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PROJECT SQUID TECHNICAL REPORT MIT-89-PU

EXPERIMENTAL AND THEORETICAL STUDIES OF CHEMICAL DYNAMICS AND INSTABILITIES IN IRREVERSIBLE PROCESSES

JOHN ROSS AND F. G. KEYES CHEMISTRY DEPARTMENT MASSACHUSETTS INSTITUTE OF TECHNOLOGY CAMBRIDGE, MASSACHUSETTS 02139

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Project SQUID is a cooperative program of basic research relating to Jet Propulsion. It is sponsored by the Office of Naval Research and is administered by Purdue University through Contract N00014-75-C-1143, NR-098-038.

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Technical Report MIT-89-PU

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A COOPERATIVE PROGRAM OF FUNDAMENTAL RESEARCH AS RELATED TO JET PROPULSION OFFICE OF NAVAL RESEARCH, DEPARTMENT OF THE NAVY

CONTRACT NO0014-75-C-1143 NR-098-038

EXPERIMENTAL AND THEORETICAL STUDIES OF CHEMICAL DYNAMICS AND INSTABILITIES IN IRREVERSIBLE PROCESSES

by

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May 1978

Project SQUID Headquarters Chaffee Hall Purdue University West Lafayette, Indiana

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ABSTRACT

The final report summarizes the work accomplished under the subcontract. The overall objectives of the investigation were as follows: The determination of molecular properties of chemical dynamics for reactions of importance to combustion and propulsion. Molecular beam techniques were used for the experimental part of this work and were accompanied by theoretical studies in chemical dynamics. The second purpose was the study of the interaction of chemical reactions with transport processes and flows in gases in which instabilities may occur.

PROJECT SQUID FINAL REPORT

A. Identification

Principal Investigator: John Ross, F.G.Keyes Professor of Chemistry

Contractor: Massachusetts Institute of Technology

Contract No.: Sub 4965-10 under Contract N00014-67-0226-0005

Title:

Experimental and Theoretical Studies of Chemical Dynamics and Instabilities in Irreversible Processes

B. Duration:

October 1, 1967 - December 31, 1977

C. Participation

Other Support:

Work has been supported in part by the National Science Foundation (30%) and M.I.T. (20%).

Names of Investigators who contributed to research:

† Robert K. Brown

- * Randolph H. Burton Rashmikant C. Desai
- * Michele Flicker George P. Flynn
- * John A. Gracki Hong-sup Hahn Raymond Kapral
- *Jennifer Makowski

*David L. McFadden Charles Mims Abraham Nitzan Peter Ortoleva

* Lawrence G. Piper Itamar Procaccia George Schatz

1

1 M. A. *Ph. D.

D. Object

Purpose of Research: The determination of molecular properties of chemical dynamics for reactions of importance to combustion and propulsion. Molecular beam techniques were used for the experimental part of this work and were accompanied by theoretical studies in chemical dynamics. The second purpose was the study of the interaction of chemical reactions with transport processes and flows in gases in which instabilities may occur.

E. Achievement

Extensive progress has been made in the study of chemical dynamics both experimentally and theoretically. We developed simple but effective methods of estimating reaction cross sections which include the details of rotational and vibrational distribution of the energy of reaction in the reaction products.

The study of chemical instabilities proceeded along both theoretical and experimental lines. We demonstrated the theoretical feasibility of a large number of new phenomena such as light-induced spatial structures, chemical oscillations, and multiple stationary states. On the enclosed publication list, research sponsored in part by Project SQUID is marked with an asterisk.

Publications of Professor John Ross

- "Diffusion Coefficients of the Systems CO₂-CO₂ and CO₂-N₂O," J. Chem. Phys., 20, 436 (1952). (With I. Amdur, J. W. Irvine, Jr., and E. A. Mason.)
- "Intermolecular Potentials for the Systems CO₂-CO₂ and CO₂-N₂O," J. Chem. Phys., 20, 1620 (1952). (With I. Amdur and E. A. Mason.)
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- "Statistical Mechanical Theory of Transport Processes. IX. Contribution to the Theory of Brownian Motion," J. Chem. Phys., 24, 375 (1956).
- 5. "Energy of Interaction Between a Hydrogen Atom and a Helium Atom," J. Chem. Phys., 25, 626 (1956). (With E. A. Mason and P. N. Schatz.)
- 6. "The Energy of Interaction of He⁺ and H⁻," The Astrophysical J., 124, 485 (1956). (With E. A. Mason.)
- "Temperature Dependence of Distribution Functions in Quantum Statistical Mechanics," Phys. Rev., 107, 28 (1957). (With I. Oppenheim.)
- 8. "The Statistical Mechanical Basis of the Boltzmann Equation," Contribution to International Symposium on Statistical Mechanics and Transport Processes. Interscience Publishers, New York, 1958. (With J. G. Kirkwood.)
- 9. "Transport Equation in Quantum Gases," Phys. Rev., <u>109</u>, 1877 (1958). (With H. Mori.)
- "On the Calculation of Properties of Gases at Elevated Temperatures," Combustion and Flame, 2, 412 (1958). (With I. Amdur.)
- "Statistical Mechanical Theory of Transport Processes. XII. Dense Rigid Sphere Fluids," J. Chem. Phys., 31, 575 (1959). (With S. A. Rice, J. G. Kirkwood, and R. W. Zwanzig.)
- "Variation of a Chemical Reaction Cross Section with Energy," J. Chem. Phys., <u>32</u>, 940 (1960). (With E. F. Greene and R. W. Roberts.)

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