## Contents

**Preface to the Second Edition** *XIII*

**Preface to the First Edition** *XV*

**1 Preliminaries of the Interaction of Light with Matter** *1*

**2 Weak-Field Photodissociation** *5*
2.1 Photoexcitation of a Molecule with a Pulse of Light *6*
2.2 State Preparation During the Pulse *8*
2.3 Photodissociation *13*
2.3.1 General Formalism *13*
2.3.2 Electronic States *20*
2.3.3 Energy-Resolved Quantities *21*
2.A Appendix: Molecular State Lifetime in Photodissociation *22*

**3 Weak-Field Coherent Control** *25*
3.1 Traditional Excitation *25*
3.2 Photodissociation from a Superposition State *26*
3.2.1 Bichromatic Control *28*
3.2.2 Energy Averaging and Satellite Contributions *31*
3.3 The Principle of Coherent Control *33*
3.4 Interference between *N*-Photon and *M*-Photon Routes *35*
3.4.1 Multiphoton Absorption *35*
3.4.2 One- vs. Three-Photon Interference *39*
3.4.2.1 One- vs. Three-Photon Interference: Three-Dimensional Formalism *41*
3.4.3 One- vs. Two-Photon Interference: Symmetry Breaking *50*
3.5 Polarization Control of Differential Cross Sections 56
3.6 Pump-Dump Control: Few Level Excitation *57*
3.A Appendix: Mode-Selective Chemistry *68*

**4 Control of Intramolecular Dynamics** *71*
4.1 Intramolecular Dynamics *71*
4.1.1 Time Evolution and the Zero-Order Basis *72*
4.1.2 Partitioning of the Hilbert Space *73*

Bibliografische Informationen
http://d-nb.info/1013143930

digitalisiert durch DEUTSCHE NATIONAL BIBLIOTHEK

4.1.3 Initial State Control and Overlapping Resonances *75*
4.1.3.1 Internal Conversion in Pyrazine *77*
4.1.3.2 Intramolecular Vibrational Redistribution: OCS *78*

**5 Optimal Control Theory** *83*
5.1 Pump-Dump Excitation with Many Levels: the Tannor–Rice Scheme *83*
5.2 Optimal Control Theory *89*
5.2.1 General Principles of Optimal Control Theory *89*

**6 Decoherence and Its Effects on Control** *95*
6.1 Decoherence *95*
6.1.1 Master Equations *98*
6.2 Sample Computational Results on Decoherence *100*
6.2.1 Electronic Decoherence *100*
6.2.2 Vibrational Decoherence in Condensed Phases *102*
6.2.3 Decoherence: Towards the Classical Limit *106*
6.3 Environmental Effects on Control: Some Theorems *109*
6.3.1 Environment Can Limit Control *109*
6.3.2 Environment Can Enhance Control *112*
6.3.2.1 Environmentally Assisted Transport *112*
6.3.3 Environmentally Assisted One-Photon Phase Control *114*
6.3.4 Isolated Molecules *116*
6.3.5 Nonisolated Systems *117*
6.4 Decoherence and Control *119*
6.4.1 The Optical Bloch Equation *120*
6.4.1.1 Decoherence Effects in One-Photon vs. Three-Photon Absorption *121*
6.4.2 Countering Collisional Effects *126*
6.4.3 Additional Control Studies *129*
6.4.4 State Stability against Decoherence *133*
6.4.5 Overlapping Resonances and Decoherence Control: Qualitative Motivation *135*
6.4.6 Control of Dephasing *139*
6.5 Countering Partially Coherent Laser Effects in Pump-Dump Control *142*
6.6 Countering CW Laser Jitter *149*
6.6.1 Laser Phase Additivity *150*
6.6.2 Incoherent Interference Control *151*

**7 Case Studies in Coherent Control** *153*
7.1 Two-Photon vs. Two-Photon Control *153*
7.1.1 Experimental Implementation *160*
7.2 Control over the Refractive Index *169*
7.2.1 Bichromatic Control *171*
7.3 The Molecular Phase in the Presence of Resonances *176*

7.3.1 Theory of Scattering Resonances *178*
7.3.2 Three-Photon vs. One-Photon Coherent Control in the Presence of Resonances *182*
7.3.2.1 Case (a): an Indirect Transition to an Isolated Resonance *184*
7.3.2.2 Case (b): a Purely Direct Transition to the Continuum *184*
7.3.2.3 Case (c): an Indirect Transition to a Set of Overlapping Resonances *185*
7.3.2.4 Case (d): a Sum of Direct and Indirect Transition to an Isolated Resonance *185*
7.4 Control of Chaotic Dynamics *186*

**8 Coherent Control of Bimolecular Processes** *191*
8.1 Fixed Energy Scattering: Entangled Initial States *191*
8.1.1 Issues in the Preparation of the Scattering Superposition *193*
8.1.2 Identical Particle Collisions *195*
8.1.3 Sample Control Results *198*
8.1.3.1 $m$ Superpositions *198*
8.1.3.2 Control in Cold Atoms: Penning vs. Dissociative Ionization *201*
8.1.3.3 Control in Electron Impact Dissociation *207*
8.1.4 Experimental Implementation: Fixed Total Energy *211*
8.1.5 Optimal Control of Bimolecular Scattering *212*
8.1.5.1 Optimized Bimolecular Scattering: the Total Suppression of a Reactive Event *214*
8.1.6 Sculpted Imploding Waves *216*
8.2 Time Domain: Fast Timed Collisions *217*
8.2.1 Nonentangled Wave Packet Superpositions: Time-Dependent Scattering *217*
8.2.2 Entangled or Wave Packets? *220*

**9 The Interaction of Light with Matter: a Closer Look** *223*
9.1 Classical Electrodynamics of a Pulse of Light *223*
9.1.1 The Classical Hamiltonian *223*
9.1.2 The Free Light Field *226*
9.2 The Dynamics of Quantized Particles and Classical Light Fields *228*

**10 Coherent Control with Quantum Light** *233*
10.1 The Quantization of the Electromagnetic Field *233*
10.1.1 Light–Matter Interactions *235*
10.2 Quantum Light and Quantum Interference *236*
10.2.1 One-Photon vs. Two-Photon Quantum Field Control *238*
10.2.1.1 Use of Number States *238*
10.2.1.2 Use of Coherent States *239*
10.2.2 Pump-Dump Coherent Control *240*

10.2.2.1 Results with Quantized Light *240*
10.2.2.2 Results with Classical Fields *242*
10.2.3 Phase-Independent Control *243*
10.3 Quantum Field Control of Entanglement *245*
10.3.1 Light–Matter Entanglement *245*
10.3.2 Creating Entanglement between a Chain of Molecules and a Radiation Field *247*
10.4 Control of Entanglement in Quantum Field Chiral Separation *250*

**11 Coherent Control beyond the Weak-Field Regime: Bound States and Resonances** *253*
11.1 Adiabatic Population Transfer *253*
11.1.1 Adiabatic States, Trapping, and Adiabatic Following *254*
11.1.2 The Multistate Extension of STIRAP *260*
11.2 An Analytic Solution of the Nondegenerate Quantum Control Problem *261*
11.3 The Degenerate Quantum Control Problem *266*
11.4 Adiabatic Encoding and Decoding of Quantum Information *271*
11.5 Multistate Piecewise Adiabatic Passage *275*
11.5.1 Multistate Piecewise Adiabatic Passage – Experiments *280*
11.5.1.1 Chirped Adiabatic Passage *281*
11.5.1.2 Rabi Flopping *283*
11.6 Electromagnetically Induced Transparency *290*
11.6.1 EIT: a Resonance Perspective *291*
11.6.2 EIT as Emerging from the Interference between Resonances *293*
11.6.2.1 Unstructured Continua *300*
11.6.2.2 Structured Continua *300*
11.6.3 Photoabsorption *301*
11.6.4 The Resonance Description of Slowing Down of Light by EIT *306*

**12 Photodissociation Beyond the Weak-Field Regime** *315*
12.1 One-Photon Dissociation with Laser Pulses *315*
12.1.1 Slowly Varying Continuum *318*
12.1.2 Bichromatic Control *319*
12.1.3 Resonance *319*
12.2 Computational Examples *325*

**13 Coherent Control Beyond the Weak-Field Regime: the Continuum** *329*
13.1 Control over Population Transfer to the Continuum by Two-Photon Processes *329*
13.1.1 The Adiabatic Approximation for a Final Continuum Manifold *330*
13.2 Pulsed Incoherent Interference Control *335*
13.3 Resonantly Enhanced Photoassociation *345*

13.3.1 Theory of Photoassociation of a Coherent Wave Packet *346*
13.3.2 Photoassociation by the Consecutive Application of APC and STIRAP *353*
13.3.3 Interference between Different Pathways *357*
13.3.4 Experimental Realizations *359*
13.4 Laser Catalysis *363*
13.4.1 The Coupling of a Bound State to Two Continua by a Laser Pulse *364*

**14 Coherent Control of the Synthesis and Purification of Chiral Molecules** *373*
14.1 Principles of Electric Dipole Allowed Enantiomeric Control *374*
14.2 Symmetry Breaking in the Two-Photon Dissociation of Pure States *376*
14.3 Purification of Racemic Mixtures by "Laser Distillation" *381*
14.4 Enantiomer Control: Oriented Molecules *395*
14.5 Adiabatic Purification of Mixtures of Right-Handed and Left-Handed Chiral Molecules *397*
14.5.1 Vibrational State Discrimination of Chiral Molecules *399*
14.5.2 Spatial Separation of Enantiomers *404*
14.5.3 Internal Hamiltonian and Dressed States *405*
14.5.4 Laser Configuration *408*
14.5.5 Spatial Separation Using a Cold Molecular Trap *409*
14.A Appendix: Computation of $B-A-B'$ Enantiomer Selectivity *413*

**15 Strong-Field Coherent Control** *415*
15.1 Strong-Field Photodissociation with Continuous Wave Quantized Fields *415*
15.1.1 The Coupled-Channels Expansion *419*
15.1.2 Number States vs. Classical Light *423*
15.2 Strong-Field Photodissociation with Pulsed Quantized Fields *425*
15.2.1 Light-Induced Potentials *426*
15.3 Controlled Focusing, Deposition, and Alignment of Molecules *429*
15.3.1 Focusing and Deposition *429*
15.3.2 Strong-Field Molecular Alignment *435*

**16 Coherent Control with Few-Cycle Pulses** *443*
16.1 The Carrier Envelope Phase *443*
16.2 Coherent Control and the CEO Frequency Measurement *445*
16.3 The Recollision Model *446*
16.3.1 Step 1: Tunnel Ionization *447*
16.3.2 Step 2: Classical "Swing" Motion *448*
16.3.3 Step 3: Recollision *448*
16.3.4 Step 4: Emission of a Photon *450*
16.4 CEP Stabilization and Control *451*
16.4.1 The Attosecond Streak Camera *452*
16.5 Coherent Control of Sample Molecular Systems *453*

16.5.1 One-Photon vs. Two-Photon Control with Few-Cycle Pulses *453*
16.5.1.1 Backward-Forward Asymmetry in the Dissociative Photoionization of $D_2$. *453*
16.5.2 Control of the Generation of High-Harmonics *455*
16.5.3 Control of Electron Transfer Processes *456*
16.5.4 Electron Transfer in Alkali Halides *457*

**17 Case Studies in Optimal Control** *463*
17.1 Creating Excited States *463*
17.1.1 Using Prepared States *467*
17.2 Optimal Control in the Perturbative Domain *468*
17.3 Adaptive Feedback Control *471*
17.4 Analysis of Adaptive Feedback Experiments *480*
17.4.1 *trans–cis* Isomerization in 3,3′-Diethyl-2,2′-thiacyanine Iodide *480*
17.4.2 Controlled Stokes Emission vs. Vibrational Excitation in Methanol *486*
17.5 Interference and Optimal Control *487*

**18 Coherent Control in the Classical Limit** *491*
18.1 The One-Photon vs. Two-Photon Scenario Revisited *491*
18.1.1 Resonant Regime *491*
18.1.2 Off-Resonant Extension *492*
18.1.3 A Three-State Example *494*
18.1.4 Quantum Features *495*
18.2 The Quartic Oscillator *496*
18.3 Control in an Optical Lattice *499*
18.3.1 Equivalence with Dipole Driving *501*
18.3.2 Computational Results *502*

**Appendix Common Notation Used in the Book (in Order of Appearance)** *507*

**References** *513*

**Subject Index** *537*