

Contents

1 Kinematics of Atomic and Molecular Fragmentation Reactions

V.P. Shevelko and J. Ullrich	1
1.1 General Considerations	1
1.1.1 Definitions and Parameters	2
1.1.2 Fragmentation Reactions for Atomic Targets	4
1.2 Particle Kinematics: Fragmentation of Atoms	6
1.2.1 Momentum and Energy Conservation Equations in the Nonrelativistic Case	6
1.2.2 Transverse- and Longitudinal-Momentum Balances	9
1.2.3 Fast Ion-Atom Collisions: Small Momentum, Energy, and Mass Transfers	10
1.2.4 Fast Ion-Atom Collisions: Recoil-Ion Momenta	12
1.2.5 Relativistic Case	14
1.3 Ion-Atom Collisions: Illustrative Examples	15
1.3.1 Single-Electron Capture	15
1.3.2 Target Ionization	18
1.4 Photon-Atom Collisions	20
1.4.1 Photoeffect	20
1.4.2 Compton Effect	24
1.5 Particle Kinematics: Fragmentation of Molecules	25
1.5.1 Many-Body Dissociation of Fast Molecular Beams	25
1.5.2 Longitudinal Fragment Velocity Components in the Approximation $z_0 = 0$	28
1.5.3 Transverse Fragment Velocity Components in the Approximation of Zero-Beam Extension	28
1.5.4 Transverse Fragment Velocity Components in the Approximation of Zero-Beam Divergence	29
1.5.5 Fragmentation into Two Particles with Equal Masses	29
References	31

2 Recoil-Ion Momentum Spectroscopy and “Reaction Microscopes”

R. Moshhammer, D. Fischer, H. Kollmus	33
2.1 Introduction	33
2.2 Imaging Spectrometers for Ions	33
2.2.1 Time Focusing	37
2.2.2 Reconstruction of Momentum Components	37
2.2.3 Spectrometers with Position-Focusing	39
2.2.4 Electric-Field Distortions and Calibration	40
2.3 Target Preparation	41
2.3.1 Supersonic Jets	41
2.3.2 Atomic Traps (MOTRIMS)	44
2.4 Position-Sensitive Detectors	45
2.4.1 Wedge and Strip Anodes	45
2.4.2 Delay-Line Anodes	47
2.4.3 Multiple-Hit Detection	48
2.5 Imaging Spectrometers for Electrons	50
2.5.1 Direct Imaging of Electrons	51
2.5.2 Reaction Microscopes: Magnetic Guiding of Electrons . . .	52
2.5.3 Reconstruction of Electron Momenta	53
2.6 New Developments	55
References	57

3 Multiparticle Imaging of Fast Molecular Ion Beams

D. Zajfman, D. Schwalm, A. Wolf	59
3.1 Introduction	59
3.2 Basic Concepts	59
3.2.1 Distances and Times: Order of Magnitude	59
3.2.2 Three-Dimensional vs. Two-Dimensional Imaging	61
3.3 Detector Concepts and Development	62
3.3.1 Optical Detection	63
3.3.2 Electrical Detection	64
3.4 Image Reconstruction	65
3.4.1 Two-Body Fragmentation	65
3.4.2 Three-Body Channel	67
3.5 Conclusion and Outlook	68
References	69

4 Neutral-Atom Imaging Techniques

U. Müller, H. Helm	71
4.1 Introduction	71
4.2 Fast-Beam Apparatus	72
4.3 Detector Requirements and Specifications	73

4.4	Multihit Methods and Readout System	74
4.5	Data-Reduction Algorithms	76
4.5.1	Two-Body Decay	76
4.5.2	Many-Body Decay	77
4.6	Projection of the Multidimensional Cross Sections	79
4.6.1	Two-Body Decay	79
4.6.2	Three-Body Decay	80
	References	81

5 Collisional Breakup in Coulomb Systems

	T.N. Rescigno, C.W. McCurdy	83
5.1	Introduction	83
5.2	Exterior Complex Scaling: Circumventing Asymptotic Boundary Conditions	84
5.3	Scattered-Wave Formalism: Options for Computing the Wave Function	86
5.3.1	Time-Independent Approach: Linear Equations	86
5.3.2	Time-Dependent Approach: Wavepacket Propagation	88
5.4	Extraction of Physical Cross Sections	90
5.4.1	Flux-Operator Approach	91
5.4.2	Formal Rearrangement Theory and Scattering Amplitudes for Three-Body Breakup	92
5.5	Multielectron Targets	95
5.5.1	Asymptotic Subtraction	97
5.6	Conclusion	99
	References	100

6 Hyperspherical $\mathcal{R}$ -Matrix with Semiclassical Outgoing Waves

	L. Malegat, P. Selles, A.K. Kazansky	103
6.1	The Double-Electronic Continuum Problem	103
6.2	The ‘ γ , $2e$ ’ Case: a Stationary Formulation	104
6.3	Outline of the Three-Step HRM-SOW Resolution Scheme	105
6.4	First Step: Extraction of the Solution at R_0	107
6.4.1	$\mathcal{R}$ -Matrix Relation	107
6.4.2	Local Properties of the Adiabatic Channels	107
6.4.3	Frame Transformation	108
6.4.4	Discussion	109
6.5	Second Step: Propagation of the Solution from R_0 to $R_{\max}$	110
6.5.1	Defining a Semiclassical Treatment of the R -motion	110
6.5.2	Solving the Resulting R -Propagation Equation	112
6.6	Third Step: Extraction of the DPI Cross Sections at $R_{\max}$	113
6.6.1	Definition of the DPI Cross Sections	113
6.6.2	The Flux-Based Extraction: A Specificity of HRM-SOW .	114

6.6.3	Structure of the Outgoing Flux at Intermediate Distances	116
6.7	Extracting the HRM-SOW Single-Ionization Cross Sections	116
6.8	Comments on the Current State-of-the-Art in the Field	117
	References	120

7 Convergent Close-Coupling Approach to Electron-Atom Collisions

I. Bray, A.T. Stelbovics	121
7.1 Introduction	121
7.2 Electron-Hydrogen Collisions	121
7.2.1 Structure	122
7.2.2 Scattering	123
7.2.3 S-Wave Model: Proof-of-Principle	128
7.2.4 Full Calculations	131
7.3 Conclusions and Future Directions	134
References	134

8 Close-Coupling Approach to Multiple-Atomic Ionization

A.S. Kheifets	137	
8.1	Introduction: Photoionization vs. Electron–Ion Scattering	137
8.2	Two-Electron Photoionization	138
8.2.1	Shake-Off and Two-Step Mechanisms	138
8.2.2	Ground-State Correlation and Gauge Invariance	139
8.2.3	Total Cross Sections for Ionization-Excitation and Double Photoionization of Helium Isoelectronic Sequence	140
8.2.4	Angular Correlation in Two-Electron Continuum: TDCS and Circular Dichroism	142
8.2.5	Symmetrized Amplitudes of Double Photoionization and “Practical Parametrization” of TDCS	144
8.2.6	Beyond Two-Electron Targets: Double Photoionization of Beryllium and Triple Photoionization of Lithium	147
8.3	Two-Electron Charged Particle Impact Ionization	148
8.3.1	($\gamma, 2e$) and ($e, 3e$) Reactions on Helium	148
8.3.2	Second Born Corrections	149
8.4	Conclusion: Towards Larger Dynamical Freedom	150
References		150

9 Numerical Grid Methods

K.T. Taylor, J.S. Parker, D. Dundas, K.J. Meharg, L.R. Moore, E.S. Smyth, J.F. McCann	153
9.1 Introduction	153

9.2	Solution of the Time-Dependent Schrödinger Equation Using Numerical Spatial Grids and High-Order Time Propagation	153
9.2.1	Time Propagation using Taylor Series and Arnoldi Propagator Methods	154
9.2.2	Grid Methods: Finite-Difference and Discrete Variable Representation (DVR) Methods for Spatial Variables	155
9.3	Mixed Finite-Difference and Basis-Set Techniques for Spatial Variables in Spherical Geometry with Application to Laser-Driven Helium	158
9.4	Mixed Finite-Difference and DVR Techniques for Spatial Variables in Cylindrical Geometry with Application to Laser-driven H_2	163
9.5	Conclusions	167
	References	168

10 S-Matrix Approach to Intense-Field Processes in Many-Electron Systems

F.H.M. Faisal	169
10.1 Introduction	169
10.2 The Rearranged Many-Body S-Matrix Theory	169
10.3 Applications to Intense-Field Ionization Dynamics	171
10.3.1 Intense-Field Ionization of Atoms	171
10.3.2 Recoil-Momentum Distributions for Nonsequential Double Ionization	172
10.3.3 Intense-Field Ionization of Molecules	177
10.4 Conclusion	181
References	182

11 Quantum Orbits and Laser-Induced Nonsequential Double Ionization

W. Becker, S.P. Goreslavski, R. Kopold, S.V. Popruzhenko	185
11.1 Introduction	185
11.2 The S -Matrix Element	186
11.2.1 Volkov Wave Functions	186
11.2.2 The S Matrix in the Strong-Field Approximation	187
11.3 Quantum Orbits	188
11.3.1 The Saddle-Point Approximation	188
11.3.2 The Simple-Man Model	190
11.3.3 Classical Cutoffs	191
11.3.4 The Long and the Short Orbits	192
11.3.5 An Analytical Approximation to the Complex Quantum Orbits	193

11.4	Results	194
11.4.1	The Choice of the Electron–Electron Interaction Potential	195
11.4.2	The Case of the Contact Electron–Electron Interaction ..	196
11.4.3	Distribution of the Ion Momentum and the Electron Momenta	197
11.5	Resonances and the Effects of Orbits with Long Travel Time	198
11.5.1	Channel Closings	200
11.5.2	Relation to Formal Scattering Theory	201
11.6	Conclusions	202
	References	202

12 Time-Dependent Density Functional Theory in Atomic Collisions

H.J. Lüdde	205
12.1 Introduction	205
12.2 Basic Concepts of Time-Dependent Density-Functional Theory . .	206
12.3 Time-Dependent Kohn-Sham Equations	207
12.3.1 Kohn-Sham Potential	208
12.3.2 Numerical Solution of the Kohn-Sham Equations	210
12.4 Extraction of Observables	211
12.4.1 Exact Functionals	211
12.4.2 Approximate Functionals	213
12.5 Applications	214
12.5.1 Many-Electron Atoms in Strong Laser Fields	214
12.5.2 Ion-Atom Collisions Involving Many Active Electrons . .	215
12.5.3 Fragmentation of Atomic Clusters in Collisions with Ions	216
12.6 Conclusion	216
References	218

13 Electronic Collisions in Correlated Systems: From the Atomic to the Thermodynamic Limit

J. Berakdar	221
13.1 Introduction	221
13.2 Two Charged-Particle Scattering	221
13.3 Three-Particle Coulomb Continuum States	222
13.3.1 Coulomb Three-Body Scattering in Parabolic Coordinates	223
13.3.2 Remarks on the Structure of the Three-Body Hamiltonian	224
13.3.3 Dynamical Screening	227
13.4 Theory of Excited N -Particle Finite Systems	228
13.5 Continuum States of N -Charged Particles	228
13.6 Green' Function Theory of Finite Correlated Systems	230
13.6.1 Application to Four-Body Systems	231

13.6.2	Thermodynamics and Phase Transitions in Finite Systems	231
13.7	Collective Response Versus Short-Range Dynamics	232
13.7.1	Manifestations of Collective Response in Finite Systems .	233
13.8	The Quantum Field Approach: Basic Concepts	233
13.8.1	The Single-Particle Green's Function for Extended Systems	234
13.8.2	Particle-Particle and Hole-Hole Spectral Functions	237
13.8.3	The Two-Particle Photocurrent	239
13.9	Conclusion	242
	References	242

14 From Atoms to Molecules

	R. Dörner, H. Schmidt-Böcking, V. Mergel, T. Weber, L. Spielberger, O. Jagutzki, A. Knapp, and H.P. Bräuning	245
14.1	Introduction	245
14.2	Double Ionization of Helium by Photoabsorption	245
14.2.1	Energy, Momentum, and Angular Momentum Considerations	245
14.2.2	Probability and Mechanisms of Double Ionization	246
14.2.3	Electron and Ion Momentum Distributions	249
14.2.4	Fully Differential Cross Sections	250
14.3	Double Ionization of Helium by Compton Scattering	253
14.4	Double Ionization of H_2	254
14.5	Conclusions and Open Questions	256
	References	257

15 Vector Correlations in Dissociative Photoionization of Simple Molecules Induced by Polarized Light

	D. Dowek	261
15.1	Introduction	261
15.2	$(\mathbf{V}_e, \mathbf{V}_{A^+}, \hat{\mathbf{e}})$ Photoelectron-Photoion Vector Correlations in Dissociative Photoionization of Small Molecules	263
15.2.1	Experimental Approaches	263
15.2.2	Electron-Ion Kinetic Energy Correlation	264
15.2.3	$(\mathbf{V}_e, \mathbf{V}_{A^+}, \hat{\mathbf{e}})$ Angular Correlations	266
15.3	$(\mathbf{V}_{A^+}, \mathbf{V}_{B^+}, \dots \hat{\mathbf{e}})$ Vector Correlations in Multiple Ionization of Small Polyatomic Molecules	276
15.4	Conclusion and Perspectives	277
	References	278

16 Relaxation Dynamics of Core Excited Molecules Probed by Auger-Electron–Ion Coincidences

Marc Simon, Catalin Miron, and Paul Morin	283
16.1 Introduction	283
16.2 Experimental Approaches	284
16.2.1 Description of the DTA (Double–Toroidal Analyzer)	286
16.2.2 Mass Spectrometer and Coincidence Regime	287
16.3 Nuclear Motion in Competition with Resonant Auger Relaxation	288
16.3.1 Mapping Potential Energy Surfaces by Core Electron Excitation: BF_3	289
16.3.2 Molecular Dissociation Mediated by Bending Motion in the Core–Excited CO_2	292
16.4 Selective Photofragmentation	293
16.5 Dynamical Angular Correlation of the Photoelectron and the Auger Electron	296
16.6 Conclusions and Perspectives	297
References	299

17 Laser-Induced Fragmentation of Triatomic Hydrogen

H. Helm, U. Müller	303
17.1 Introduction	303
17.2 Signatures of Many-Body Interactions in Predissociation	305
17.2.1 Scalar Observables	306
17.2.2 Observation of Vector Correlations	306
17.3 Imaging Molecular Dynamics in Triatomics	307
17.3.1 Two-Body Decay	308
17.3.2 Three-Body Decay	310
17.3.3 Interpretation of Experimental Maps of Nonadiabatic Coupling	311
17.4 Outlook	314
References	315

18 Nonsequential Multiple Ionization in Strong Laser Fields

H. Rottke	317
18.1 Introduction	317
18.2 Strong–Field Single Ionization	319
18.3 Electron Rescattering on the Ion Core	321
18.4 Is Momentum Conserved in a Strong Laser Pulse?	323
18.5 An Experimental Setup	326
18.5.1 The Momentum Spectrometer	327
18.5.2 The Laser System	329
18.6 Neon Multiple Ionization	330
18.7 Argon Double Ionization: The Differences Compared to Neon	334

18.8	Conclusions and Perspectives	335
	References	336
19	Helium Double Ionization in Collisions with Electrons	
A. Dorn		339
19.1	Introduction	339
19.2	Experimental Setup	341
19.3	Low Momentum Transfer Collisions	342
19.4	Impulsive Collisions with Large Momentum Transfer	347
19.5	Conclusion and Outlook	349
	References	350
20	Fast p-He Transfer Ionization Processes: A Window to Reveal the Non-s² Contributions in the Momentum Wave Function of Ground-State He	
H. Schmidt-Böking, V. Mergel, R. Dörner, H.J. Lüdde, L. Schmidt, T. Weber, E. Weigold, A.S. Kheifets		353
20.1	Introduction	353
20.2	Experimental Technique	356
20.3	Experimental Results and Discussion of Observed Momentum Patterns	359
20.4	Shake-Off Process From Non-s ² Contributions	370
20.5	Conclusions	374
	References	377
21	Single and Multiple Ionization in Strong Ion-Induced Fields	
J. Ullrich		379
21.1	Introduction	379
21.2	Interaction of Ion-Generated Strong Fields with Atoms and Single Ionization	380
21.2.1	Ion-Generated Fields	380
21.2.2	Single Ionization at Small Perturbations	381
21.2.3	Single Ionization for Strong Ion-Induced Fields at Large Perturbations	386
21.3	Double Ionization in Ion-Generated Strong Fields	394
21.3.1	Basic Mechanisms of Double Ionization	394
21.3.2	Double Ionization at Strong Perturbations	395
21.4	Multiple Ionization in Ion-Generated Strong Fields	402
21.5	A View Into the Future	405
21.5.1	Experiments in Storage Rings	405
21.5.2	Laser-Assisted Collisions	405
	References	406

22 Coulomb-Explosion Imaging Studies of Molecular Relaxation and Rearrangement

R. Wester, D. Schwalm, A. Wolf, D. Zajfman	411
22.1 Foil-induced Coulomb-Explosion Imaging	411
22.2 Experimental Procedure	416
22.3 Selected Results	419
22.3.1 Radiative Vibrational Relaxation of HD^+	419
22.3.2 The Quasilinear Molecule CH_2^+	423
22.4 Outlook	425
References	426

23 Charged-Particle-Induced Molecular Fragmentation at Large Velocities

A. Cassimi, M. Tarisien, G. Laurent, P. Sobocinski, L. Adoui, J.Y. Chesnel, F. Frémont, B. Gervais, D. Hennecart	429
23.1 Introduction	429
23.2 Molecular Fragmentation	430
23.2.1 Branching Ratios and Multielectron Removal Cross Sections	430
23.2.2 Orientation Effect	432
23.3 Fragmentation Dynamics	434
23.3.1 Non-Coulombic Fragmentation	435
23.3.2 Polyatomic Molecules	436
23.3.3 Projectile Momentum Transfer	439
23.4 Conclusion and Future Trends	443
References	443

24 Electron-Interaction Effects in Ion-Induced Rearrangement and Ionization Dynamics: A Theoretical Perspective

T. Kirchner	447
24.1 Introduction	447
24.2 Classification of Electron-Interaction Effects: A Density Functional Approach	448
24.2.1 Effects Associated with the Kohn-Sham Potential	449
24.2.2 Effects Associated with the Density Dependence of Observables	451
24.3 Identification of Electron-Interaction Effects: Comparison with Experiment	452
24.3.1 Static Exchange Effects	452
24.3.2 Response Effects	454
24.3.3 Pauli Blocking	457
24.3.4 What Lies Beyond: Correlation Effects	459
24.4 Concluding Remarks	460

References	461
25 Ionization Dynamics in Atomic Collisions	
S.Y. Ovchinnikov, J.H. Macek, Y.S. Gordeev, G.N. Ogurtsov	463
25.1 Introduction	463
25.2 Theory of Hidden Crossings	464
25.2.1 General Formalism	465
25.2.2 S-Ionization and Superpromotion	466
25.2.3 T-Ionization and Saddle-Point Electrons	468
25.2.4 D-Ionization and Radial Decoupling	472
25.3 Sturmian Theory	473
25.3.1 Scale Transformation of Solov'ev-Vinitsky	473
25.3.2 Sturmian Basis	474
25.3.3 Wave Functions and Transition Amplitudes in Fourier Space	474
25.4 Results of Calculations	475
25.4.1 Differential Cross Sections	476
25.4.2 Total Cross Sections	479
25.5 Conclusions	481
References	482
26 Fragment-Imaging Studies of Dissociative Recombination	
A. Wolf, D. Schwalm, D. Zajfman	485
26.1 Dissociative Recombination	485
26.2 Experimental Method	486
26.3 Diatomic Molecules	488
26.3.1 Branching Ratios for the Hydrogen Molecular Ion	488
26.3.2 Noncrossing Mode Recombination (HeH^+)	494
26.3.3 Metastable States in CH^+	494
26.3.4 O_2^+ and Similar, Atmospherically Relevant Species	496
26.4 Small Polyatomic Molecules	498
26.4.1 General Experimental Aspects	498
26.4.2 Triatomic Hydrogen (H_3^+)	499
26.4.3 The Water Ion	505
26.5 Conclusions and Outlook	506
References	506
Index	509

Many-Particle Quantum Dynamics in Atomic and
Molecular Fragmentation

Ullrich, J.; Shevelko, V. (Eds.)

2003, XXVIII, 515 p. 165 illus., 11 illus. in color.,
Hardcover

ISBN: 978-3-540-00667-1