

# CONTENTS

<i>Preface</i>	<i>vii</i>
<i>Foreword</i>	<i>ix</i>
<i>Future Contributions</i>	<i>xiii</i>
<b>1. Theory and Data Analysis in Time-Resolved Electron Diffraction</b>	<b>1</b>
1.1. Basic Assumptions and Approximations	1
1.2. Illustration of the Diffraction Signatures of Excited Molecules	7
1.3. Simplified Cumulant Analysis	10
1.4. Complete Cumulant Analysis	15
1.5. Manifestation of Chaotic Nuclear Dynamics in TRED Studies	17
References	23
<b>2. Structural Dynamics in Isolated Molecules</b>	<b>27</b>
2.1. The Development of TRED	30
2.2. The Time Dependence of the Intensity of Electrons Scattered by Photoexcited Molecules	54
2.3. Coherent Nuclear Dynamics of Spatially Oriented Molecules in the Laser Field	72
2.4. Dynamics of Spectrally Invisible Structures	82
Conclusion	90
References	92
<b>3. Ultrafast Electron Crystallography and Nanocrystallography</b>	<b>101</b>
3.1. Experimental Setups	102
3.2. Structural Dynamics in Condensed Phase	109
3.3. Time-Resolved Electron Nanocrystallography	117
Conclusion	137
References	139
<b>4. Coherent Dynamics of Nuclei and Electrons: Femtosecond and Attosecond Resolution in the TRED Technique</b>	<b>145</b>
4.1. Experimental Technique	149
4.2. Theory	164
4.3. Solution of the Inverse Problem	171
4.4. The Dynamics of the Wave Packets	173
4.5. Dynamics of Electrons—Attosecond Temporal Resolution of the TRED	197

Conclusion	218
References	218
<b>5. Ultrafast Electron Microscopy</b>	<b>231</b>
5.1. Transmission and Scanning Electron Microscopy	232
5.2. Ultrafast Electron Microscopy	235
5.3. Examples of Instrument Designs	240
5.4. Applications of Time-Resolved Microscopy	244
5.5. Future Trends	251
Conclusions	257
References	259
<i>Contents of Volumes 151–183</i>	263
<i>Index</i>	269



# Structural Dynamics in Isolated Molecules

## Contents

2.1. The Development of TRED	30
2.1.1. Early Implementations of TRED	31
2.1.2. Further Development of Experimental Techniques for TRED	34
2.1.3. Determination of Time-Zero in TRED	40
2.1.4. The Method of Reference Frames and Synchronization of Structures	41
<i>Difference Method for Time-Dependent Diffraction Data Analysis</i>	42
2.1.5. The Influence of the Coulomb Interaction in the Electron Bunch on the Determined Structural Parameters	43
<i>Description of the Model</i>	44
<i>Method for Calculating Corrections to the Electron Scattering Angle Based on the Conservation of Energy Law</i>	45
<i>Estimation of the Distortion of the Measured Parameters of the Molecules</i>	48
2.1.6. Effects of Space Charge and Energy Distribution in the Electron Bunch	51
2.2. The Time Dependence of the Intensity of Electrons Scattered by Photoexcited Molecules	54
2.2.1. Coherent Nuclear Dynamics	55
<i>Stochastic Approach to TRED Data Analysis</i>	56
2.2.2. Molecular Quantum State Tomography	58
2.2.3. Photodissociation of CS <sub>2</sub>	63
2.2.4. Unimolecular Photodissociation of 1,2-diiod-tetrafluoroethane	66
<i>Structural Dynamics of the Elimination Reaction of Iodine Atoms From the Free C<sub>2</sub>F<sub>4</sub>I<sub>2</sub> Molecules Under Collisionless Conditions</i>	66
<i>Structural Dynamics of Iodine Cleavage Reaction in C<sub>2</sub>H<sub>4</sub>I<sub>2</sub> and C<sub>2</sub>F<sub>4</sub>I<sub>2</sub> Molecules in Solution</i>	68
2.3. Coherent Nuclear Dynamics of Spatially Oriented Molecules in the Laser Field	72
2.3.1. Theory	73
2.3.2. Spatially Oriented Intermediate Structures in the Absence of External Orienting Fields	79
2.4. Dynamics of Spectrally Invisible Structures	82
<i>Pyridine, 2-Methylpyridine, and 2,6-Dimethylpyridine</i>	83
Conclusion	90
References	92



# Ultrafast Electron Crystallography and Nanocrystallography

## Contents

3.1. Experimental Setups	102
3.2. Structural Dynamics in Condensed Phase	109
3.2.1. Surfaces and Crystals	112
3.2.2. Conversion of Graphite into Diamond	113
3.2.3. Fatty Acid Crystal Bilayers: Molecular Ensembles	115
3.3. Time-Resolved Electron Nanocrystallography	117
3.3.1. Basic Concepts	119
3.3.2. Sample Preparation	120
3.3.3. Initial Data Analysis	123
3.3.4. Experimental Results	129
<i>Photoinduced Structural Changes in Gold Nanocrystals</i>	129
<i>4D-Refinement of the Structure by the Inverse Monte-Carlo Method</i>	134
Conclusion	137
References	139



# Coherent Dynamics of Nuclei and Electrons: Femtosecond and Attosecond Resolution in the TRED Technique

## Contents

4.1. Experimental Technique	149
4.1.1. Diffraction of Relativistic Electron Bunches	149
4.1.2. Diffraction of Single Electrons	158
4.1.3. Temporary Lens and the Nonorthogonal Intersection of the Wave Front of Optical Exciting Pulse and the Electron Probe <i>The Experiments in the Reflection mode for the Study of the Structural         Dynamics at the Surface</i>	160 163
4.2. Theory	164
4.3. Solution of the Inverse Problem	171
4.4. The Dynamics of the Wave Packets	173
4.4.1. Photoelectron Spectroscopy with Temporal Resolution	175
4.4.2. UED <i>The Dynamics of the Dissociation</i> <i>The Dynamics of the Photopredissociation</i>	181 182 185
4.4.3. Quantum-Dynamical Simulation of Dissociation	190
4.4.4. Supplementary of the Methods of Spectroscopy of the Transient State and UED	193
4.5. Dynamics of Electrons—Attosecond Temporal Resolution of the TRED	197
4.5.1. Ultrashort Pulses of Electromagnetic Radiation <i>The First Steps in Attosecond Physics</i> <i>Ultrashort Pulses and Quantum Control</i>	197 198 200
4.5.2. Molecular Dynamics and Tomography of the Electron Density	204
4.5.3. Laser-Induced Electron Diffraction <i>Alignment of Molecules in a Strong Laser Field</i>	212 216
Conclusion	218
References	218



# Ultrafast Electron Microscopy

## Contents

5.1. Transmission and Scanning Electron Microscopy	232
5.2. Ultrafast Electron Microscopy	235
5.3. Examples of Instrument Designs	240
5.4. Applications of Time-Resolved Microscopy	244
5.4.1. Phase Transitions in Nanoparticles	244
5.4.2. Laser-Induced Crystallization	244
5.4.3. Musical Nanoscale Instruments: A Drum, a Harp, and a Piano	245
5.4.4. 4D Electron Tomography	249
5.5. Future Trends	251
5.5.1. Spatial and Temporal Electron Microscopy with Additional Spectral Resolution	251
5.5.2. Controlling the Motions of Free Electrons by Femtosecond Light Pulses	252
Conclusions	257
References	259