

Contents

Part I Control of Electronic Processes with Strong Laser Fields

1 Strong-Field Induced Atomic Excitation and Kinematics	3
U. Eichmann	
1.1 Introduction	3
1.2 Strong Field Excitation of Atoms by Frustrated Tunneling Ionization (FTI).	5
1.2.1 Linearly Polarized Laser Fields.	5
1.2.2 Elliptically Polarized Laser Fields	11
1.2.3 Intermediate Conclusion	11
1.2.4 Detection of Excited Atoms	12
1.3 Frustrated Tunneling Ionization in Strong-Field Fragmentation of Molecules	14
1.3.1 Hydrogen Molecule.	14
1.3.2 Small Molecules	15
1.3.3 Dimers	16
1.4 Kinematic Effects on Atoms	18
1.4.1 Acceleration of Neutral Atoms in Strong Laser Fields	18
1.4.2 Rydberg Atoms in Strong Laser Fields	21
1.5 Summary and Outlook.	23
References.	23
2 Few-Cycle-Laser-Pulse Induced and Assisted Processes in Atoms, Molecules, and Nanostructures	27
Dejan B. Milošević	
2.1 Introduction	27
2.2 Definition of Few-Cycle Laser Pulse Parameters	28
2.3 Phase Space Path Integral and Transition Matrix Element	29
2.4 Above-Threshold Ionization by Few-Cycle Pulses	33
2.5 High-Order Harmonic Generation by Few-Cycle Pulses	39

2.6	Few-Cycle-Laser-Pulse Assisted Processes	41
2.7	Concluding Remarks	44
	References	45
3	Angular Streaking for Strong Field Ionization of Molecules—Attosecond Physics Without Attosecond Pulses	49
	Jian Wu and Reinhard Dörner	
3.1	Coincidence Angular Streaking	49
3.2	Phase-Dependent Directional Molecular Bond Breaking in a Symmetric Laser Pulse	51
3.3	Electron Tunnelling Site in Electron Localization-Assisted Enhanced Ionization	54
3.4	Orientation-Dependent Single Ionization of CO Molecule	55
3.5	Sequencing Multiple Ionization of a Multicenter Molecular Cluster	57
3.6	Conclusions	60
	References	60
4	Control of Ultrafast Electron Dynamics with Shaped Femtosecond Laser Pulses: From Atoms to Solids	63
	Matthias Wollenhaupt, Tim Bayer and Thomas Baumert	
4.1	Introduction	64
4.2	Fundamentals of Femtosecond Pulse Shaping	65
4.2.1	Theoretical Description	65
4.2.2	Experimental Implementation	83
4.2.3	Adaptive Optimization	83
4.3	Isolated Model Systems	85
4.3.1	Coherence Transfer from Light to Matter	86
4.3.2	Control by Polarization-Shaped Laser Pulses	88
4.3.3	Strong Field Control	98
4.4	Control of Ionization Processes in Dielectrics	114
4.5	Summary and Conclusion	117
	References	119
Part II	Attosecond Pulses for Inducing and Probing Electronic Processes	
5	XUV Attosecond Photoionization and Related Ultrafast Processes in Diatomic and Large Molecules	125
	Victor Despré, Alexandre Marciniak, Thomas Barillot, Vincent Loriot, Arnaud Rouzée, Marc. J.J. Vrakking and Franck Lépine	
5.1	Introduction	126

5.2	The First Attoseconds of the Light-Matter Interaction: Attosecond Control of Molecular Ionization	127
5.3	Photo-Dissociation: Attosecond Control of Dissociation Pathways	131
5.4	Attosecond Control of the Charge Localization	133
5.5	Ultrafast XUV Physics Extended to Large Molecular Species: Case of PAH and Femto-Astrochemistry	135
5.6	The Ionization Step: Attosecond Delay in Photoemission in the C ₆₀ Surface Plasmon Resonance	137
5.7	Conclusion	139
	References	140
6	Attosecond Electron Spectroscopy in Molecules	143
	Francesca Calegari, Jason Greenwood, Candong Liu, Matteo Lucchini, Maurizio Reduzzi, Giuseppe Sansone, Andrea Trabattoni and Mauro Nisoli	
6.1	Introduction	144
6.2	Temporal Gating Techniques for the Generation of Isolated Attosecond Pulses	145
6.3	Streaking Spectroscopy and Carrier-Envelope Phase of Attosecond Pulses	147
6.4	Velocity Map Imaging Spectroscopy of Diatomic Molecules	150
6.5	Electron Dynamics in Biomolecules	155
	References	158
7	Controlling Atomic Photoabsorption by Intense Lasers in the Attosecond Time Domain	161
	Xiao-Min Tong and Nobuyuki Toshima	
7.1	Introduction	161
7.2	Theoretical Method	163
7.2.1	Working Equation	164
7.2.2	Interpretation of the Working Equation	165
7.2.3	Photoionization	166
7.2.4	Photoexcitation (Photoabsorption)	167
7.3	Results	169
7.3.1	IR Assisted Photoionization	169
7.3.2	IR Assisted Photoexcitation	173
7.4	Summary	174
	References	174

8 Photoionization Time Delays	177
J. Marcus Dahlström, Morgane Vacher, Alfred Maquet, Jérémie Caillat and Stefan Haessler	
8.1 Introduction	178
8.2 Phase-Shifts and Time-Delays	179
8.2.1 Formal Definition of a Photoionization Delay	179
8.2.2 Ionization Dynamics in Numerical Experiments	182
8.3 Analysis of Two-Photon XUV +IR Ionization	187
8.3.1 Asymptotic Approximation for ATI Transition Amplitudes	190
8.3.2 Extracting Time-Delay Information from Laser-Assisted Photoionization Signals	192
8.4 Review of Experimental Delay Measurements	196
8.4.1 Atomic-Delay Measurements Using Attosecond Pulse Trains	196
8.5 Conclusions	199
References	200

Part III Surfaces, Nanostructures and Solids in Strong Laser Fields

9 Ultrafast Nanoplasmonic Photoemission	205
Péter Dombi	
9.1 Introduction	205
9.1.1 Introduction to Surface Plasmon Enhanced Electron Phenomena	205
9.1.2 Surface Plasmons	206
9.2 Novel Nanoplasmonic Photoemission Phenomena	208
9.2.1 Linear Versus Nonlinear Photoemission and Photocurrents	208
9.2.2 Scale Parameters in Photoemission Processes	209
9.2.3 Mechanisms of Photoemission and Related Phenomena	209
9.2.4 Electron Acceleration Phenomena in Plasmonic Fields	219
9.2.5 Surface Plasmon Induced Electron Acceleration in the Mid-infrared	224
9.3 Conclusions and Outlook	228
References	229

10 Highly Nonlinear and Ultrafast Optical Phenomena in Metallic Nanostructures	233
L. Wimmer, M. Sivis, G. Herink, S.V. Yalunin, K.E. Echternkamp and C. Ropers	
10.1 Introduction	234
10.2 Photoelectron Dynamics at Sharp Metal Nanotips	234
10.2.1 Nonlinear Photoemission	235
10.2.2 Sub-cycle Electron Dynamics in Highly Localized Electric Fields	237
10.2.3 Photoemission from Gold Nanotips Induced by Near- and Mid-infrared Femtosecond Pulses	239
10.2.4 Nanostructure Streaking with Ultrashort THz Pulses	242
10.3 Extreme-Ultraviolet Light Generation in Plasmonic Nanostructures	247
10.3.1 Strong-Field EUV Light Generation from Gas Atoms	248
10.3.2 Experimental Methods	250
10.3.3 Results and Discussion	251
References	255

11 Attosecond XUV Pulses and Surface Plasmon Polaritons: Two Case Studies	259
Mattia Lupetti and Armin Scrinzi	
11.1 Introduction	259
11.2 Surface Plasmon Polaritons	260
11.2.1 Excitation of SPPs	262
11.2.2 Standard SPP Imaging Techniques	262
11.3 A Plasmon Enhanced Attosecond Extreme Ultraviolet Source	264
11.3.1 Spatial Structure of the Plasmonic Field	266
11.3.2 Geometry of the Tapered Nanoplasmonic Waveguide	266
11.3.3 Wave-Guiding of XUV Pulses by the Tapered Waveguide	268
11.3.4 PEAX Temporal Characterization	272
11.3.5 PEAX Spatial Properties	273
11.3.6 Comparison with Traditional Gas Harmonics	274
11.3.7 Discussion and Experimental Issues	275
11.4 Attosecond Photoscopy of Surface Excitations	276
11.4.1 Experimental Setup	277
11.4.2 Theory of Attosecond Photoscopy	278

11.4.3	Low-Speed Approximation	280
11.4.4	Approximation of the Photoelectron Distribution Function	281
11.4.5	Numerical Simulation of the Photoscopic Spectrogram	283
11.4.6	Analytic Model for the SPP Field on a Grating	284
11.4.7	Origin of Plasmon Dark and Bright Modes	288
11.4.8	Results of the Plasmon Imaging	289
11.5	Conclusions	290
	References	291
12	Ultrafast Control of Strong-Field Electron Dynamics in Solids	295
	Vladislav S. Yakovlev, Stanislav Yu. Kruchinin, Tim Paasch-Colberg, Mark I. Stockman and Ferenc Krausz	
12.1	Introduction	295
12.2	Main Theoretical Concepts	297
12.2.1	Wannier–Stark Resonances	298
12.2.2	Accelerated Bloch States	301
12.2.3	Nonresonant Interband Transitions	303
12.3	Strong-Field-Driven Electron Dynamics in Crystals	305
12.3.1	A Numerical Example	305
12.3.2	Ultrafast Injection and Control of Current in Dielectrics	307
12.4	Summary and Outlook	312
	References	313

Part IV Atoms and Molecules Driven and Probed by Intense X-Ray Pulses

13	Atomic and Molecular Systems Under Intense X-Ray Radiation	319
	Maria Krikunova, Nicusor Timneanu and Jakob Andreasson	
13.1	Introduction	319
13.2	Temporal Diagnostics of Individual FEL Pulses	322
13.2.1	Solid Surface Cross-Correlation Technique	323
13.3	Ultrafast Ionization Dynamics of Small Quantum Systems	327
13.3.1	XUV Pump—NIR Probe Experiments of Multi-electron Relaxation Dynamics	328
13.4	The Role of Ionization Dynamics for High Resolution Imaging of Bio- and Bio-like Nanoparticles	331
13.5	Automated and Unsupervised Identification and Classification of Single-Shot Single-Particle CDI Data	334

13.6	Future Perspectives of AMO Science at Novel Light Sources	336
	References	337
14	Probing Molecular Photoexcited Dynamics by Soft X-Rays	341
	Markus Gühr	
14.1	Introduction	341
14.2	Molecular Processes	343
14.2.1	Experimental Work on Molecular Dynamics Outside the BOA Framework	346
14.3	Probing Molecular Electronic Structure by Soft X-Rays	348
14.3.1	X-Ray Absorption	350
14.3.2	X-Ray Emission	352
14.3.3	Auger Electron Emission and Fragmentation	353
14.3.4	X-Ray Photoelectron Spectroscopy	355
14.4	Sources for Ultrafast X-Ray Spectroscopy	356
14.5	Ultrafast X-Ray Probing of Photoexcited Molecular Dynamics	359
14.6	Outlook	363
	References	364
	Index	373