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Abstract

A dynamical theory that connects electronic motions and the nonlinear optical response of conjugated polyenes is developed by introducing the concept of electronic normal modes. A novel picture for the mechanism of optical nonlinearities is obtained by identifying the few dominant modes. This quasiparticle electron-hole representation establishes a close analogy with small semiconductor particles (quantum dots), and is very different from the traditional approach based on the electronic eigenstates. The effective conjugation length (coherence size), which controls the scaling and saturation of the static third order susceptibility $\chi^{(3)}$ with the number of double bonds, is related to the coherence of the relative motion of electron-hole pairs created upon optical excitation.

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Shaul Mukamel
University of Rochester
Department of Chemistry
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Publications in Reviewed Journals

1. "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.
2. "Many-body Effects In Nonlinear Susceptibilities; Beyond the Local-field Approximation", S. Mukamel, in Nonlinear Optical Properties of Organic Molecules and Crystals, Volume 3, J. Zyss, Editor, Academic Press, New York (1993).
3. "Interplay of Excitonic and Phonon-Mediated Stark Effects in Quantum Wells", J. R. Kuklinski and S. Mukamel, J. Lum. **53**, 97-100, 1992.
4. "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).
5. "Size Scaling and Exciton Coherence-size in the Optical Nonlinearities of Conjugated Polyenes", S. Mukamel and H.X. Wang, in Optics of Semiconductor Nanostructures, F. Henneberger, S. Schmitt-Rink and E. O. Göbel, Editors, (Akademie Verlag, Berlin, 1993) p. 361-395.
6. "Quasiparticle Exciton Representation of Frequency Dispersed Optical Nonlinearities of Conjugated Polyenes", H.X. Wang and S. Mukamel, J. Chem Phys., **97**, 8019-8036, (1992).
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8. "Real-Time Path Integral Approach to Quantum Coherence and Dephasing in Nonadiabatic Transitions and Nonlinear Optical Response", Y. Tanimura and S. Mukamel, Phys. Rev. A, **46**, 118-136 (1992).
9. "Simulation of Nonlinear Electronic Spectroscopy in the Condensed Phase", L.E. Fried and S. Mukamel, Adv. Chem. Phys., **84**, 435-516 (1993).
10. "Quantum Electrodynamics of Molecular Nanostructures", J. Jenkins and S. Mukamel, J. Chem Phys., **98**, 7046-7058 (1993).
11. "Simulation of the Intermolecular Vibrational Spectra of Liquid Water and Water Clusters", W.B. Bosma, L.E. Fried, and S. Mukamel, J. Chem Phys. **98**, 4413-4421 (1993).
12. "Transient Grating Spectroscopy of Exciton Sound Waves in Dense Exciton Fluids", Jan A. Leegwater and Shaul Mukamel, Chem. Phys. Lett., **203**, 125-130 (1993).
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14. "Quantum Confinement and Nonlinear Optical Response of Conjugated Molecules", H. X. Wang, A. Takahashi, M. Hartmann and S. Mukamel, SPIE OE/LASE 93, Los Angeles, January 16 - 23 (1993)
15. "Femtosecond Four Wave Mixing Spectroscopy of Conjugated Polymers", M. Hartmann and S. Mukamel, J. Chem. Phys., **99**, 1597-1606 (1993)

Graduate Students Supported:

Hong Xiang Wang, Peoples Republic of China	50%
Currently a Postdoctoral Fellow at Argonne National Laboratory	
Wayne Bosma, US citizen	50%
Currently a Postdoctoral Fellow at the University of Texas, Austin	

Postdoctoral Fellows Supported:

Dr. Jonathan Jenkins, Ph.D. University College London, UK.	25%
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External Honors:

Fellow of the American Physical Society (elected 1987)

Fellow of the Optical Society of America (elected 1989)

Organization of a symposium on Physical Chemistry in Restricted Geometries at the American Chemical Society National Meeting, Denver (March 1993)

Presentations:

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.

SPIE International Symposium on Advanced Electronic and Optoelectronic Materials, "Nonlinear Optical Response of Conjugated Systems : Electron-Hole Anharmonic-Oscillator Picture," Los Angeles, California, January 1993.

ACS National Meeting, "Nonlinear Optical Response of Conjugated Polyenes," Denver, Colorado, March 1993.

CLEO/QELS Quantum Electronics and Laser Science Conference, "Third Order Nonlinear Spectra of a Polydiacetylene: The Quasi-Particle Exciton Representation", Baltimore, Maryland, May 1993.

French-German-Israeli Symposium on *Dynamical Processes in Condensed Molecular Systems*, "Cooperative Effects in the Nonlinear Optical Response of Confined Excitons," Garchy, France, May 1993.

Royal Netherlands Academy Colloquium on *Femtosecond Reaction Dynamics*, "Multimode Brownian Oscillator Analysis of Ultrafast Nonlinear Optical Response," Amsterdam, Netherlands, May 1993.

Sixth International Conference on *Time-Resolved Vibrational Spectroscopy*, "Two-Dimensional Off-Resonant Femtosecond Spectroscopy of Liquids", Berlin, Germany, May 1993.

Max-Planck-Gesellschaft zur Förderung der Wissenschaften, Arbeitsgruppe *Halbleitertheorie*, "Nonlinear Optical Response of Molecular and Semiconductor Nanostructures", Berlin, Germany, May 1993.

ETH, Laboratorium für Physikalische Chemie, "Optical Nonlinearities and Femtosecond Spectroscopy of Confined Excitons", Zürich, Switzerland, May 1993.

Canadian Society for Chemistry Conference, "Cooperative and Ultrafast Nonlinear Optical Response of Confined Excitons", Montreal, Canada, June 1993.

DPC '93, 9th International Conference on the Dynamical Processes in Excited States of Solids, "Retardation Effects in the Nonlinear Optical Response; Pump Probe Spectroscopy of Nanostructures, Cambridge, Massachusetts, August 1993.