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1. AGENCY USE ONLY (Leave blank)		2. REPORT DATE 16 Sep 93	3. REPORT TYPE AND DATES COVERED FINAL 1 Nov 89 - 28 Feb 93	
4. TITLE AND SUBTITLE MOLECULAR NONLINEAR OPTICAL SUSCEPTIBILITIES IN CONDENSED PHASES			5. FUNDING NUMBERS 61102F 2303/B3	
6. AUTHOR(S) Prof Shaul Mukamel			7. PERFORMING ORGANIZATION REPORT NUMBER AFOSR-TR-93 0708	
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) University of Rochester Department of Chemistry Rochester, New York 14627-0216			8. PERFORMING ORGANIZATION REPORT NUMBER	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) AFOSR/NC 110 DUNCAN AVENUE SUITE B115 BOLLING AFB DC 20332-0001			10. SPONSORING/MONITORING AGENCY REPORT NUMBER AFOSR-90-005	
11. SUPPLEMENTARY NOTES				
12a. DISTRIBUTION/AVAILABILITY STATEMENT APPROVED FOR PUBLIC RELEASE DISTRIBUTION IS UNLIMITED			12b. DISTRIBUTION CODE	
13. ABSTRACT (Maximum 200 words) A microscopic theory for the nonlinear optical response and susceptibilities of molecular materials with localized and delocalized electronic states (e.g. molecular assemblies, conjugated polymers, aggregates and monolayers) was developed. The theory, which properly accounts for many-body effects, is based on the derivation of coupled reduced equations of motion for the material variables, which determine the optical response. The present formulation maps the calculation of optical nonlinearities onto solving the dynamics of coupled nonlinear oscillators and overcomes the difficulties associated with the local-field approximation. An operational definition of the nonlinear coherence-size N_c , which controls the cooperative enhancement of the optical response is developed. A real-space representation of the optical response of conjugated polyenes is developed by using the Wannier representation to derive equations of motion for coupled two-site oscillators representing correlated electron-hole pairs. The resulting elementary				
14. SUBJECT TERMS			15. NUMBER OF PAGES 10	
17. SECURITY CLASSIFICATION OF REPORT UNCLASSIFIED			16. PRICE CODE	
18. SECURITY CLASSIFICATION OF THIS PAGE UNCLASSIFIED		19. SECURITY CLASSIFICATION OF ABSTRACT UNCLASSIFIED		20. LIMITATION OF ABSTRACT

DTIC
OCT 05 1993

93-23116
208

NSN 7540-01-280-5500

93 10 1 2 2 7

Standard Form 298 (Rev. 2-89)
Prescribed by ANSI Std. Z39-18
298-102

Molecular Nonlinear Optical Susceptibilities in Condensed Phases Final Technical Report

submitted to the
Air Force Office of Scientific Research

Grant: AFOSR-90-0054

Period 11/1/89 to 10/31/92

Summary

A microscopic theory for the nonlinear optical response and susceptibilities of molecular materials with localized and delocalized electronic states (e.g. molecular assemblies, conjugated polymers, aggregates and monolayers) was developed. The theory, which properly accounts for many-body effects, is based on the derivation of coupled reduced equations of motion for the material variables, which determine the optical response. The present formulation maps the calculation of optical nonlinearities onto solving the dynamics of coupled nonlinear oscillators and overcomes the difficulties associated with the local-field approximation. An operational definition of the nonlinear coherence-size N_c , which controls the cooperative enhancement of the optical response is developed. A real-space representation of the optical response of conjugated polyenes is developed by using the Wannier representation to derive equations of motion for coupled two-site oscillators representing correlated electron-hole pairs. The resulting elementary excitations are shown to be intermediate between the molecular (Frenkel) and the semiconductor (Wannier) excitons, and clearly resemble charge transfer excitons. Theoretical analysis of resonant and off-resonant femtosecond pump-probe absorption experiments on polydiacetylene-para-toluene sulfonate (PTS) was carried out using a 3-mode Brownian oscillator model for the nuclear dynamics. The role of spectral diffusion process was explored.

This grant resulted in 46 publications (which are listed below). In addition, a graduate level textbook entitled "Nonlinear Optical Spectroscopy: The Density Matrix Perspective" by

S. Mukamel is being completed. The book develops a unified theoretical microscopic treatment of the optical nonlinear response of molecular, conjugated-polymer, and semiconductor materials.

Publications Resulting from the AFOSR Award

1. "Cooperative Nonlinear Optical Response of Molecular Aggregates: Crossover to Bulk Behavior", F. C. Spano and Shaul Mukamel, Phys. Rev. Lett., **66**, 1197-1200 (1991).
2. "Excitons in Confined Geometries: Size Scaling of Nonlinear Susceptibilities", F.C. Spano and S.Mukamel, J. Chem. Phys., **95**, 7526-7440, (1991).
3. "Exciton Coherence-size and Phonon-mediated Optical Nonlinearities in Restricted Geometries", S. Mukamel and Oleg Dubovsky, J. Chem Phys., **95**, 7828-7844, (1991).
4. "Biexciton States and Two-Photon Absorption in Molecular Monolayers", F. C. Spano, V. Agranovich, and S. Mukamel, J. Chem. Phys., **95**, 1400-1409 (1991).
5. "Temperature Dependent Superradiant Decay of Excitons in Small Aggregates", F.C. Spano, J.R. Kuklinski and S. Mukamel, Phys. Rev. Lett. **65**, 211-214 (1990).
6. "Cooperative Radiative Dynamics in Molecular Aggregates", F.C. Spano, J.R. Kuklinski and S. Mukamel, J. Chem. Phys., **94**, 7534-7544 (1991).
7. "Note on Superradiance of Excitonic Molecules", V.M. Agranovich and S. Mukamel, Phys. Lett. A **147**, 155-160 (1990).
8. "Temperature Dependent Superradiant Decay in Molecular Aggregates", F.C. Spano, J.R. Kuklinski and S. Mukamel in Ultrafast Phenomena VII, C.B. Harris, E. P. Ippen, G.A. Mourou, and A.H. Zewail, eds., Springer-Verlag, Berlin, 507-509, (1990).
9. "Transient Gratings, Four-Wave Mixing and Polariton Effects in Nonlinear Optics", J. Knoester and S. Mukamel, Physics Reports, **205**, 1-58 (1991).
10. "Intermolecular Forces, Spontaneous Emission and Superradiance in a Dielectric Medium: Polariton -Mediated Interactions", J. Knoester and S. Mukamel, Phys. Rev. A, **40**, 7065-7080 (1989).
11. "Nonlinear Optical Response in Condensed Phases: A Microscopic Theory Using the Multipolar Hamiltonian", J. Knoester and S. Mukamel, Phys. Rev. A, **41**, 3812-3821 (1990).
12. "Note on Retardation Effects in the Dielectric Optical Response", V. M. Agranovich and S. Mukamel, Solid State Communication, **80**, 85-88 (1991).
13. "Polaron and Size Effects in Optical Lineshapes of Molecular Aggregates", N. Lu and S. Mukamel, J. Chem. Phys., **95**, 1588-1607 (1991).

14. "Optical Properties of Wannier Excitons in the Linear and Weakly Nonlinear Regime", J.R. Kuklinski and S. Mukamel, Phys. Rev. B, **42**, 2959-2976 (1990)
15. "Real-Versus-Virtual Excitonic Stark Effect in Semiconductor Quantum Wells", J.R. Kuklinski and S. Mukamel, Phys. Rev. B, **42**, 11938-11941.(1990).
16. "Generalized Semiconductor Bloch Equation: Local Fields and Transient Gratings", J. R. Kuklinski and S. Mukamel, Phys. Rev. B, **44**, 253-259 (1991).
17. "Femtosecond Pump-Probe Spectroscopy of Polyatomic Molecules in Condensed Phases", Y.J. Yan and S. Mukamel, Phys. Rev. A **41**, 6485-6504 (1990).
18. "A Classical Theory of Pump-Probe Photodissociation for Arbitrary Pulse Durations", L.E. Fried and S. Mukamel, J. Chem. Phys. **93**, 3063-3071 (1990).
19. "Intramolecular and Solvent Dynamics in Femtosecond Pump-Probe Spectroscopy", W.B. Bosma, Y.J. Yan and S. Mukamel, J. Chem. Phys. **93**, 3863-3873 (1990).
20. "Femtosecond Optical Spectroscopy: A Direct Look at Elementary Chemical Events", S. Mukamel, Ann. Rev. Phys. Chem. **41**, 647-681 (1990).
21. "Manipulation of Molecular Motions Using Femtosecond Pulse Sequences", S. Mukamel and Y.J. Yan, J. Phys.Chem. **95**, 1015-1016 (1991).
22. "Pulse Shaping and Coherent Raman Spectroscopy in Condensed Phases", Y.J. Yan and S. Mukamel, J. Chem. Phys. **94**, 997-1005 (1991).
23. "Femtosecond Optical Spectroscopies of Solvated Polyatomic Molecules", S. Mukamel and Y.J. Yan in Ultrafast Phenomena VII, C.B. Harris, E. P. Ippen, G.A. Mourou, and A.H. Zewail, eds., Springer-Verlag, Berlin, 390-392, (1990).
24. "Solvation Structure in the Time Resolved Stokes Shift and Adiabatic Electron Transfer", L.E. Fried and S. Mukamel, Molecular Crystals and Liquid Crystals **194**, 263-268 (1991).
25. "Impulsive Pump-Probe and Photon Echo Spectroscopies of Dye Molecules in Condensed Phases", W.B. Bosma, Y.J. Yan and S. Mukamel, Phys. Rev. A **42**, 6920-6923 (1990).
26. "Photon Echoes of Polyatomic Molecules in Condensed Phases", Y.J. Yan and S. Mukamel, J. Chem. Phys., **94**, 179-190 (1991).
27. "Stochastic Theory of Time-Resolved Four-Wave Mixing in Interacting Media", S. Schmitt-Rink and S. Mukamel; K. Leo and J. Shah, and D. S. Chemla, Phys. Rev. A, **44**, 2124-2129 (1991).
28. "Melting and the Electronic Absorption of Benzene-Argon Clusters", L. E. Fried and S. Mukamel, Phys. Rev. Lett. **66**, 2340-2343 (1991).

29. "Simulation of the Ultrafast Optical Response of Water", L. E. Fried and S. Mukamel, in Time-Resolved Vibrational Spectroscopy V, H. Takahashi, Editor (Springer-Verlag, Berlin) (1992) pp. 295-298.
30. "Photon Echoes and Related Four Wave Mixing Spectroscopies using Phase-Locked Pulses", M. Cho, N. F. Scherer, G. R. Fleming, and S. Mukamel, J. Chem. Phys., **96**, 5618-5629 (1992).
31. "Brownian Oscillator Analysis of Femtosecond Pump-Probe Spectroscopy of Polydiacetylene", W.B. Bosma, S. Mukamel, B.I. Greene, and S. Schmitt-Rink, Synthetic Metals, **49**, 71-76 (1992).
32. "Interplay of Excitonic and Phonon-Mediated Stark Effects in Quantum Wells", J. R. Kuklinski and S. Mukamel, J. Lum., **53**, 97-100 (1992).
33. "Structure, Dynamics, and the Electronic Absorption of Benzene-Argon Clusters", L.E. Fried and S. Mukamel, J. Chem Phys., **96**, 116 (1992).
34. "Excitons in Confined Geometries: Size Scaling of Nonlinear Susceptibilities", F.C. Spano and S. Mukamel, J. Chem. Phys., **95**, 7526-7440, (1991).
35. "Exciton Coherence-size and Phonon-mediated Optical Nonlinearities in Restricted Geometries", O. Dubovsky and S. Mukamel, J. Chem Phys., **95**, 7828-7844, (1991).
36. "Radiative Decay in Semiconductor Quantum Dots at Finite Temperatures", J.R. Kuklinski and S. Mukamel, Chem Phys. Lett., **189**, 119-127 (1992).
37. "Note on Retardation Effects in the Dielectric Optical Response", V. M. Agranovich and S. Mukamel, Solid State Comm., **80**, 85-88 (1991).
38. "Generalized Semiconductor Bloch Equation: Local Fields and Transient Gratings", J. R. Kuklinski and S. Mukamel, Phys. Rev. B., **44**, 253-259 (1991).
39. "Solvation Dynamics in Electron-transfer and Femtosecond Nonlinear Spectroscopy", S. Mukamel and W. Bosma, in Dynamics and Mechanisms of Photoinduced Transfer and Related Phenomena, N. Mataga, T. Okada and H. Mashuhara (Editors), pp 195-210 (Elsevier, Amsterdam, 1992).
40. "Simulation of the Femtosecond Optical Response of a Solute in Water", L. E. Fried, N. Bernstein and S. Mukamel, Phys. Rev. Lett., **68**, 1842-1845 (1992).
41. "Femtosecond Pump-Probe Spectroscopy of Conjugated Polymers; Coherent and Sequential Contributions", W.B. Bosma, S. Mukamel, B.I. Greene, and S. Schmitt-Rink, Phys. Rev. Lett., **68**, 2456 (1992).

42. "Nonlinear Optical Response of Conjugated Polymers; Electron-Hole Anharmonic-Oscillator Picture", S. Mukamel and H.X. Wang, Phys. Rev. Lett., **69**, 65-69 (1992).
43. "Charge-Transfer Excitons and $\chi^{(2)}$ in Molecular Assemblies", O. Dubovsky and S. Mukamel, J. Chem. Phys., **96**, 9201-9211 (1992).
44. "Optical Response of Conjugated Polyenes; Electrons, Holes and Intermediate Excitons", H.X.Wang and S. Mukamel, Chem. Phys. Lett., **192**, 417-424 (1992).
45. "Exciton Scattering Mechanism for Enhanced Nonlinear Response of Molecular Nanostructures", J.A. Leegwater and S. Mukamel, Phys. Rev. A **46**, 452-464 (1992).
46. "Many-body Effects In Nonlinear Susceptibilities; Beyond the Local-field Approximation", S. Mukamel, to be published in Nonlinear Optical Properties of Organic Molecules and Crystals, Volume 3, J. Zyss, editor, Academic Press New York, (1992).

List of Personnel involved in the AFSOR Sponsored Research

Graduate Students

Hong Xiang Wang, Ph.D. (1993)
Expected to take a postdoctoral position in January of 1994

Wayne B. Bosma, Ph.D. (1992)
Postdoctoral Fellow
University of Texas, Austin (with Professor P. Rossky)

Postdoctoral Fellows

Dr. Jasper Knoester, Presently Huygens Fellow at the University of Groningen;

Dr. Frank Spano, Presently an Assistant Professor at Temple University;

Dr. Y.J. Yan, Presently at the University of California at San Diego;

Dr. Jan Rafal Kuklinski, Presently at the Institute for Theoretical Physics, Warsaw;

Dr. L.E. Fried, Presently a Postdoctoral Fellow at the Lawrence Livermore National Laboratory, California

Dr. N. Lu, Presently a Research Scientist at the Program Development Corporation, White Plains, New York

Visiting Scientists

Prof. V. Agranovich, Institute of Spectroscopy, Moscow;

Dr. Oleg Dubovsky, Institute of Physics and Energetics, Obninsk.

External Honors

Fellow of the American Physical Society (elected 1987)

Fellow of the Optical Society of America (elected 1989)

Advisory Editor, Chemical Physics Letters (1990 - Present)

Guest Editor, Special Issue of *Journal of Physical Chemistry*, "Charge Transfer in Restricted Geometries" (April, 1992)

Committees and Chairmanships

Chairman of a conference on Charge Transfer in Restricted Geometries, Science and Technology Center, University of Rochester, July 1991

Member, Steering Committee of NSF Center for PhotoInduced Charge Transfer, University of Rochester (1991-Present)

Member of a National Science Foundation Review Panel on Material Science 1992

Invited Presentations

University of Rochester, Rochester, New York, "Nonlinear Molecular Susceptibilities: Beyond the Local Field Approximation", 1989 ACS Spring Symposium, April 1989

Columbia University, New York, New York, "Solvation Dynamics in Nonlinear Optical Processes and Electron Transfer", May 1989

Israel, The 22nd Jerusalem Symposium on Quantum Chemistry and Biochemistry: *Perspectives in Photosynthesis*, "Unified Description of Electron Transfer and Nonlinear Optical Spectroscopy", May 1989

Universität Bern, Institut für Anorganische und Physikalische Chemie, Switzerland, "Solvation Dynamics in Nonlinear Optical Lineshapes and Electron Transfer", May 1989

Cambridge University, Cambridge, England, "Solvation Dynamics and Intermolecular Forces in Nonlinear Optical Susceptibilities in Condensed Phases", May 1989

4th International Conference on Time-Resolved Vibrational Spectroscopy, Princeton, New Jersey, "Unified Description of Time and Frequency Resolved Four Wave Mixing Spectroscopies in Solution", June 1989

AT&T Bell Laboratories, Murray Hill, New Jersey, "Polaritons and Superradiance in Nonlinear Susceptibilities of Molecular Crystals and Aggregates", June 1989

Coherence and Quantum Optics VI Conference, "Polaritons and Retarded Interactions in Nonlinear Optical Susceptibilities", Rochester, New York, June 1989

Vanderbilt University, "Femtosecond Spectroscopy: A Probe for Chemical and Solvation Dynamics", Nashville, Tennessee, July 1989

Gordon Conference on Nonlinear Optics and Lasers, Wolfeboro, New Hampshire, "Nonlinear Optics in Molecular Aggregates and Crystals", July 1989

American Chemical Society National Meeting, "Femtosecond Pump-Probe and Related Spectroscopies: A Liouville-Space Theory", Miami, Florida, September 1989

University of Rochester Science and Technology Center, "Electron Transfer and Solvation Dynamics in Condensed Phases", Rochester, New York, September 1989.

Air Force Annual Contractors Meeting, "Nonlinear Susceptibilities in Molecular Aggregates: Is $\chi^{(3)}$ Enhanced by Size?", Long Beach, California, September 1989.

University of California, Los Angeles, "Femtosecond Nonlinear Spectroscopy: Coherent Vibrations, Bond Breaking and Solvation Dynamics", California, September 1989.

University of Michigan, "Femtosecond Nonlinear Spectroscopy: A Unified View of Bond Breaking, Solvation Dynamics and Electron Transfer", Ann Arbor, Michigan, October 1989.

University of Waterloo, Regional Symposium on Chemical Physics, "Solvation Dynamics in Electron Transfer and Nonlinear Optical Susceptibilities: A Unified Description", Waterloo, Ontario, Canada, November 1989.

University of Pennsylvania, "Femtosecond Pump-Probe Spectroscopy and Four Wave Mixing in Solvated Molecules and Molecular Aggregates", Philadelphia, Pennsylvania, November, 1989.

Princeton University, "Femtosecond Nonlinear Spectroscopy: Coherent Vibrations, Bond Breaking and Solvation Dynamics", Princeton, New Jersey, November 1989.

University of California, San Diego, "Femtosecond Pump-Probe Spectroscopy and Four-Wave Mixing in Solvated Molecules and Molecular Aggregates, San Diego, California, December 1989.

University of California, Riverside, "Femtosecond Nonlinear Spectroscopy: A Unified View of Bond Breaking, Solvation Dynamics and Electron Transfer", Riverside, California, December 1989.

University of Bayreuth, Emil Warburg Symposium on Dynamical Processes in Condensed Molecular Systems, "Femtosecond Nonlinear Spectroscopy of Solvated and Aggregated Polyatomic Molecules", Bayreuth, Germany, April 1990.

Institut für Theoretische Physik der Universität Frankfurt, "Nonlinear Optics and Superradiance in Molecular and Semiconductor Aggregates", Frankfurt, Germany, April 1990.

Max-Planck-Institut für Biophysikalische Chemie, "Femtosecond Spectroscopy, Nonlinear Optics and Superradiance in Solvated and Aggregated Molecules", Göttingen, Germany, April 1990.

Optical Society of America, Ultrafast Phenomena Topical Meeting, Monterey, California, "Femtosecond Pump-Probe and Photon Echo Spectroscopy of Solvated and Aggregated Polyatomic Molecules", May 1990.

Optical Society of America, International Quantum Electronic Conference, Anaheim, California, "Nonlinear Susceptibilities of Solvated Molecules and Molecular Aggregates", May 1990.

University of Rochester, Center for Photoinduced Charge Transfer Symposium on Photoinduced Charge Transfer, "Optical Nonlinearities in Semiconductors and Microstructures", Rochester, New York, June 1990.

Air Force Annual Contractors Meeting, "Nonlinear Susceptibilities of Molecular and Semiconductor Microstructures", Washington, DC, June 1990.

Seventh American Conference on Theoretical Chemistry, "Cooperative Effects and Coherence Size in Nonlinear Susceptibilities of Molecular Assemblies", San Diego, California, July 1990

American Physical Society, Sixth Interdisciplinary Laser Science Conference (ILS-VI), "Ultrafast Nonlinear Spectroscopy of Isolated, Solvated and Aggregated Molecules", Minneapolis, Minnesota, September 1990.

American Chemical Society Fall Symposium, University of Rochester, "Femtosecond Spectroscopy of Solvated and Aggregated Molecules", Rochester, New York, October 1990.

AT&T Bell Laboratories Murry Hill, "Cooperative Effects and Superradiance in Nonlinear Susceptibilities", January 1991.

Eastman Kodak Company, "Nonlinear Optical Response of Solvated and Aggregated Dyes", February 1991

Sanibel Symposium on Photodissociation and Fast Spectroscopy, "Femtosecond Spectroscopy of Isolated and Solvated Polyatomic Molecules", St. Augustine, Florida, March 1991.

IBM Corporation - Thomas J. Watson Research Center, "Nonlinear Optical Response of Isolated and Aggregated Molecules", Yorktown Heights, New York, April 1991.

NATO Conference on Coherence Phenomena in Atoms and Molecules in Laser Fields, "Femtosecond Spectroscopy of Isolated, Solvated and Aggregated Molecules", Niagara, Ontario, Canada, May 1991.

29th Yamada Conference on Dynamics and Mechanisms of Photoinduced Electron Transfer and Related Phenomena. "Solvation Dynamics in Electron Transfer and Femtosecond Nonlinear Spectroscopy", Osaka, Japan, May 1991.

Institute of Molecular Science, "Nonlinear Optical Response of Isolated and Aggregated Molecules", Okazaki, Japan, May 1991.

NTT Electrical Communication Laboratory, "Electron Coherence-size and Optical Nonlinearities in Restricted Geometries", Tokai, Ibaraki, Japan, May 1991.

NTT Research Laboratory, "Cooperative Effects and Superradiance in Molecular and Semiconductor Clusters", Musashino-shi, Tokyo, Japan, May 1991.

University of Tokyo, "Electron Coherence-Size and Optical Nonlinearities in Restricted Geometries", Tokyo, Japan, May 1991.

Fuji Photo Ashigara Research Labs, "Coherence-size and Superradiance in the Radiative Decay of J-Aggregates", Odawara, Japan, May 1991.

Fifth International Conference on Time Resolved Vibrational Spectroscopy. "Ultrafast Four Wave Mixing Spectroscopy of Solvated Polyatomics and Molecular Aggregates", Waseda University, Tokyo, Japan, June 1991.

Workshop on Pulse Shapes and Quantum Control in External Fields, "Nonlinear Optical Response in Liouville Space", Telluride, Colorado, July 1991.

International Conference on Optical Probes of Conjugated Polymers, "Exciton Coherence Length and Nonlinear Optical Response of Conjugated Polymers", Snowbird, Utah, August 1991.

American Chemical Society National Meeting - Symposium on Energy Transfer and Relaxation in Condensed Phases, "Exciton Coherence-Size and Optical Nonlinearities in Restricted Geometries", New York City, August 1991.

Eighth International Conference on Dynamical Processes in Excited States of Solids, "Coherence Length, Nonlinear Optical Response and Superradiance of Confined Excitons", Leiden, The Netherlands, August 1991.

Air Force Nonlinear Optical Polymers Contractors Meeting, "Nonlinear Optical Response of Conjugated Polymers; Electron-Hole Anharmonic-Oscillator Picture", Dayton, Ohio, December 1991.

Cornell University, "Coherence-Size and Cooperative Nonlinear Optical Response of Conjugated and Nonconjugated Molecular Assemblies", Ithaca, February 1992.

University of California, San Diego, "Exciton Coherence-Size and Cooperative Non-linear Optical Response in Restricted Geometries", San Diego, February 1992.

American Physical Society Annual Meeting, "Coherence-Size and Cooperative Non-Linear Optical Response in Confined Systems", Indianapolis, March 1992.

Air Force Office of Scientific Research, "Nonlinear Optical Response of Conjugated Polymers: Electron-Hole Anharmonic-Oscillator Picture", Washington, DC, May 1992.

Sandia National Laboratories, "Quasiparticle Exciton Representation of Frequency Dispersed Optical Nonlinearities of Conjugated Polyenes", Albuquerque, New Mexico, May 1992.

Second French-Israeli Symposium on Nonlinear Optics, "Nonlinear Optical Response of Molecular and Semiconductor Nanostructures", Port-Barcarès, France, May 1992.

Gordon Conference on Electronic Processes in Organic Materials, "Coherence Size and Nonlinear-Optical Response in Confined Systems", Andover, New Hampshire, July 1992.

US China Binational Workshop on Photoinduced Charge Transfer, "Charge Transfer Excitons in Optical Nonlinearities of Molecular Nanostructures and Conjugated Polymers", Academia Sinica, Beijing, China, August 1992.

University of Beijing, Department of Physics, "Cooperative Nonlinear Optical Response of Molecular and Semiconductor Nanostructures" Beijing, China, September 1992.

University of Michigan, Department of Chemistry, "Cooperative and Ultrafast Nonlinear Optical Spectroscopy of Confined Excitons and Molecular Nanostructures", Ann Arbor, Michigan, October 1992.

University of Indiana, Department of Chemistry, "Femtosecond Optical Spectroscopy of Molecular Systems in Condensed Phases" Bloomington, Indiana, November 1992.

University of Pittsburgh, Department of Chemistry, "Nonlinear Femtosecond Spectroscopy of Molecular Systems in Condensed Phases", Pittsburgh, Pennsylvania, November 1992.

Joint Harvard - MIT Seminar, "Cooperative and Ultrafast Nonlinear Optical Response of Confined Excitons and Molecular Nanostructures", Cambridge, Massachusetts, December 1992.