CONTENTS

COLLECTIVE ORIENTATIONAL RELAXATION IN DENSE DIPOLAR LIQUIDS 1

By Biman Bagchi and Amalendu Chandra

MECHANISTIC CLASSIFICATION OF CHEMICAL OSCILLATORS AND THE ROLE OF SPECIES 127

By Markus Eiswirth, Albrecht Freund, and John Ross

COMPLEX SCALING AND DYNAMICAL PROCESSES IN AMORPHOUS CONDENSED MATTER 201

By C. A. Chatzidimitriou-Dreismann

CHEMICAL KINETICS OF FLUE GAS CLEANING BY IRRADIATION WITH ELECTRONS 315

By H. Mätzing

A THEORETICAL STUDY OF ORIGINS OF RESONANCE RAMAN AND RESONANCE FLUORESCENCE USING A SPLIT-UP OF THE EMISSION CORRELATION FUNCTION 403

By H. Kono, Y. Nomura, and Y. Fujimura

Author Index 463

Subject Index 475

COLLECTIVE ORIENTATIONAL RELAXATION IN DENSE DIPOLAR LIQUIDS

BIMAN BAGCHI and AMALENDU CHANDRA

Solid State and Structural Chemistry Unit, Indian Institute of Science, Bangalore India

CONTENTS

I. Introduction
II. Equilibrium and Time-Dependent Orientational Correlation Functions
III. Relationship with Experimental Observables
  A. Dielectric Relaxation
  B. Dynamic Light Scattering
  C. Magnetic Resonance Experiments
  D. Neutron Scattering
  E. Raman and Infrared Line Shapes
  F. Quadrupolar Relaxation
  G. Solvation Dynamics (Stokes Shift)
  H. Kerr Relaxation
IV. Extended Hydrodynamic Description of Orientational Motion
V. Simple Markovian Theory of Collective Orientational Relaxation
VI. Dielectric Relaxation: The Markovian Theory
  A. $k \to 0, \omega \to 0$
  B. $k \to 0, \omega$ Fixed
  C. $\omega \to 0, k$ Fixed
VII. Inertial Effects in Polarization and Dielectric Relaxation: Collective Excitations in Dipolar Liquids
VIII. Single-Particle Orientational Dynamics and the Dielectric Friction
IX. Memory Effects in Collective Orientational Relaxation
X. Relationship Between Microscopic and Macroscopic Orientational Relaxations
XI. Role of Collective Orientational Relaxation in Solvation Dynamics and Electron Transfer Reactions
  A. Solvation Dynamics
  B. The Electron Transfer Reactions

Advances in Chemical Physics Volume LXXX, Edited by I. Prigogine and Stuart A. Rice
ISBN 0-471-53281-9 © 1991 John Wiley & Sons, Inc.

XII. Kerr Relaxation
XIII. Orientational Relaxation in a Liquid of Ellipsoidal Molecules
XIV. Orientational Relaxation in a Supercooled Dipolar Liquid
XV. Orientational Relaxation in Dipolar Mixtures
A. Collective Orientational Relaxation
B. The Dielectric Relaxation in a Binary Liquid
XVI. Collective Orientational Relaxation in the Presence of an External Field
XVII. Future Problems
A. Relaxation in a Nonhomogeneous State
B. Relaxation in Hydrogen-Bonded Liquids
C. Relaxation in a Supercooled Liquid
D. Relaxation of Molecules in Orientationally Disordered Crystals
E. Orientational Relaxation in Nematic Liquid Crystals
F. Reorientational Dynamics of Molecules with an Internal Degree of Freedom
G. Polarizable Fluids
XVIII. Conclusion
Acknowledgments
References

MECHANISTIC CLASSIFICATION OF CHEMICAL OSCILLATORS AND THE ROLE OF SPECIES

MARKUS EISWIRTH*, ALBRECHT FREUND**, and JOHN ROSS

Department of Chemistry, Stanford University, Stanford, California

CONTENTS

I. Introduction
II. Essential and Nonessential Species
  A. Definition of Nonessential Species
  B. Identification of Nonessential Species
III. Classification of Oscillators and Role of Essential Species
  A. Category 1: Critical Current Cycles
  B. Category 2: Strong Current Cycles
  C. A Remark about Enzyme Reactions
IV. Application to Mechanistic Studies
  A. General Procedure
  B. Examples
V. Discussion
VI. Summary
Appendix A: Directory of Oscillators
Appendix B: Diagrammatic Stability Analysis
Acknowledgments
References

COMPLEX SCALING AND DYNAMICAL PROCESSES IN AMORPHOUS CONDENSED MATTER

C. A. CHATZIDIMITRIOU-DREISMANN

Iwan N. Stranski-Institut für Physikalische und Theoretische Chemie
Technische Universität Berlin
Berlin, FRG

CONTENTS

I. Introduction
  A. General Introductory Remarks
  B. On the Physical Basis of Coherent-Dissipative Structures: CSM and Microscopic Irreversibility
  C. Outline
II. The Complex Scaling Method
  A. Introductory Remarks
  B. CSM, Unbounded Similarity Transformations, and Complex Symmetric Forms
III. Subdynamics in the Light of CSM
IV. Coherence, ODLRO, and Fermionic Density Matrix
  A. Introductory Remarks
  B. Box-and-Tail Form and Eigenvalues of the Fermionic $\Gamma^{(2)}$
V. Complex Scaling and Statistical Mechanics: Coherent-Dissipative Structures
  A. Introductory Remarks
  B. Formalism of Coherent-Dissipative Structures
  C. Spontaneous Creation of Coherent-Dissipative Structures: Physical Considerations
VI. Applications to Dynamical Process in Condensed Matter
  A. Remarks on "Correlations" and "Mechanisms"
  B. Proton Transfer and Proton Delocalization in Water and Aqueous Solutions
  C. Experimental Evidence for H^+ and D^+ Delocalization in H_2O/D_2O Mixtures
  D. Further Proposed Experiments on H^+ Delocalization

Advances in Chemical Physics Volume LXXX, Edited by I. Prigogine and Stuart A. Rice
ISBN 0-471-53281-9 © 1991 John Wiley & Sons, Inc.

E. Quantum Correlations in Molten Alkali Chlorides
F. Quantum Correlations in the Spin Dynamics of Paramagnetic Gd at High Temperatures
G. Quantum Correlation Effects in High T_c Superconductivity
VII. Outlook
Appendix A
Appendix B
Acknowledgments
References

CHEMICAL KINETICS OF FLUE GAS CLEANING BY IRRADIATION WITH ELECTRONS

H. MÄTZING

Kernforschungszentrum Karlsruhe GmbH, Laboratorium für Aerosolphysik und Filtertechnik I, Karlsruhe, FRG

CONTENTS

PART I: CHEMICAL KINETICS OF FLUE GAS CLEANING BY ELECTRON BEAM

I. Introduction
II. Radiolysis
  A. The Fate of Fast Electrons
  B. The Fate of Primary Species
III. Gas-phase Chemistry: Excited Species, Primary Radicals, and Ions
  A. Modeling Active Species Generation
  B. Reactions of Primary Excited Species
  C. Reactions of Primary Radicals
  D. Reactions of Primary Ions
  E. Positive Ion Chemistry
  F. Negative Ion Chemistry
  G. Ion Mutual Neutralization
  H. Overview on the Chemistry of Primary Species
IV. Gas-phase Chemistry: Positive Ions and Radicals
  A. NO_x Oxidation by Positive Ions
  B. Radical Reactions
  C. Oxidation versus Reduction
  D. A Simplified Reaction Scheme
V. The Chemistry of Ammonia
  A. Ammonia and Radicals
  B. The Formation of Ammonium Salts
  C. Ammonia and Sulfur Dioxide
  D. Effects of Ammonia Addition

Advances in Chemical Physics Volume LXXX, Edited by I. Prigogine and Stuart A. Rice
ISBN 0-471-53281-9 © 1991 John Wiley & Sons, Inc.

VI. Heterogeneous Chemistry
A. The Aerosol Surface
B. Heterogeneous Reactions
C. Effects of Gas–particle Reactions
VII. Summary
References

PART II: THE AGATE-CODE

I. Introduction
II. The Structure of the AGATE-Code
III. Data Representation
Table I. Radiolytic Processes
Table II. Positive Ion–Neutral Reactions
Table III. Negative Ion–Neutral Reactions
Table IV. Ionic Recombination Reactions in the Gas Phase
Table V. Reactions of Excited Species
Table VI. Bimolecular Reactions of (Ground-State) Neutrals
Table VII. Termolecular Reactions
Table VIII. Particle Formation and Heterogeneous Reactions
Acknowledgments
References

A THEORETICAL STUDY OF ORIGINS OF RESONANCE RAMAN AND RESONANCE FLUORESCENCE USING A SPLIT-UP OF THE EMISSION CORRELATION FUNCTION

H. KONO

Department of Basic Technology, Faculty of Engineering, Yamagata University, Yonezawa, Japan

Y. NOMURA and Y. FUJIMURA

Department of Chemistry, Faculty of Science, Tohoku University, Sendai, Japan

CONTENTS

I. Introduction
II. Theoretical Background
III. Divided Emission Correlation Functions
IV. Analytical and Numerical Results
A. The Fluorescencelike Component due to Molecular Pure Dephasing: The Correlation Function G_S^{PF}
B. The Fluorescencelike Components due to Spectral Broadening: The Correlation Functions G_S^{EF} and G_S^{TF}
C. The Ramanlike Component: The Correlation Function G_S^R
D. The Interferencelike Components: The Correlation Functions G_S^M and G_S^T
V. The Raman State and the Proper Excited State
VI. The Spectral Broadening Mechanisms and the Molecular Pure Dephasing Mechanism
VII. Non-Markovian Effect: An Analysis Based on the Stochastic Model
A. Derivation of Emission Correlation Functions
B. The Jump Process due to Frequency Modulation
C. Rearrangement of Emission Correlation Functions
D. Line Shape Calculation
VIII. Summary and Conclusion
Acknowledgments
References

Advances in Chemical Physics Volume LXXX, Edited by I. Prigogine and Stuart A. Rice ISBN 0-471-53281-9 © 1991 John Wiley & Sons, Inc.