. Soileau, Optical Society of America Florida Section Quarterly Meeting, Melbourne, Fl., 1987.

2. "Ultrafast Phase Conjugation in Semiconductors", E. Canto, D.J. Hagan, and E.W. Van Stryland, E. Miesak, and M.J. Soileau, Optical Society of America Florida Section Quarterly Meeting, Orlando, Fl., 1987.

III. SUMMARY OF PROGRESS

We have shown that nonlinear optical interactions are an effective means for passively controlling the spatial energy (or power) distribution of optical beams. Optical self-action (self-focusing and self-defocusing), multiphoton absorption, laser-induced breakdown or plasma formation, and various combinations of these nonlinear interactions have all been used to make optical power limiters (OPL) during this program. These devices can limit the focal plane irradiance (or fluence) of optical systems. Media investigated by us to date include Kerr liquids, liquid crystals, other organic liquids, semiconductors and liquid suspensions of solid particles. However, there remain several major problems that often prevent the effective utilization of nonlinear optical interactions for spatial control of optical beams. These include the lack of materials with (1) proper or desired nonlinear response, i.e., materials with large, broadband, fast nonlinearities; (2) materials with suitable characteristics in the various spectral regions of interest; and (3) materials with a large dynamic range. Here we define the dynamic range to be the irradiance or fluence region between where spatial beam control begins and the material fails due to irreversible laser-induced damage or breakdown. In the case of liquid OPL's that self-heal, the laser induced breakdown limit on the dynamic range is determined by damage to the cell window. In liquids it is also important to determine the self-healing rate. For example, we have observed large hysteresis effects in liquid/particle suspension based limiters even at a laser repetition rate of 1 Hz.

Figure 1 shows the ideal limiter response characteristics (solid line). The limiter has linear response at low input and constant output above a particular input which we call P_C. This could be input power or energy. The response of CS_2 in the limiting configuration depicted in Fig. 2 shows nearly ideal limiting, and in fact this system works remarkably well. However $P_C \simeq 8$kw for the input wavelength of 0.53μm. Note that Fig. 2 shows a defocusing nonlinearity characteristic of a semiconductor, while CS_2 has a self-focusing nonlinearity. For many applications

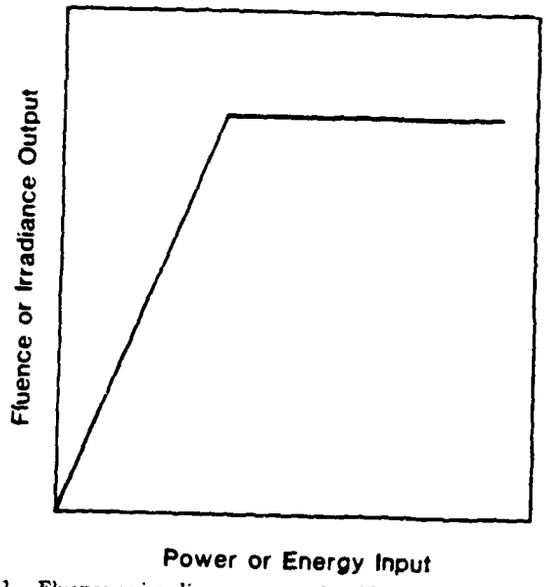


Fig. 1. Fluence or irradiance output of an ideal optical limiter as a function of the input power or energy.

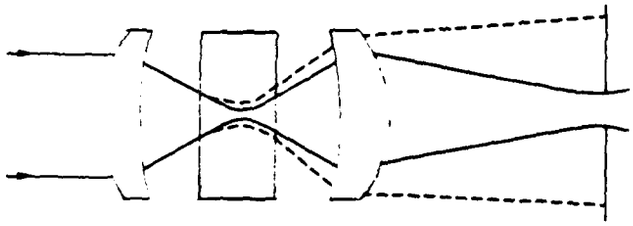


Fig. 2. Schematic drawing of the thick limiter geometry. The solid lines show linear beam propagation for low inputs, and the dashed lines show the beam for high inputs.

this P_c is too large. Because the mechanism in CS_2 is self-focusing from the optical Kerr effect, there is no way to lower P_c in CS_2. Another material must be found. We have found materials with somewhat smaller values of P_c, but often the characteristics of the "ideal" curve of Fig. 1 are sacrificed. Such materials problems are dependent on many parameters such as pulsewidth and wavelength. For that reason we separately considered the visible to near infrared region, and the infrared spectral region.

The 0.3 to 1.5 micron region has the advantage of having a large variety of materials for consideration. In addition, highly transparent solvents are readily available for use as vehicles for artificial Kerr media, solutions, liquid suspensions, and colloidal suspensions. Perhaps, more importantly, this spectral region is free from most vibrational resonances, such as those associated with hydrogen bonding. This means that many organic materials are highly transparent in this region. This is particularly appropriate in light of recent developments in the area of new organic materials with enhanced optical nonlinearities. These new organics have been demonstrated to have extremely high second order nonlinearities and large third order susceptibilities have also been reported. However, great care in the interpretation and extrapolation of published results toward use in optical limiters and other sensor protection schemes must be excercised. For example, the material MNA (methyl nitroanaline, an organic polymer in a solid host) was studied using nsec 4-wave mixing experiments. The reported χ^3 was several times the nonlinear susceptibility of CS_2. In separate measurements using femtosecond pulses, this material was reported to show a fast nonlinearity but the magnitude was not reported. We determined that the larger nsec signal was due to a linear absorption induced change in index, a slow thermal effect, and that the fast effect is presumably electronic. However, we have measured the magnitude of this nonlinearity to be no larger than one-tenth that of CS_2.

A standardized set of experiments to determine magnitudes, response times,

and physical mechanisms is necesssary in order to assess a material's potential for optical limiting. Under this program we have set up experimen.. and developed facilities to perform these measurements. We have also worked on the design of optical limiters to best utilize the measured nonlinearities.

This program has resulted in the development of extensive laboratory facilities for detailed, quantitative measurements of nonlinear absorption and nonlinear refraction in optical materials. Present capabilities cover the spectral range from the 10.6 μm region to the UV and the pulsewidth range from cw to femtoseconds. We have established formal collaborative arrangements with various material suppliers (e.g., CNVEO, Kent State's Liquid Crystal Institute, Purdue University, Hughes Research Labs, Battelle Columbus, etc.) and other groups studying nonlinear optical properties of materials. In 1985, Professor Van Stryland spent a sabbatical at Heriot-Watt University in Scotland (one of the world's leading institutions in this field) and we maintain active collaborations with that group. Dr. David Hagan of Heriot-Watt first joined the group at North Texas State University and has since joined the faculty at CREOL participating in this research. Also in December, 1988, Dr. Alan Miller of RSRE in Malvern, England will join CREOL. In addition, we have undertaken joint projects and exchanges with Dr. Viv Ropper's group at RSRE (the U.K.'s leading laboratory for optical power limiter research). This has been particularly fruitful in looking at nonlinearities in the IR of HgCdTe. In addition, they have recently grown, by molecular beam epitaxy, thin film structures of HgCdTe which we have been examining.

CREOL facilities and expertise were used as a test and evaluation facility to provide quantitative characterization of the nonlinear optical properties of new materials. This is helping to provide a standardized data base from which we can assess progress in materials development. Especially unique is our capability to determine absolute values of nonlinear coefficients since we can accurately determine optical irradiance levels. This latter point may seem trivial, however, errors in the

measurement of nonlinear parameters are nonlinearly dependent on the beam quality and errors in linear beam characterization. A notorious example of discrepancies between nonlinear material parameters reported in the literature are the values of β_2, the 2PA coefficient. The reported value for GaAs varies by nearly three orders of magnitude. Work in our laboratories has explained these discrepancies as due to (a) inadequate laser characterization and (b) not taking into account two other nonlinearities that can occur simultaneously with 2PA, i.e., free carrier absorption and free carrier refraction. Clearly, careful laser beam characterization is a necessary, but not sufficient condition for obtaining accurate nonlinear material parameters. The addition during this program of a psec 4-wave mixing apparatus for experiments at the Nd:YAG fundamental and harmonics adds greatly to our ability in determining the response times and nonlinear mechanisms in the various materials studied. Examples of the types of information that can be extracted from such an apparatus are given in Appendix A using bulk ZnSe as a sample. Varying beam polarization and varying the temporal delay of one of the 3 input beams at a time can allow an unraveling of the various nonlinear responses.

Our previous work in collaboration with researchers at RSRE has shown complicated transmission characteristics in HgCdTe that have been qualitatively associated with various orders of absorption (i.e., 1, 2, or 3 photon absorption). This work has led to the lowest limiting energy yet observed at 10.6 μm. A complete report of this work has been published by the British Ministry of Defense. Our contribution is included in the attatched reprints.

Mixtures, solutions, and suspensions which we refer to as combination media offer interesting possibilities as OPL materials. Research has shown that many highly nonlinear materials unfortunately have relatively high linear losses. Mixing approaches allow one to form artificial Kerr media and to use dilute solutions of materials which in their pure form would have too much linear absorption. In addition, our research has shown that the addition of a small amount of linear

absorber has little effect on the short pulse nonlinear response while greatly affecting the cw nonlinear response. The cw limiting occurs because of the slow and cumulative thermally induced refractive index changes. The cumulative thermal effect is why this type of nonlinearity is one of the largest measured to date. Thus, these approaches allow one to tailor the OPL performance to include response to cw as well as pulsed output.

Optical Damage:

The work on optical damage to solid materials led to the development of a solid state optical limiter. The work on understanding of damage mechanisms in solid materials including self-lensing is published in references 1, 2, 9, 25, 28, 29, 35, 40, and has been presented at conferences as listed under "Papers", reference numbers 1, 2, 8, 10, 12, 16, 17, 26, 39, 47 and 51.

Using the experimental apparatus for measuring 2PA in semiconductors with psec pulses we also monitored laser-induced damage to wide-gap insulators that were irradiated with γ-rays to produce mid-gap defect levels. We found that there is an excellent correlation between the damage thresholds and extrinsic linear absorption at twice the fundamental frequency, indicating 2PA from defects was initiating damage.

The knowledge gained in these damage studies helped in formulating a model for limiting in liquid suspensions of solid particles (ie. we found that damage in normally transparent materials began by linear or nonlinear absorption of defects or impurities which subsequently initiated an avalanche). We now find a similar process in particle suspensions.

In order to understand and interpret data a great deal of effort was focused on understanding nonlinear propogation in these solids. In particular, the role of self-focusing in damage experiments was determined. These studies have also helped our understanding of beam propagation in semiconductors needed to build and design optical limiters as discussed next.

Optical Limiting in Liquids (Visible and Near IR)

The results of research performed in this area are described in detail in publications 3, 4, 7, 10, 12, 18, 27, 33, and 34. This work was also presented at conferences as listed. See references under "Papers", numbers 3, 4, 6, 14, 15, 24, 42, 45, and 49.

The liquids studied included Kerr liquids, (e.g., CS_2), mixtures of Kerr liquids, liquid crystals above the Freedricks transition temperature (e.g., MEBBA, BUPBUB, etc.) and in BiPhenyl compounds. Limiting in Kerr liquids is now well understood and predictable. Unfortunately, CS_2 has the highest nonlinearity in conjunction with a fast response time (2 psec) and limiting still only begins at $\simeq$ 8kw at 532 nm (32 kw at 1.06 mm scaling as the wavelength squared). For most applications, this power is too high and it can only be raised not lowered.

We then studied a series of organic compounds that are liquid crystals at relatively low temperatures. Since they scatter light efficiently in the liquid crystal state and we were interested in transparent media with fast nonlinear response, we worked above the liquid crystal transition temperature.

We find that in liquid crystals 2PA is large but self-refraction is dominated by a relatively small self-focusing consistent with the absence of free carriers. Limiting was $\simeq$ 20 times less effective at 1.06 nm where the materials were not two-photon absorbers. While this result was disappointing, we feel that there is still a great deal of work to do in this area since engineering of the molecular structure and, therefore, nonlinearities are possible. Comparisons of nonlinearities observed in liquid crystals and semiconductors helps us to clearly separate effects due to self-refraction. This is particularly important in propagation through "thick" media where solutions to Maxwell's equations become numerically difficult. Appendix A gives recent theoretical calculations for limiting in thick media.

Optical Limiting in CS_2 at 10 um

Few liquids transmit at 10 μm (the CO_2 laser wavelength) and it appears that

solids, promising for limiting such as InSb, must be cooled to obtain high linear transmission. Solids also can suffer irreversible damage. The self-healing property of liquids makes them appealing for limiter applications. We found with nanosecond pulses that electrostriction plus thermal nonlinearities dominate the nonlinear response. These results are published in references 13, 17, 23, 24, 31, and have been presented at conferences as listed under "Papers" 19, 41, 50 and 56. We will present a talk at the 1988 OSA conference entitled "Thermal Lensing in CS_2 at 10 um" further describing the thermal nonlinearities in CS_2. While the first published results indicated a power of $\simeq$ 160 kw was needed to initiate limiting, recent results show that thermal nonlinearities can begin to limit as low as a few kilowatts with response faster than 50 nsec. The thermal nonlinearity is initiated by a small residual linear absorption. In order to understand how this absorption affected other nonlinearities we performed a separate study published in reference 23.

Optical Limiting with Semiconductors

During this program we succeeded in developing small, solid state, completely passive, fast response (psec) broadband optical limiters. Such limiters can be made in a monolithic form and are self-protected from optical damage. This overcomes the problem that made self-healing liquid limiters so attractive. We have succeeded in building devices that limit at $\simeq$300 Watts for 30 psec pulses and $\simeq$80 Watts for 12 nsec pulses. These devices should be extremely broadband and we are preparing to verify this prediction. This work has been published in reference numbers 5, 6, 8, 11, 15, 16, 21, 22, 30, 32, 37, 39, and 41. This work has been presented at conferences as listed under papers numbers 7, 22, 25, 29, 31, 34, 37, 38, 48, 52, and 53. Fundamental studies that led to the development of these semiconductor limiters are on two-photon absorption and self-defocusing by photogenerated carriers which are discussed in detail in reference 14, 19, and 26, as well as reviewed in references 5 and 22. These fundamental studies (Fig. 3) were

presented as listed under papers number 5, 11, 13, 18, 21, 35, 36, 40, 44, and 55.

The present device uses a combination of two-photon absorption and the associated self-defocusing in semiconductors. We use a geometry in which we tightly focus into the bulk of semiconductors as shown in Fig. 2. In this geometry the limitation of the device due to laser induced damage is eliminated. Additionally, the limiting can be made to occur at lower fluence levels.

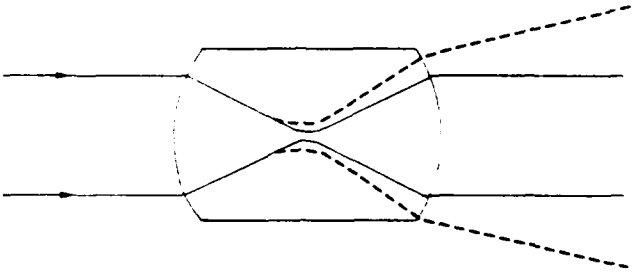


Fig. 3. Schematic of the monolithic optical limiter showing optical paths for low (solid lines) and high (dashed lines) inputs.

Fig. 3. Experimental configuration of "thick" optical power limiter.

The fluence at the surface remains low and two-photon absorption in the bulk lowers the irradiance as the beam focuses. Additionally, and more importantly, the excited carriers created in the bulk via two-photon absorption generate a nonlinear refraction of negative sign (Drude plus band-blocking). The resulting defocusing prevents the beam from reaching damaging irradiances. The beam paths for linear (solid) and nonlinear (dotted) propagation are shown schematically in Fig. 3.

The results of using this design with 30 psec 0.53 μm pulses and ZnSe as the active medium are shown in Fig. 4. Note the change of scale in Fig. 3. Limiting action is clearly seen for inputs of 10 nJ, i.e., the transmission is decreased by a

factor of 2. The output is clamped to a value below ~5 nJ for all inputs. The overall low input transmission including reflection losses is ~10% at the laser wavelength. For longer wavelengths the linear absorption should be less. We have tested the devices up to 1/2 mJ inputs without bulk damage (greater than 4 orders of magnitude dynamic range). The device is only limited by damage to the front surface of the semiconductor which can be made "arbritarily" high by increasing the thickness and aperture. Additionally, since the two-photon absorption coefficient in semiconductors is nearly constant over a wide range of frequencies above the gap, the optical limiter is a broad band device.

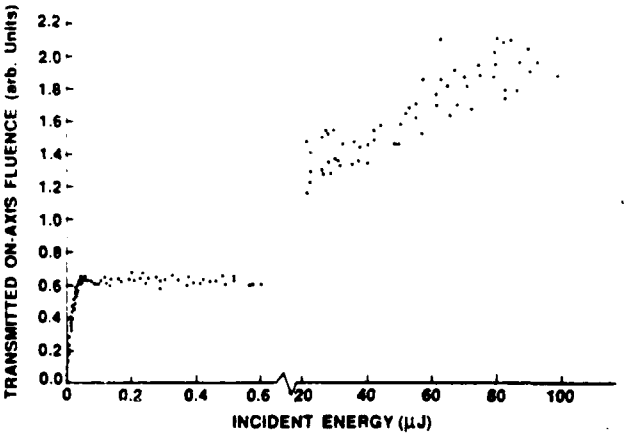


Fig. 4. Plot of the output of a monolithic ZnSe limiter (interpreted as the on-axis fluence detected through an aperture) as a function of the input energy of 30-psec FWHM, 0.53-μm pulses.

Fig. 4. Transmission versus input energy of 0.53 μm 30 psec pulses through the "thick" limiter configuration of Fig.2 using ZnSe

We are also performing a fundamental study of nonlinearities in these semiconductors using picosecond degenerate four-wave-mixing as discussed in reference 35 and 55 under "Papers" and discussed in more detail in appendix B. We are applying this technique to other nonlinear materials.

One problem remaining to be solved for the solid state limiter is that for long pulses thermal lensing can dominate and lead to damage. Materials with a negative thermal lensing are currently under investigation.

Fundamental Study of Two-Photon Absorption

We have developed a predictive capability for determining the two-photon absorption coefficients in semiconductors given only a few materials parameters. Our conclusion from a systematic study of ten different semiconductors is that near-intrinsic values of the two-photon absorption coefficient β_2 can be determined with the proper experimental technique for relatively pure samples. Also, both the scaling laws for β_2 and the absolute values of β_2 predicted by theory are confirmed by our experiments. We find our data is consistent with:

$$\beta_2(\text{cm/GW}) = (3.1 \pm 0.5) \times 10^3 \, E_p \, F_2 \, (2\hbar\omega/E_g) \, n^{-2} \, E_g^{-3}, \quad \ldots\ldots\ldots\ldots \quad (1)$$

where n is the refractive index, E_g is the band-gap energy and $F_2(x)=(x-1)^{3/2}/x^5$. E_P is nearly constant (21 eV) for the materials studied and is given by $E_P = 2P^2m/\hbar^2$, where P is the Kane momentum parameter and m is the electron mass. This relation can now be used to predict β_2 for other materials at other wavelengths. For example, if we extend our result to 2PA in InSb at 10.6 μm (300 K), we obtain a value of $\beta_2 = 6.8$ cm$(\text{MW})^{-1}$. This value is in excellent agreement with the value of 8 cm $(\text{MW})^{-1}$ measured previously. Given this relation, we can now look for deviations from the predicted behavior as a function of various extrinsic effects such as doping levels. It is hoped that the addition of near resonant intermidiate states by doping will enhance 2PA and thus, lower limiting thresholds.

The scaling law Eq. 1 holds for the ten different semiconductors studied except for the ZnTe where two photons couple states only 3 percent above the band

edge. Here it is expected that exciton effects or impurity effects might become important, and indeed the measured value of β_2 is considerably larger than predicted by Eq. 1. In addition, as $2\hbar\omega/E_g$ approaches unity, the allowed-forbidden transitions upon which Eq. 1 is based become small (as evidenced by F_2) and eventually this leads to allowed-allowed transitions becoming dominant. We should note here, however, that using photoconductivity techniques on GaAs we find the spectral dependence just above the gap obeys Eq. 1 which neglects exciton enhancement and allowed-allowed transitions. We plan to study this discrepancey in the future.

Figure 5 shows the data and Eq. 1 plotted to emphasize the E_g^{-3} dependence of β_2. The value of the constant 3.1 x 10^3 was determined by performing a least-squares fit of the data, excluding ZnTe, to the parametric dependence of Eq. 1. If a more sophisitcated band structure, including the split-off bands and band nonparabolicity is included, the absolute values are on average within 26 percent of the calculated values. Although the algebraic expression for F_2 becomes quite complicated, the parametric dependencies remain unchanged.

The agreement with theory is remarkable in light of previous results, especially when it is noted that the samples are different semiconductors having different structures. It shows that while the true band structure is quite complex, the intermediate states that are most important can be accurately modeled with a simple band structure. In fact the two-band model predictions (see Eq. (1)) are quite adequate. This is a very fortuitous result since it means that refinements in band structures do not lead to large changes in β_2, nor to changes in scaling. The important question raised for the future is: can the 2PA be significantly changed by the addition of impurities (mid-gap levels), structural changes, etc.?

In addition to the work presented on nonlinear absorption, a good deal of effort went into the study of nonlinear refraction since it turned out that a major source of error in the determination of 2PA coefficients was due to nonlinear beam propagation effects caused by self-lensing. In short, both the 2PA and associated

self-defocusing are quantitatively understood in semiconductors. Again, this will allow prediction of nonlinearities (both absorptive and refractive), useful in the design of nonlinear optical devices. such as optical power limiters.

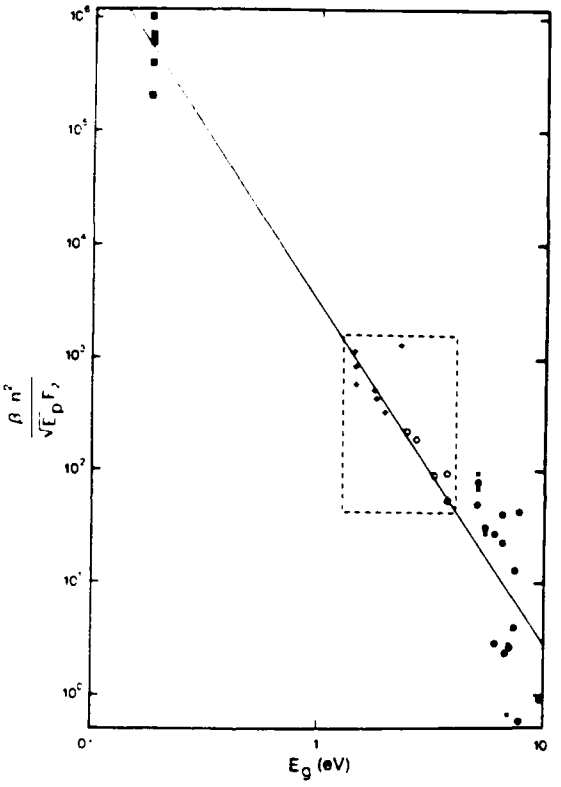


Figure 5. A log-plot of the scaled two-photon absorption coefficient versus bandgap energy. The solid line is given by equation (1) and has slope -3.

Novel Limiting Schemes

We have been investigating alternative methods for passive optical limiting by using the field enhancement that occurs in interference filters (Fabry-Perot's). While this work is still in its infancy, some interesting results are published in

references 20 and 36 and have been presented as listed under "Papers", references 23, 27, 28, 30, and 33. We have found $\simeq 10^3$ enhancement of two-photon absorption within the spacer layer of the interference filter. However, the "thinness" of the layer still prevents substantial changes in transmission.

We have developed the pulse delay modulation technique using shot-noise-limited detection for these interference filter 2PA measurements. This method lends itself well to the study of optical nonlinearities of thin films. The use of thin films as nonlinear elements in optical devices is gaining considerable attention as interest in optical computing grows. The operation of such thin film devices is critically dependent on nonlinear material used for the film. We have developed a sensitive method for monitoring a nonlinearly induced transmission change that is particularly useful for observation of fast nonlinearities in thin film structures. This technique allows assessment of the nonlinearity using a beam oriented perpendicular to the fulm surface rather than in the guided wave geometry.

Using this technique, we have observed transmission changes of a probe beam induced by two-photon absorption (2PA), of picosecond 0.6 μm dye laser pulses in a 0.4 μm thick ZnSe film sandwiched between reflective coatings. These Fabry-Perot structures have been used with cw lasers to demonstrate bistability induced by the slow thermal refractive index change. In order to observe 2PA in thin films, we have utilized two separate methods to enhance the signal-to-noise ratio. A dual modulation scheme enhances the sensitivity for observing small transmission changes while discriminating against the large thermal nonlinearities. The Fabry-Perot cavity enhances the irradiance, thus directly increasing the transmission change. The results of calculations of the 2PA enhancement in a Fabry-Perot are consistent with experimental results, and indicate that three orders of magnitude enhancement of 2PA is readily obtainable. Combining these two methods, we are able to detect signals so small that they correspond to a calculated single pass change in transmission of $\sim 10^{-10}$.

In addition, we are studying possible applications of the nonlinearities present in semiconductor doped glasses for optical limiting. See reference 36 under "Papers."

Optical Limiting With Liquid Suspensions of Carbon Black

We have begun an intensive investigation of the limiting properties of a suspension of carbon particles in liquid (SCP). We have developed a preliminary explanation of the optical limiting characterizations of these suspensions. We find that the limiting depends primarily on the input optical fluence $\left(\frac{J}{cm^2}\right)$ rather than irradiance $\left(\frac{W}{cm^2}\right)$. Therefore, limiting works well for long pulses ($\geq$10 nsec) but is less effective for short pulses (~ psec). Additionally, the SCP rapidly degrades with repetitive laser firings. Thus, flowing or moving the liquid between firings is necessary. Portions of this work have been published in reference 38 and presented in papers number 9, 32, and 54.

We have monitored transmission, optical scattering, and the photoacoustic response of the SCP simultaneously. We observe nonlinearities in all three signals as displayed in Fig. 6. These data have led us to the conclusion that the carbon first linearly absorbs the input light very efficiently (carbon is a good absorber). The carbon is rapidly heated, vaporizes and ionizes to form a rapidly expanding microplasma. This plasma absorbs and scatters subsequent light, thus limiting the transmission. Note, that the volumetric expansion of the plasma is necessary as the volume percent of carbon is small and they are already highly absorbing. The nonlinear scattered light which appears to be the dominant nonlinearity is evidence for the existence of these plasmas.

Fig. 7 shows the scattered signal (■ 's) transmittance (x's) and absorption for a fixed input energy of 1.2 mJ for each 12 nsec (FWHM) laser pulse at 1.06 μm. The time between pulses was 0.1 sec. We see the transmission increases by approximately a factor of 2, the absorption (from the photoacoustic signal) decreases by a factor of ~2, while the initially high scattered light signal decreases by more than a factor of 40. This shows that the nonlinear response for this input is

dominated by nonlinear scattering. The decrease in nonlinearity seen in all three signals shows that the laser energy is causing a physical change in the sample (i.e., of the carbon particles since no nonlinearity is seen in the liquid by itself). Thus, in essence, we are performing a laser induced damage experiment, and we have prepared a material with an extremely low damage threshold. This is consistent with having high linear absorption by the very small carbon particles (~350A). By waiting 30 seconds between laser firings, we see little or no change in nonlinearity after repeated irradiations as shown in Fig. 7. This simply indicates that normal convection and diffusion within the liquid replenishes the irradiated carbon by unirradiated carbon within the beam spot size of ~125 μm $\left[\text{HW } \frac{1}{e^2\text{M}}\right]$. We have also found similar results by flowing the liquid.

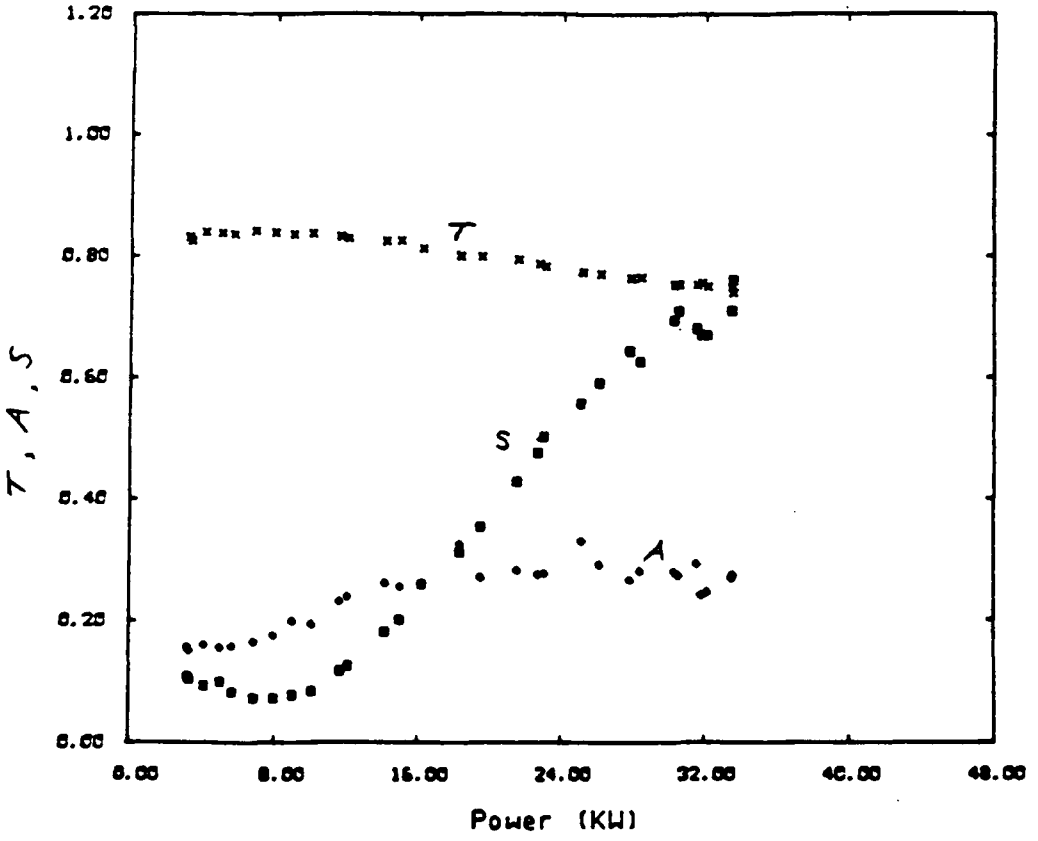
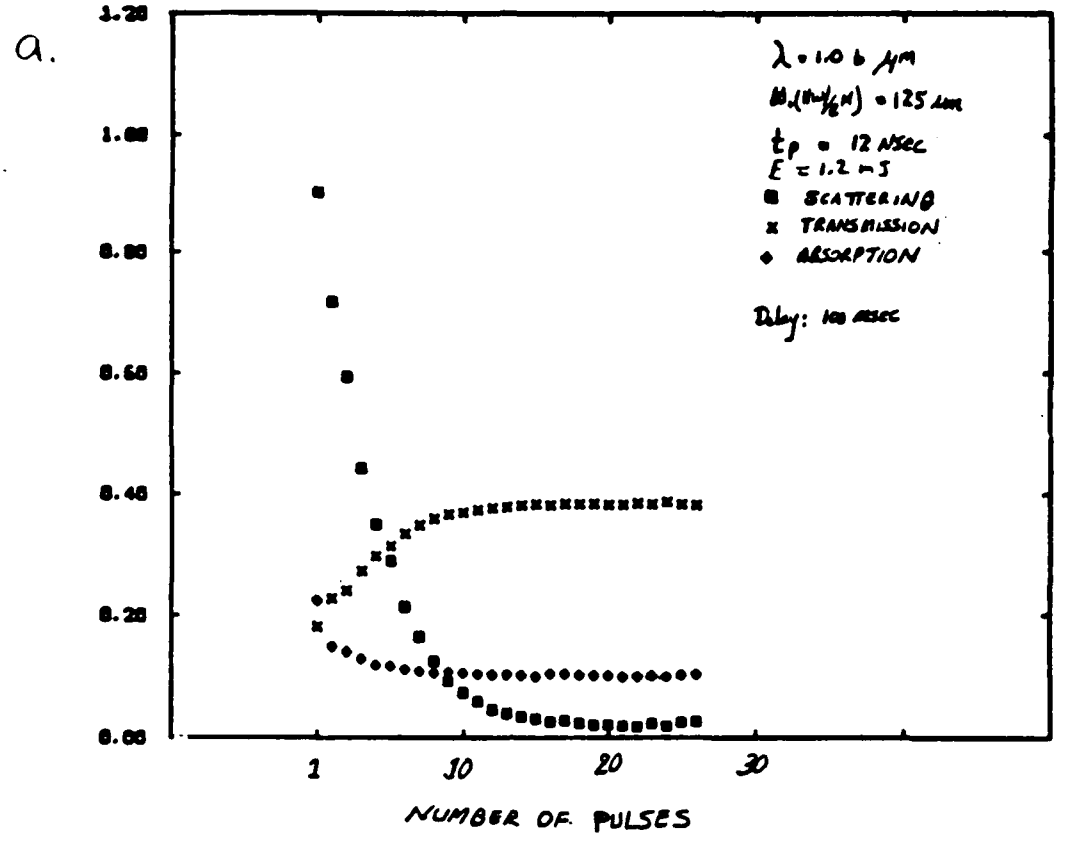


Fig. 6. Transmittance x, absorbance....◆...., and scattering....■.... versus input power for 1.06 μm, 12 nsec (FWHM) pulses focused to 125 μm $\left[\text{HW } \frac{1}{e} \mu\right]$.



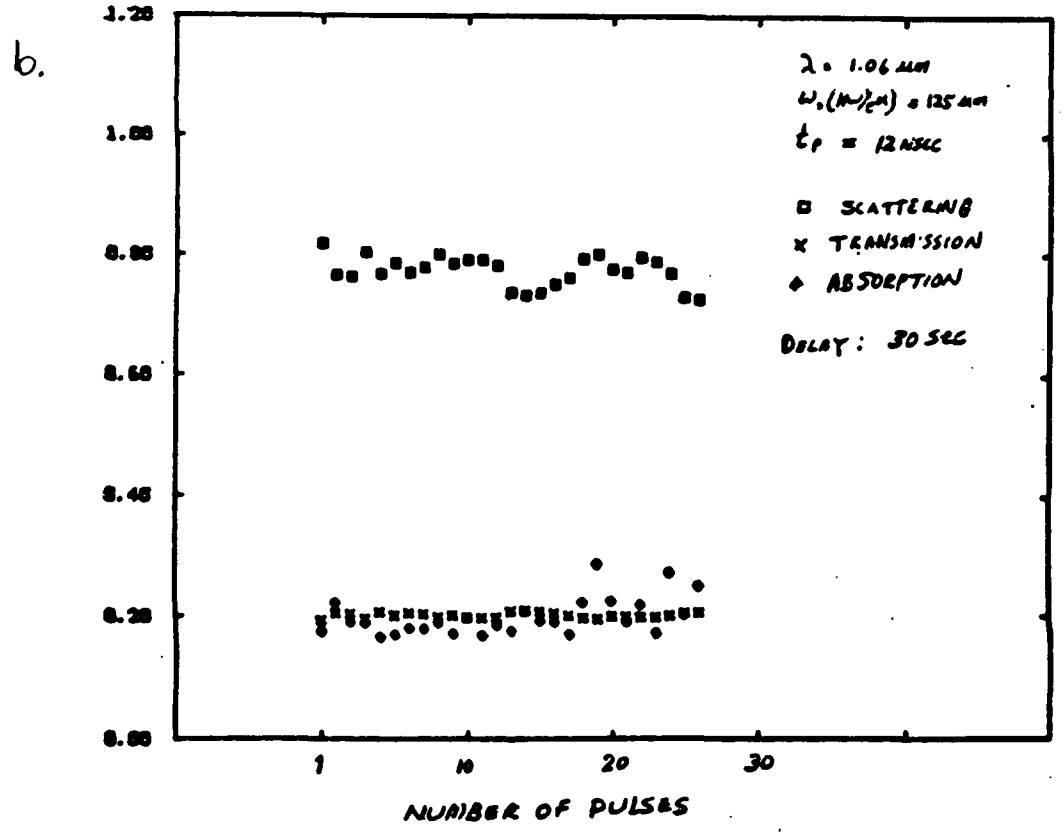


Fig. 7 Simultaneous measurement of transmission, absorption and scattering of the carbon suspended liquid for fixed input energy as a function of number shots for .1 sec (a.) and 30 sec (b.) time delay between input pulses

Additionally, the photoacoustic experiments show an increase in absorption at the onset of limiting, which quickly saturates with higher irradiance to a new, higher absorption level. This is shown in Fig. 6 along with the side scattered light which shows an increase in scatter at the onset of limiting which continues to increase nonlinearly with increasing power. Note that these data are obtained at low input levels where only small changes in transmission are observed. Fig. 6 clearly shows the dominance of scattering. The decrease in transmission at high powers is due almost exclusively to nonlinear scattering since the absorption ceases to increase. This increased scattering agrees with what is expected from expanding microplasms.

Initially, the scattering contribution to the overall transmittance is only a few percent for the small particle sizes used. All data shown are for a 70 transmitting 1 cm long cell. Mie theory predicts increased scattering with increasing particle size, but Mie theory can only qualitatively describe the optical properties of carbon black since it consists of irregularly shaped agglomerated particle clumps. In our experiments we filtered the dispersion through a $\frac{1}{4}$ μm filter and the largest agglomerates seen were 0.21 μm with an individual particle size of ~35 nm.

A light induced microplasma expands rapidly and the scattering increases rapidly with size. Why the absorption doesn't continue to increase is less clear. However, the original high absorption of the carbon particle will not continue since the surrounding plasma will scatter the light before it reaches what is left of the carbon. The plasma, of course, will both scatter and absorb. The relative values depend on size and charge density. The data are insufficient to estimate either the size or density at this time.

We can estimate the temperature rise of the carbon paticles at the limiting threshold given the laser beam parameters and optical properties of the carbon. Ravey et al gives the complex index of carbon black as $\tilde{n} = 1.96 - i\ 0.66$ which gives an absorption coefficient of $\simeq 3 \times 10^4$ cm^{-1}. Donnet and Voet in "Carbon

Black", pub. Marcel Dekker, 1976, show that the variation of absorption with the different types (sizes, etc.) of carbon particles varies by factors of no more than 2 to 3 and scales linearly with concentration. It is also observed that the optical properties of a carbon black dispersion in a liquid behave similarly to those of the powder form. This is true since the powder form generally has a void volume of over 90 so that it may be considered a dispersion in air. This, in turn, implies that the absorption which must be used to calculate a temperature rise for a single small particle is more than an order of magnitude larger than the reported value (i.e., $3x10^5 cm^{-1}$).

If we use the value of $\alpha \sim 3x10^5\ cm^{-1}$ and assume single carbon particles (350 A diameter) with no thermal diffusion, we calculate a temperature increase of $\sim 10^5$ °C using 12 nsec, 20 $\frac{MW}{cm^2}$ pulses at threshold. Thermal diffusion lowers this temperature increase by ~ 10. 10,000 degrees is enough to give thermionic emission and create carriers to start an avalanche. The irradiance of 20 $\frac{MW}{cm^2}$ gives a field of $\sim 10^5\ \frac{V}{cm}$ for the avalanche ionization process.

While the evidence obtained is insufficient to give details of this damage process, we feel that the basic underlying physical mechanisms are reasonably well understood. We note that the broadband nature of the nonlinearity is primarily due to the fact that carbon is black, i.e., it absorbs all wavelengths. Unfortunately, this also doesn't leave a lot of room for improving the performance of optical limiters based on this nonlinearity, since we can't start with larger linear absorption, and going to smaller particle sizes doesn't help since thermal diffusion is already significant.

III 1a. Abstract of Dissertation

Woodall, Milton A., Nonlinear Absorption Techniques and Measurements in Semiconductors. Doctor of Philosophy (Physics), August, 1985, 290 pp., 7 tables, 56 illustrations, bibliography, 95 titles.

We have conducted a detailed experimental and theoretical study of nonlinear absorption in semiconductors. Experimental measurements were made on a variety of materials at wavelengths of 2.06 and 0.53 microns using a picosecond Nd:YAG laser. Both two- and three-photon processes were investigated. Values of nonlinear absorption coefficients extracted from these measurements show excellent agreement with recent theory and scaling rules.

Our theoretical investigation has been carried out for two-, three-, and n-photon absorption, for both continuous and pulsed sources. Expressions are obtained for the transmission of the sample in terms of the incident irradiance for each case. The physical interpretation of these results is discussed.

We have also considered the effects of the photogenerated carriers on the measurements. Equations are developed that include linear absorption by these carriers. We have observed severe distortions on the transmitted beam, caused by changes in the refractive index of the material, due to the presence of these carriers. We present a model that accurately describes these effects in terms of the photo-generated carrier density.

We have developed several novel techniques for monitoring nonlinear absorption. In particular, we have adapted the photoacoustic technique to the measurement of nonlinear absorption in semiconductors. We have also developed a technique employing irradiance modulation to greatly enhance the sensitivity to nonlinear processes and simultaneously discriminate against linear background signals. A related technique has been used to observe coherent mixing effects in semiconductors with cw, modelocked dye lasers.

III 1b. Scientific Collaborators

Principal Investigator: M.J. Soileau, Professor, University of Central Florida; Adjunct Professor, North Texas State University

Principal Investigator: E.W. Van Stryland, Professor, University of Central Florida; Adjunct Professor, North Texas State University

Research Associate: D.J Hagan, Assisstant Professor, CREOL, University of Central Florida

Research Associate: H.J. Mackey, Professor, North Texas State University

PhD Research Scientists: M. Sheik-Bahae, M. Junnarkar, J. Young

Graduate Students: M.A. Woodall (currently graduated with PhD), Steven A. Miller (dissertation in progress), Edesly Canto, Y.Y. Wu, A. Said, K. Mansour, T. Wei, C.B. Park

In addition to the researchers listed above a number of collaborators woking at other institutions have contributed to this effort including; Dr. Alan Miller RSRE and his associates , researchers at Kent State's Liquid Crystal Institute, the Jet Propulsion Laboratory, Mcdonnell Douglas and Battelle. We have also greatly benefitted from our interactions with the researchers at CNVEO including Dr. E. Sharp, Dr. J. Pohlmann and G. Wood.

APPENDIX A

Analysis of "thick" optical limiters

Our previous experimental work on optical power limiting has shown that some of the the most effective limiters are those in which defocusing (or focusing) occurs inside the limiting medium itself. These are termed "thick" optical limiters, due to the long propagation paths involved. However, so far we have only been able to do useful theoretical analysis on thin limiters.

A rigorous and exact theoretical analysis of the wave propagation for thick limiters is computationally cumbersome and extremely time consuming when the thickness of the nonlinear medium is comparable to or larger than the Rayleigh range of the focussed beam ($L > Z_0$). A simple and efficient model, presented here, eases this task by regarding the nonlinear medium as a stack of nonlinear lenses whose focal lengths depend on the local beam irradiance (see Figure 1-b). In particular for a Kerr effect (nonlinear index γ) where the index of refraction change $\Delta n(I) = \gamma I$, or for a thermal nonlinearity (nonlinear index η) where $\Delta n(I) = \eta \int I dt$, one obtains an equivalent focal length f_i for the i-th element as:

$$f_i = \frac{a\,\omega^2_i}{4\,\Delta n(I)\,\Delta l} \qquad \text{for} \quad 3 < a < 5$$

where Δl is the thickness of each element, and ω_i is the laser beam waist at the i'th element. This is based on the Gaussian-parabolic approximation of $1-e^{-x^2} \simeq \frac{x^2}{a}$ which appears works quite well under our experimental conditions. The ABCD matrix of the system is then obtained as:

$$\begin{pmatrix} A & B \\ C & D \end{pmatrix} = \begin{pmatrix} 1 & D_2 \\ 0 & 0 \end{pmatrix} x \begin{pmatrix} 1 - \frac{\Delta l}{f_i} & \Delta l \\ -\frac{1}{f_i} & 1 \end{pmatrix} \ldots x \ldots \begin{pmatrix} 1-\frac{\Delta l}{f_2} & \Delta l \\ -\frac{1}{f_2} & 1 \end{pmatrix}$$

$$x \begin{pmatrix} 1-\frac{\Delta l}{f_1} & \Delta l \\ -\frac{1}{f_1} & 1 \end{pmatrix} x \begin{pmatrix} 1 & D_1 \\ 0 & 1 \end{pmatrix} x \begin{pmatrix} 1 & 0 \\ -\frac{1}{f} & 1 \end{pmatrix}$$

where D_1=f-Z-L/2 and D_2=D+Z-L/2.

The essential features of such an optical limiter are fairly predictable by applying this model. For example, a comparison of calculated results and the measured nonlinear refraction in CS_2, (see Figure 2) is indicative of a theromo-optic coefficient $\simeq -1 \times 10^{-3\ o} C^{-1}$ in excellent agreement with the previously reported value.

In general the calculated results point out a number of important considerations in designing an OPL device:

1. For a negative nonlinearity (such as thermal), the lowest limiting threshold is obtained by focussing at the first surface of the NL medium.
2. For a positive nonlinearity (e.g. electrostriction, Kerr effect) this is achieved by focussing at the second surface.
3. For a given focal length, increasing the sample length above the Rayleigh range would not increase the effective nonlinear refraction.
4. For a thin sample ($L << Z_0$), maximum limiting occurs when the sample is placed at a distance $\simeq \frac{Zo}{2}$ before or after the focus depending on the sign of the nonlinearity.

In conclusion, we have been able to reduce the wave propagation problem in a "thick" nonlinear index material to a simple ABCD matrix formalism. This algorithm can be applied to study the behavior of an optical limiting device based on nonlinear refraction. The computational efficiency and qualitative accuracy of this model are found to be quite satisfactory.

APPENDIX B

Here we show the geometry used in our psec 4-wave mixing experiments and data taken using bulk polycrystalline ZnSe. In Figure B1 is shown the three separate polarization combinations for the probe P, and forward F and backward B pump beams. Performing experiments in these different combinations and varying the temporal delay of the three input beams yeilds different information on the nonlinear interaction responsible for the production of the phase-conjugate signal traveling backward along the probe beam (the beam going to the right at an angle in Fig. B1). Figures B2, 3 and 4 show the various polarization configurations used in ZnSe and, in addition, show the variation in signal with backward pump beam temporal delay. These data indicate that for short times, a fast χ^3 process is dominant while for longer times, the cummulative effects of free carrier refraction become dominant.

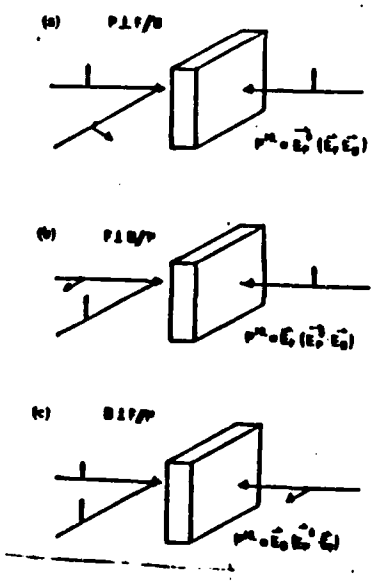


Figure B-1 Polarization selection of four-wave mixing phase-conjugate source terms. (a) yields only the two-photon coherence term, while (b) and (c) give the short and large period grating terms, respectively.

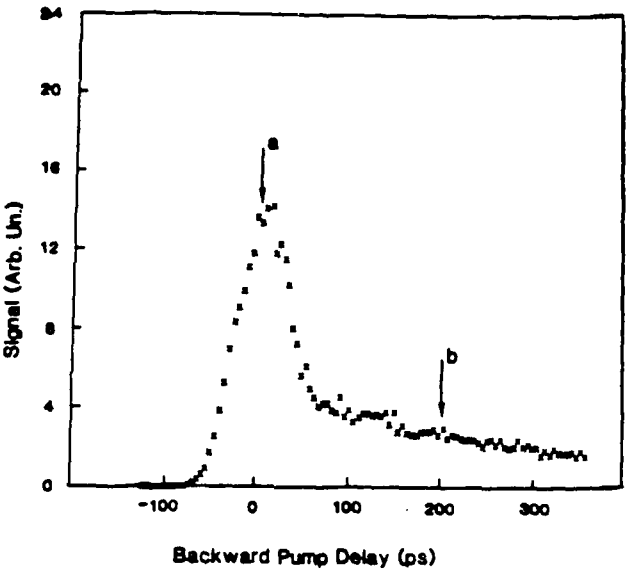


Figure B-2 Phase-conjugate signal of 532 nm light from ZnSe versus backward pump pulse delay. The peak shows a cubic power law, indicating a fast χ^3 process, while the 'tail' at -100 to -200 ps shows a fifth power dependence, indicating refraction due to carriers excited by two-photon absorption. All beams have parallel polarization.

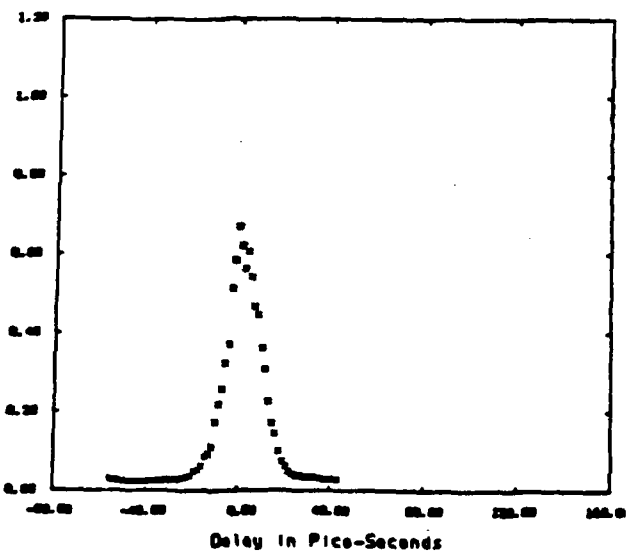


Figure B-3 As figure B-2, but with forward pump polarized perpendicular to the other two beams, thus eliminating the large period grating. The short period grating decays rapidly by diffusion.

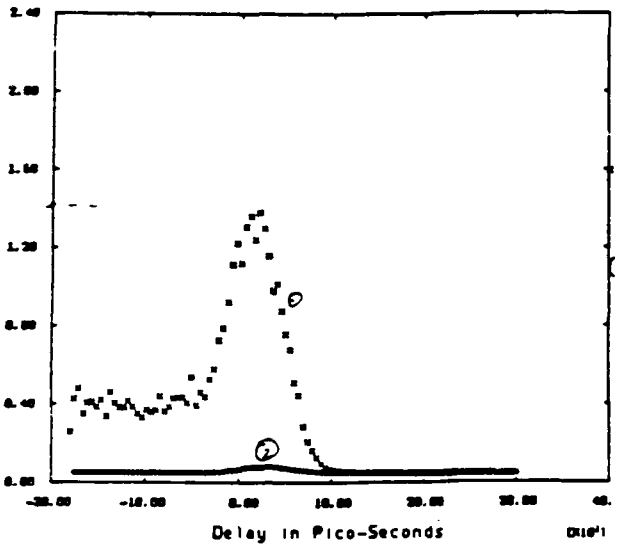


Figure B-4 As figure B-3, but with backward pump (crosses) or probe (diamonds) polarized perpendicular showing only a small two-photon coherence.

The work performed dealt with development of techniques for, and measurement of, two-photon absorption (2PA) and associated nonlinearities, as well as applications of these nonlinearities to the design of optical devices. A technique was developed that is extremely sensitive to nonlinear absorption induced changes in transmission that is even applicable to the study of 2PA in thin films (e.g., optical coatings). A systematic study of 2PA in ten different semiconductors led to verification of a simple 2-band model which allows prediction of 2PA magnitudes in other materials given only a few material parameters. These experiments also clarified the reasons for previous disagreements appearing in the literature as to the magnitudes of 2PA coefficients. These studies showed the importance of nonlinear refraction by 2PA generated carriers even under circumstances where the free-carrier absorption was negligible. This led to the development of passive optical limiting devices that have applications in sensor protection. Such devices have high linear transmission for low input power or energy and have low transmission at high inputs such that above some threshold input, the output fluence or irradiance is clamped. We have designed and tested one such device that has a limiting input

energy of 12 nJ (300W) for 30 picosecond 0.53 μm input pulses and 80 Watts for 18 nsec input pulsewidths.