

2

SECURITY CLASSIFICATION OF THIS PAGE

REPORT DOCUMENTATION PAGE

188

1a. REPORT SECURITY CLASSIFICATION Unclassified	1b. RESTRICTION
2a. SECURITY CLASSIFICATION AUTHORITY DTIC ELECTED FEB 07, 1990	3. DISTRIBUTION STATEMENT APPROVAL unlimited
2b. DECLASSIFICATION / DOWNGRADING SCHEDULE DTIC B	

AD-A217 743

4. PERFORMING ORGANIZATION REPORT NUMBER(S) 114	5. MONITORING ORGANIZATION REPORT NUMBER(S) AFOSR-TR-86-0000
--	---

6a. NAME OF PERFORMING ORGANIZATION Depts. Chemistry & Physics State University of New York	6b. OFFICE SYMBOL (if applicable)	7a. NAME OF MONITORING ORGANIZATION AFOSR/NC
---	--------------------------------------	---

6c. ADDRESS (City, State, and ZIP Code) Fronczak Hall, Amherst Campus Buffalo, New York 14260	7b. ADDRESS (City, State, and ZIP Code) Building 410 Bolling AFB, D.C. 20332-6448
---	---

8a. NAME OF FUNDING / SPONSORING ORGANIZATION AFOSR	8b. OFFICE SYMBOL (if applicable) NC	9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER F49620-86-C-0009
--	--	---

8c. ADDRESS (City, State, and ZIP Code) Building 410 Bolling AFB, D.C. 20332-6448	10. SOURCE OF FUNDING NUMBERS		
	PROGRAM ELEMENT NO. 61102F	PROJECT NO. 2303	TASK NO. B3

11. TITLE (Include Security Classification) Molecular Dynamics and Spectroscopy at Gas-Solid Interfaces
--

12. PERSONAL AUTHOR(S) Thomas F. George
--

13a. TYPE OF REPORT Final	13b. TIME COVERED FROM 10/1/85 to 1/30/89	14. DATE OF REPORT (Year, Month, Day) January 1990	15. PAGE COUNT 11
------------------------------	--	---	----------------------

16. SUPPLEMENTARY NOTATION

17. COSATI CODES	18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number) LASER EXCITED ADSPECIES CHEMICAL VAPOR DEPOSITION STRUCTURED SURFACES PHOTOCHEMISTRY POLYMERIC SYSTEMS NONLINEAR OPTICS
FIELD	
GROUP	
SUB-GROUP	

19. ABSTRACT (Continue on reverse if necessary and identify by block number) Progress was made in the development of theories and computational programs for the following topics: infrared-laser-excited adspecies, including energy and phase relaxation, and desorption; ultraviolet-laser-induced chemical vapor deposition; resonance fluorescence at flat surfaces; photochemistry at structured surfaces, including gratings and thin films; phase-conjugated surfaces; spectroscopy in solid matrices; and nonlinear optical process in polymeric systems.

20. DISTRIBUTION / AVAILABILITY OF ABSTRACT <input checked="" type="checkbox"/> UNCLASSIFIED/UNLIMITED <input checked="" type="checkbox"/> SAME AS RPT. <input type="checkbox"/> DTIC USERS	21. ABSTRACT SECURITY CLASSIFICATION Unclassified	
22a. NAME OF RESPONSIBLE INDIVIDUAL Major Larry P. Davis	22b. TELEPHONE (Include Area Code) (202) 767-4963	22c. OFFICE SYMBOL AFOSR/NC

00 02 06 225

COMPLETED PROJECT SUMMARY

TITLE: Molecular Dynamics and Spectroscopy at Gas-Solid Interfaces

PRINCIPAL INVESTIGATOR: Thomas F. George
Departments of Chemistry and Physics & Astronomy
State University of New York at Buffalo
Buffalo, NY 14260

INCLUSIVE DATES: 1 October 1985 - 30 September 1989

CONTRACT/GRANT NUMBER: F49620-86-C-0009

COSTS AND FY SOURCE: \$123,000 FY86; \$132,000 FY87;
\$142,000 FY88; \$109,000 FY89

SENIOR RESEARCH

PERSONNEL: Henk F. Arnoldus Hyeong R. Lee
Sung G. Chung Peter T. Leung
Azizul Haque Lakshmi N. Pandey
Daniel A. Jelski Devaraj Sahu
Andy Langner Zhong-Chao Wu
Isidore Last

JUNIOR RESEARCH

PERSONNEL: Xinfu Xia

PUBLICATIONS:

"Thermalization in Multiphoton Excitation of Molecules," X. Y. Huang and T. F. George, Phys. Lett., 114A, 34-8 (1986).

"Convergent Scheme for Light Scattering from an Arbitrary Deep Metallic Grating," D. Agassi and T. F. George, Phys. Rev. B, 33, 2393-400 (1986).

"Vibrational Dephasing of Admolecules by Phonons," M. Hutchinson and T. F. George, Chem. Phys. Lett., 124, 211-5 (1986).

"Laser-Induced Bound and Metastable States in Bound-Continuum Systems," K. S. Lam and T. F. George, Phys. Rev. A, 33, 2491-503 (1986).

"Wigner Phase-Space Description Above and Below the Classical Threshold for the $H + H_2$ Reaction," H. W. Lee and T. F. George, J. Chem. Phys., 84, 6247-9 (1986).

"Analysis of Experimental Data for Neutralization of Low-Energy Ions at a Solid Surface," H. W. Lee and T. F. George, Surf. Sci., 172, 211-29 (1986).

"Surface Plasmon Dispersion Relation and Local Field Enhancement Distribution for a Deep Sinusoidal Grating," D. Agassi and T. F. George, Surf. Sci., 172, 230-56 (1986).

Approved for public release;
distribution unlimited.

"Resonance Fluorescence of a Two-Level Atom Near a Rough Metal Surface," X. Y. Huang, K. T. Lee and T. F. George, J. Chem. Phys., 85, 567-72 (1986).

"Photodissociation of Molecules at Structured Metallic Surfaces," P. T. Leung and T. F. George, J. Chem. Phys., 85, 4729-33 (1986).

"Light Scattering from a Deep Metallic Grating," D. Agassi and T. F. George, in Advances in Laser Sciences - I, W. C. Stwalley and M. Lapp, eds, AIP Conf. Proc. 146, 734-8 (1986).

"Energy and Phase Relaxation in Laser-Induced Admolecular Processes," X. Y. Huang and T. F. George, in Advances in Laser Sciences - I, W. C. Stwalley and M. Lapp, eds, AIP Conf. Proc. 146, 748-9 (1986).

"Transient Excitation of Anharmonic Adspecies by Pulsed Laser Radiation," J. Lin, X. Y. Huang and T. F. George, in Advances in Laser Sciences - I, W. C. Stwalley and M. Lapp, eds, AIP Conf. Proc. 146, 750-2 (1986).

"Thermal Relaxation of Adsorbed Atoms in an Intense Laser Field," H. F. Arnoldus, S. van Smaalen and T. F. George, Phys. Rev. B, 34, 6902-11 (1986).

"Heat Capacities of Rare Gases Adsorbed on Graphite," F. Battaglia, Y. S. Kim and T. F. George, J. Phys. Chem., 91, 414-7 (1987).

"Laser-Heating of a Transparent Crystal via Adsorbed Atoms," S. van Smaalen, H. F. Arnoldus and T. F. George, Phys. Rev. B, 35, 1142-6 (1987).

"Laser-Linewidth Effects on the Photon-Phonon Conversion Rate at a Gas-Solid Interface," H. F. Arnoldus and T. F. George, J. Opt. Soc. Am. B, 4, 195-200 (1987).

"Theory of Laser-Induced Phenomena on Conventional and Phase-Conjugated Surfaces," J. T. Lin, X. Y. Huang and T. F. George, J. Opt. Soc. Am. B, 4, 219-27 (1987).

"Multiplicative Stochastic Processes Involving the Time-Derivative of a Markov Process," H. F. Arnoldus and T. F. George, J. Math. Phys., 28, 340-6 (1987).

"Infrared-Laser Excitation of the Internal Vibrational Mode of a Diatomic Molecule Adsorbed on a Metal Surface," A. Peremans, J. Darville, J.-M. Gilles and T. F. George, Phys. Rev. B, 35, 2690-9 (1987).

"Cooperative Laser/Collision-Induced Chemical Bonds," K. S. Lam and T. F. George, Chem. Rev., 87, 155-66 (1987).

"Dynamics of a Laser-Irradiated Adatom," S. van Smaalen, A. Peremans, H. F. Arnoldus and T. F. George, Spectrochim. Acta, 43A, 201-5 (1987).

"Probe Absorption by an Atom in a Strong Finite-Linewidth Laser Field," H. F. Arnoldus and T. F. George, Phys. Rev. A, 35, 2080-8 (1987).

"Surface Distortions of Asymmetric Line Profiles," P. T. Leung and T. F. George, Chem. Phys. Lett., 134, 375-8 (1987).

ORIGINAL
COPY
FILED

Codes

Stat Serial and/or
Special

A-1

"Dynamics of Patterned Laser-Induced Chemical Vapor Deposition," D. A. Jelski and T. F. George, *J. Appl. Phys.*, 61, 2353-7 (1987).

"The Hückel Model for Small Metal Clusters. I. Geometry, Stability and Relationship to Graph Theory," Y. Wang, T. F. George, D. M. Lindsay and A. C. Beri, *J. Chem. Phys.*, 86, 3493-9 (1987).

"The Hückel Model for Small Metal Clusters. II. Orbital Energies, Shell Structures, Ionization Potentials and Extrapolation to the Bulk Limit," D. M. Lindsay, Y. Wang and T. F. George, *J. Chem. Phys.*, 86, 3500-10 (1987).

"Interaction of Xe^+ and Cl^- Ions and Their Formed Molecules with a Xe Solid Matrix," I. Last and T. F. George, *J. Chem. Phys.*, 86, 3787-94 (1987).

"Laser Pulse-Induced Molecular Collisions: Effect of the Temporal Width of the Pulse," H. W. Lee and T. F. George, in Photons and Continuum States of Atoms and Molecules, N. K. Rahman, C. Guidotti and M. Allegrini, eds, Springer-Verlag, Berlin, pp. 166-70 (1987).

"Role of Coherences in the Relaxation of Adsorbates," H. F. Arnoldus and T. F. George, *Phys. Rev. B*, 35, 5955-63 (1987).

"Quantum Adsorption for an Inverse Quadratic Potential," S. G. Chung and T. F. George, *J. Phys. Soc. Jpn. (Letters)*, 56, 1289-92 (1987).

"Confinement and Redistribution of Charges and Currents on a Surface by External Fields," H. F. Arnoldus, D. Jelski and T. F. George, *J. Math. Phys.*, 28, 1069-74 (1987).

"Generalization of Levinson's Theorem to Particle-Matter Interactions," S. G. Chung and T. F. George, *J. Math. Phys.*, 28, 1103-6 (1987).

"Low-Energy Collision-Induced Energy Transfer in the HeI^*_2 System," Y. S. Kim, M. Hutchinson and T. F. George, *J. Chem. Phys.*, 86, 5515-22 (1987).

"Collisional Redistribution Beyond the Medium-Coupling Limit," H. F. Arnoldus and T. F. George, *J. Phys. B*, 20, 2203-16 (1987).

"Determination of the Incommensurately Modulated Structure of α -Uranium Below 37 K," S. van Smaalen and T. F. George, *Phys. Rev. B*, 35, 7939-51 (1987).

"Vibrational Motions of Buckminsterfullerene," Z. C. Wu, D. A. Jelski and T. F. George, *Chem. Phys. Lett.*, 137, 291-4 (1987).

"Limit of the Image Theory for the Classical Decay Rates of Molecules at Surfaces," P. T. Leung, T. F. George and Y. C. Lee, *J. Chem. Phys.*, 86, 7227-9 (1987).

"Computational Study of Radiative Transitions in Molecular Collisions Induced by Short Laser Pulses," H. W. Lee and T. F. George, *Phys. Rev. A*, 35, 4977-87 (1987).

"The Plasmon Dispersion Relation on a Rough Surface: A Simple Approximation," D. A. Jelski and T. F. George, *J. Phys. Chem.*, 91, 3779-82 (1987).

- "Semiempirical Study of Polyatomic Rare Gas Halides: Application to the Xe_nCl Systems," I. Last and T. F. George, J. Chem. Phys., 87, 1183-93 (1987).
- "Molecular Lifetimes in the Presence of Periodically Roughened Metallic Surfaces," P. T. Leung, Z. C. Wu, D. A. Jelski and T. F. George, Phys. Rev. B, 36, 1475-9 (1987).
- "Cooperative Optical Transitions in Impurity Centers Coupled via Host Atoms," I. Last, Y. S. Kim and T. F. George, Chem. Phys. Lett., 138, 225-30 (1987).
- "Sensitivity of Atomic Line Shapes to the Laser Model," H. F. Arnoldus and T. F. George, in Spectral Line Shapes, Vol. 4, R. J. Exton, ed, A. Deepak, Hampton, Virginia, pp. 569-70 (1987).
- "The Hückel Model for Small Metal Clusters. III. Anion Structures and HMO Electron Affinities," D. M. Lindsay, L. Chu, Y. Wang and T. F. George, J. Chem. Phys., 87, 1685-9 (1987).
- "Theory of Spectroscopy and Dynamics in Laser-Irradiated Adspecies-Surface Systems," T. F. George, D. Jelski, X. Y. Huang and A. C. Beri, in Interfaces Under Laser Irradiation, L. D. Laude, D. Bäuerle and M. Wautelet, eds, Nijhoff, Dordrecht, The Netherlands, NATO ASI Series E: Applied Sciences 134, 165-83 (1987).
- "Non-Markovian Line Shapes of Physisorbed Atoms on a Crystal," H. F. Arnoldus and T. F. George, Phys. Rev. B, 36, 2987-95 (1987).
- "Energy Transfer Theory for the Classical Decay Rates of Molecules at Rough Metallic Surfaces," P. T. Leung and T. F. George, Phys. Rev. B, 36, 4664-71 (1987).
- "Theory of Laser-Stimulated Surface Processes. III. Desorption Through Vibrational Excitation by an IR Laser," A. C. Beri and T. F. George, J. Chem. Phys., 87, 4147-53 (1987).
- "Quantum Theory of Atomic Fluorescence Near a Metal Surface," H. F. Arnoldus and T. F. George, J. Chem. Phys., 87, 4263-72 (1987).
- "Vibrational Excitation of an Adbond by a Short-Pulsed Laser," S. van Smaalen and T. F. George, in Advances in Laser Sciences - II, M. Lapp, W. C. Stwalley and G. A. Kenney-Wallace, eds, AIP Conf. Proc., 160, 493-5 (1987).
- "On the Born and Markov Approximations: Phonon Relaxation and Coherent Excitation of Adsorbed Molecules," S. van Smaalen and T. F. George, J. Chem. Phys., 87, 5504-11 (1987).
- "Correlation Functions in Finite Memory-Time Reservoir Theory," H. F. Arnoldus and T. F. George, J. Math. Phys., 28, 2731-8 (1987).
- "Potential Energy Surfaces and Transition Moments of Cl Atom in Xe Solid Matrix," I. Last, T. F. George, M. E. Fajardo and V. A. Apkarian, J. Chem. Phys., 87, 5917-27 (1987).

- "Dynamical Analysis of Molecular Decay at Spherical Surfaces," P. T. Leung and T. F. George, *J. Chem. Phys.*, 87, 6722-4 (1987).
- "Coherent States for the Damped Harmonic Oscillator," K. H. Yeon, C. I. Um and T. F. George, *Phys. Rev. A*, 36, 5287-91 (1987).
- "Light Absorption by an Atom Moving Inside a Spherical Box," I. Last and T. F. George, *Chem. Phys. Lett.*, 142, 19-24 (1987).
- "Pure Dephasing of a Vibrational Adbond," S. van Smaalen and T. F. George, *J. Chem. Phys.*, 87, 7307-14 (1987).
- "Generalized Levinson's Theorem and Quantum Sticking Coefficient at 0 K," S. G. Chung and T. F. George, *J. Phys. Soc. Jpn. (Letters)*, 57, 20-3 (1988).
- "Theory of Low-Temperature Adsorption," S. G. Chung and T. F. George, *Surf. Sci.*, 194, 347-78 (1988).
- "Spontaneous Decay and Atomic Fluorescence Near a Metal Surface or an Absorbing Dielectric," H. F. Arnoldus and T. F. George, *Phys. Rev. A*, 37, 761-9 (1988).
- "Correlations Between Photons in Resonance Fluorescence Emitted by an Atom Near a Metal Surface," H. F. Arnoldus and T. F. George, *Phys. Rev. A*, 37, 770-9 (1988).
- "Surface-Enhanced Correlations Between Polarized Photons in Resonance Fluorescence," H. F. Arnoldus and T. F. George, *J. Phys. B*, 21, 431-46 (1988).
- "Classical Decay Rates for Molecules in the Presence of a Spherical Surface: A Complete Treatment," Y. S. Kim, P. T. Leung and T. F. George, *Surf. Sci.*, 195, 1-14 (1988).
- "Theory of the Tetragonal-to-Orthorhombic Structural Phase Transition in La_2CuO_4 ," D. Sahu and T. F. George, *Solid State Commun.*, 65, 1371-3 (1988).
- "Memory-Induced Extra Resonances of Adsorbates," H. F. Arnoldus and T. F. George, *Phys. Rev. Lett.*, 60, 1487-9 (1988).
- "Superconductors with Structured Surfaces: Fields and Currents," Z. C. Wu, D. A. Jelski and T. F. George, in Laser and Particle-Beam Chemical Processing for Microelectronics, D. J. Ehrlich, G. S. Higashi and M. M. Oprysko, eds, *Mater. Res. Soc. Symp. Proc.*, 101, 267-72 (1988).
- "Anomalies in the Heat-Capacity Signatures of Submonolayers Adsorbates with Attractive Lateral Interactions," Y. S. Kim, F. Battaglia and T. F. George, *J. Chem. Phys.*, 88, 7066-70 (1988).
- "Theory of Laser-Pulse-Induced Molecular Dynamics: Gas-Phase Molecular Collisions and Adbond Dynamics," H.-W. Lee, S. van Smaalen and T. F. George, in Atomic and Molecular Processes with Short Intense Laser Pulses, A. D. Bandrauk, ed, Plenum, New York, NATO ASI Series B: Physics 171, 87-95 (1988).
- "Fluorescence at a Surface," H. F. Arnoldus, P. T. Leung and T. F. George, *Kvantovaya Elektronika (Moscow)*, 15, 1161-7 (1988) [English version: *Sov. J. Quantum Electron.*, 18, 740-3 (1988)].

"Line Shape of an Atom-Crystal Bond," H. F. Arnoldus and T. F. George, *Phys. Rev. B*, 38, 978-86 (1988).

"Semiclassical Molecular Dynamics of Wavepackets in One-Dimensional Phase Space," A. Haque and T. F. George, in Condensed Matter Theories, Vol. 3, J. S. Arponen, R. F. Bishop and M. Manninen, eds, Plenum, London, pp. 115-30 (1988).

"Photochemistry at Structured Surfaces: A Classical Electromagnetic Approach," D. A. Jelski, P. T. Leung and T. F. George, *Int. Rev. Phys. Chem.*, 7, 179-207 (1988).

"Coupled Even-Parity Superconducting States," D. Sahu, A. Langner and T. F. George, *Phys. Rev. B*, 38, 2466-71 (1988).

"A Pascal-Type Triangle for the Number of Topologically Distinct Many-Electron Feynman Graphs," F. Battaglia and T. F. George, *J. Math. Chem.*, 2, 241-7 (1988).

"Electronic States of the Xe HCl Systems in Gas and Condensed Phases," I. Last and T. F. George, *J. Chem. Phys.*, 89, 3071-8 (1988).

"Large Silicon Clusters: Confirmation of Phillips' Conjecture," D. A. Jelski, Z. C. Wu and T. F. George, *Chem. Phys. Lett.*, 150, 447-51 (1988).

"Clusters: Link Between Molecules and Solids," D. A. Jelski and T. F. George, *J. Chem. Ed.*, 65, 879-83 (1988).

"Memory Effects on Infrared Adsorbate Spectra," H. F. Arnoldus and T. F. George, in Advances in Laser Sciences - III, A. C. Tam, J. L. Gole and W. C. Stwalley, eds, AIP Conf. Proc., 172, 445-7 (1988).

C. I. Um, C. W. Jun, W. H. Kahng and T. F. George, "Coefficient of First Viscosity via Three-Phonon Processes in Bulk Liquid Helium," *Phys. Rev. B* 38, 8834-7 (1988).

"Thermal Conductivity and Viscosity via Phonon-Phonon, Phonon-Roton and Roton-Roton Scatterings in Thin ⁴He Films," C. I. Um, C. W. Jun, W. H. Kahng and T. F. George, *Phys. Rev. B*, 38, 8838-49 (1988).

"Coupled s-Wave and d-Wave States in the Heavy-Fermion Superconductor U_{1-x}Th_xBe₁₃," A. Langner, D. Sahu and T. F. George, *Phys. Rev. B*, 38, 9187-90 (1988).

"Radiative Decay Rates for Molecules Near a Dielectric Sphere," P. T. Leung, Y. S. Kim and T. F. George, *J. Phys. Chem.*, 92, 6206-8 (1988).

"Coupled Even-Parity Superconducting States: Square Lattice," A. Langner, D. Sahu and T. F. George, in Superconductivity and Its Applications, H. S. Kwok and D. T. Shaw, eds, Elsevier, New York, pp. 57-62 (1988).

"Roughness-Induced Resonance for Molecular Fluorescence Near a Corrugated Metallic Surface," P. T. Leung, Y. S. Kim and T. F. George, *Phys. Rev. B*, 38, 10032-4 (1988).

"Symmetries of Spontaneous Decay for Atoms Near Any Surface," H. F. Arnoldus and T. F. George, *Surf. Sci.*, 205, 617-36 (1988).

"Remark on the Morphology-Dependent Resonance in the Decay Rate Spectrum for Molecules Near a Spherical Surface," Y. S. Kim, P. T. Leung and T. F. George, Chem. Phys. Lett., 152, 453-6 (1988).

"Propagators for Driven Coupled Harmonic Oscillators," K. H. Yeon, C. I. Um, W. H. Kahng and T. F. George, Phys. Rev. A, 38, 6224-30 (1988).

"Critical Field Nucleation at the Structured Surface of a Superconductor," Z. C. Wu, D. A. Jelski and T. F. George, Z. Phys. B, 73, 357-61 (1988).

"Molecular Spectroscopy at Corrugated Metal Surfaces," P. T. Leung and T. F. George, Spectrosc., 4, 35-41 (1989).

"Nonlocal and Quasilocal Potentials in the Spontaneous Emission of Molecular Exciplexes Coupled to the Phonon Bath of a Solid Matrix," K.-S. Lam and T. F. George, J. Chem. Phys., 90, 1048-60 (1989).

"Fresnel Coefficients for a Phase Conjugator," H. F. Arnoldus and T. F. George, J. Opt. Soc. Am. B, 6, 30-5 (1989).

"Light Scattering By a Phase Conjugator in the Four-Wave Mixing Configuration," H. F. Arnoldus and T. F. George, J. Mod. Opt., 36, 31-51 (1989).

"Effect of Finite Size on Magnetoresistance," H. R. Lee, H. G. Oh, T. F. George and C. I. Um, Phys. Rev. B, 39, 2822-5 (1989).

"Laser-Induced Metal Deposition on Semiconductors From Liquid Electrolytes," L. Nanai, I. Hevesi, F. V. Bunkin, B. S. Luk'yanchuk, M. R. Brook, G. A. Shafeev, D. A. Jelski, Z. C. Wu and T. F. George, Appl. Phys. Lett., 54, 736-8 (1989).

"Sounds in One-Dimensional Superfluid Helium," C. I. Um, W. H. Kahng, M. H. Whang, S. K. Hong, H. G. Oh and T. F. George, Phys. Rev. B, 39, 6537-43 (1989).

"Decay of Molecules at Corrugated Thin Metal Films," P. T. Leung, Y. S. Kim and T. F. George, Phys. Rev. B, 39, 9888-93 (1989).

"Model of Laser-Induced Deposition on Semiconductors From Liquid Electrolytes," Z. C. Wu, D. A. Jelski, T. F. George, L. Nanai, I. Hevesi, F. V. Bunkin, B. S. Luk'yanchuk, M. R. Brook and G. A. Shafeev, Chem. Mater., 1, 353-6 (1989).

"Recent Progress in the Theory of Laser-Assisted Collisions," H. F. Arnoldus, T. F. George, J. F. Scipione, P. L. DeVries, K. S. Lam and J. M. Yuan, in Laser Applications in Physical Chemistry, D. K. Evans, ed, Marcel Dekker, New York, pp. 329-75 (1989).

"Exact Wave Functions and Coherent States of a Damped Driven Harmonic Oscillator," H. G. Oh, H. R. Lee, T. F. George and C. I. Um, Phys. Rev. A, 39, 5515-22 (1989).

"Photoabsorption of Molecules at Corrugated Thin Metal Films," P. T. Leung, Y. S. Kim and T. F. George, J. Chem. Phys., 90, 7472-7 (1989).

"Quantum Mechanics of a Molecular System Adsorbed on a Dielectric Surface," H. G. Oh, H. R. Lee, T. F. George, C. I. Um, Y. M. Choi and W. H. Kahng, Phys. Rev. A, 40, 45-53 (1989).

"Nonresonant Interaction of a Three-Level Atom with Cavity Fields. IV. Atomic Dipole Moment and Squeezing Effects," X. S. Li, D. L. Lin, T. F. George and Z. D. Liu, Phys. Rev. A, 40, 228-36 (1989).

"Dynamics of Observed Reality: Abridged Version of Classical and Quantum Mechanics," A. Haque and T. F. George, in Condensed Matter Theories, Vol. 4, J. Keller, ed, Plenum, New York, pp. 223-36 (1989).

"Dynamics of an M-Level Atom Interacting with Cavity Fields: Effects of the Level Number on Quantum Collapse and Revival," F. Li, D. L. Lin, T. F. George and X. Li, Phys. Rev. A, 40, 1394-1401 (1989).

"Photochemistry at Corrugated Thin Metal Films: A Phenomenological Approach," P. T. Leung, Y. S. Kim and T. F. George, in Photochemistry in Thin Films, T. F. George, ed, Proc. Soc. Photo-Opt. Instrum. Eng., 1056, 139-46 (1989).

"Squeezing of Atomic Variables in the One- and Two-Photon Jaynes-Cummings Model," X. Li, D. L. Lin and T. F. George, Phys. Rev. A, 40, 2504-7 (1989).

"Electronic Energy Levels in a Quantum Well Within an In-Plane Magnetic Field," H. R. Lee, H. G. Oh, T. F. George and C. I. Um, J. Appl. Phys., 66, 2442-5 (1989).

"Density of States in a Resonant Tunneling Structure," W. Trzeciakowski, D. Sahu and T. F. George, Phys. Rev. B, 40, 6058-62 (1989).

"Dwell Time and Average Local Speed in a Resonant Tunneling Structure," L. N. Pandey, D. Sahu and T. F. George, Solid State Commun., 72, 7-11 (1989).

"Squeezing of Cavity Fields in Cascade Multiphoton Processes," F. Li, X. Li, D. L. Lin and T. F. George, J. Phys. B: At. Mol. Opt. Phys., 22, 2977-83 (1989).

"Dynamics of an M-Level Atom Interacting with Cavity Fields. II. Properties of Photon Statistics," F. Li, X. Li, D. L. Lin and T. F. George, Phys. Rev. A, 40, 5129-34 (1989).

"Optical Nutation in Polymers Irradiated by Ultrashort Laser Pulses," X. Li, D. L. Lin, T. F. George and X. Sun, Phys. Rev. B, 40, 11728-32 (1989).

"Gap States of Charged Solitons in Polyacetylene," X. Sun, D. Lu, R. Fu, D. L. Lin and T. F. George, Phys. Rev. B, 40, 12446-9 (1989).

"Electromagnetic Reaction to Molecular Relaxation and Its Effect on Absorption Near a Rough Surface," P. C. Das, A. Puri and T. F. George, Solid State Commun., in press.

"Quasi-Diffusion between Phonon and Roton Gases in Two- and Three-Dimensional Liquid Helium," C. I. Um, C. W. Jun, H. Joon Shin and T. F. George, J. Low-Temp. Phys., in press.

"Interface Phonons in Semiconductor Double Heterostructures," D. L. Lin, R. Chen and T. F. George, Solid State Commun., in press.

"Observation of Atomic Relaxation Near an Interface Through Detection of Emitted Fluorescence," H. F. Arnoldus and T. F. George, Com. At. Mol. Phys., in press.

"New Type of Optical Bistability in Polymers Mediated by Phonons," X. Li, D. L. Lin, T. F. George and X. Sun, Phys. Rev. B (Rapid Commun.), in press.

"Extraordinary Behavior of Atoms Near a Phase Conjugator," H. F. Arnoldus and T. F. George, in Coherence and Quantum Optics VI, L. Mandel and E. Wolf, eds, Plenum Press, New York, in press.

"Strongly Coupled One-Dimensional System and the Polymer," X. Sun, C. Wu, R. Fu, D. L. Lin and T. F. George, Proceedings of the Yamada Conference on Strongly Coupled Plasma Physics, S. Ichimaru, ed, Tokyo, Japan, in press.

"Light Scattering from an Atom Near the Surface of a Superlattice," X. Li, T. F. George and D. L. Lin, in Atomic and Molecular Physics, M. S. Z. Chaghtai, ed, Aligarh Muslim University, Aligarh, India, in press.

"Interference Phenomena in Atomic Emission Near an Interface: Pure Classical Effects in Quantum Radiation," H. F. Arnoldus and T. F. George, in Lasers and Applications, A. K. Popov, ed, Krasnoyarsk Institute of Physics, USSR Academy of Sciences, Krasnoyarsk, USSR, in press.

ABSTRACT OF OBJECTIVES AND ACCOMPLISHMENTS:

Progress was made in the development of theories and computational programs for the following topics: infrared-laser-excited adspecies, including energy and phase relaxation, and desorption; ultraviolet-laser-induced chemical vapor deposition; resonance fluorescence at flat surfaces; photochemistry at structured surfaces, including gratings and thin films; phase-conjugated surfaces; spectroscopy in solid matrices; and nonlinear optical processes in polymeric systems. This is summarized briefly below.

Infrared-laser-excited adspecies. The following four studies have been carried out: (1) The Born and Markov approximations for phonon relaxation and coherent excitation of adsorbed species are shown to be invalid for CO adsorbed on Ni or Cu (a strongly-bound physisorbed system) but valid for the weakly-bound system Ar on W. (2) A master equation approach which includes electron-hole excitations in the substrate reveals that the probability of finding a laser-driven adsorbed CO molecule on a Cu surface in its first excited vibrational state is 0.03, which is encouraging for experiments on laser-stimulated surface reactions involving CO. (3) The vibrational dephasing rate for OH on SiO₂ is found to be considerably faster than the energy relaxation rate, and the calculated value of 4 ps for the dephasing relaxation time (corresponding to a linewidth of 1.3 cm⁻¹) is in good agreement with experiments. (4) A master equation approach shows that a pulsed laser does not lead to a dramatic increase in the rate of desorption, and in the high-intensity limit, resonant heating and desorption reach the same saturation limit for a pulsed laser and for a continuous-wave laser.

Ultraviolet-laser-induced chemical vapor deposition. Using the Rayleigh hypothesis and determining all components of the electromagnetic field (incident laser, reflected field, image field and surface plasmon field), the dynamics of periodic structured growth of Cd on Si, resulting from photolysis of gaseous Cd(CH₃)₂, is calculated in order to explain experimental results.

Resonance fluorescence at flat surfaces. A rigorous quantum theory of atomic resonance fluorescence near a flat metallic surface is derived which allows for polarization-dependent detection involving specific transitions between degenerate substrates.

Photochemistry at structured surfaces. The following two studies have been carried out: (1) The photodissociation of I₂ above a Ag grating surface is calculated semiclassically, where there is an optimal distance of the molecule from the surface at which dissociation is a maximum. The reason for this is that the enhancement due to the surface plasmon field as the molecule is brought closer to the surface is offset by a diminution due to line-broadening effects. (2) Due to cross-coupling into long- and short-range surface plasmons and the different coupling nature between radiations from an incident laser and from the molecular dipole to a corrugated thin metallic film substrate, it is shown that enhanced photoabsorption may be achieved through control of the various film parameters.

Phase-conjugated surfaces. An atom near a phase conjugator behaves quite differently than an atom in empty space or in the vicinity of an ordinary (linear) surface, and it is shown with nonlinear optics that an atom in its ground state can fluoresce if it is sufficiently close to a phase conjugator. This phenomenon opens the door to new and novel types of surface spectroscopy.

Spectroscopy in solid matrices. To describe recent experiments on laser-induced chemical reactions in a HCl- and Cl₂-doped xenon solid, a semiempirical approach called the diatomics-in-ionic²-systems (DIIS) method is developed which accounts for the coupling between ionic and neutral species and charge delocalization among host rare-gas atoms. Calculations are carried out by treating 66 Xe matrix atoms via pairwise interactions, including polarization, with the remaining part of the matrix treated as a continuum, and it is shown that the positive charge in the ionic activated complex is distributed most often between several Xe atoms forming, for example, the Xe₁₂⁺Cl⁻, Xe₆⁺Cl⁻ and Xe₂⁺Cl⁻ molecules (Xe₂⁺Cl⁻ is the most stable ionic complex). A calculation of the excitation spectrum² of Xe₁₂Cl is in general agreement with experimental data.

Nonlinear optical processes in polymeric systems. The transient behavior of the optical susceptibility of polydiacetylene-toluene sulfonate induced by an ultrafast pump field has been investigated within a two-level model. The phenomena of optical nutation and optical tristability mediated by phonons and virtual excitons are found, which are shown to be different than in direct-gap bulk semiconductors.

AFOSR PROGRAM MANAGER: Lt Col Larry P. Davis