

## CONTENTS

<b>Chapter 1. An Introduction to the Electronic Structure of <math>\pi</math>-Conjugated Molecules and Polymers, and to the Concept of Electronic Bands</b>	1
<i>Jean-Luc Brédas, Seth R. Marder and Jean-Marie André</i>	
1. Periodicity and Translation Symmetry Along a Polymer Chain . . . . .	2
2. Bloch's Theorem and Band Theory . . . . .	2
3. The Hückel Method for $\pi$ -Conjugated Molecules . . . . .	4
4. Qualitative LCAO Band Theory . . . . .	10
5. Impact of the Presence of Electron-Donor and/or Electron-Acceptor Moieties . . . . .	13
5.1. Electron Affinity, Ionization Energy, Electronegativity, and Coulomb Integral . . . . .	13
5.2. Modification of the Orbitals of $\pi$ -Conjugated Systems by Inclusion of Electronegative Atoms and the Concepts of Donors and Acceptors . . . . .	14
References . . . . .	18
<b>Chapter 2. DFT 101 and Applications to <math>\pi</math>-Conjugated Systems</b>	19
<i>Thomas Körzdörfer and Jean-Luc Brédas</i>	
1. Introduction . . . . .	19
2. Density Functional Theory . . . . .	20
2.1. The KS Equations . . . . .	21
2.2. Exchange-correlation Functionals . . . . .	23
2.2.1. General classification . . . . .	23
2.2.2. Jacob's ladder . . . . .	23
2.3. Ground-state Calculations Using KS-DFT: Some Dangerous Pitfalls	27
2.3.1. Interpretation of KS orbitals and eigenvalues . . . . .	27
2.3.2. The band-gap problem . . . . .	29

2.3.3.	Self-interaction errors and spurious charge delocalization . . . . .	32
2.3.4.	Static correlation and multi-reference problems . . . . .	36
2.3.5.	Dispersion interactions . . . . .	36
2.3.6.	How to choose the “best” XC-functional . . . . .	38
3.	Time-dependent Density Functional Theory . . . . .	39
3.1.	Background . . . . .	39
3.2.	Linear-response Theory and Excited-state Energies . . . . .	40
3.3.	Excited-state Calculations Using TDDFT: Some Dangerous Pitfalls . . . . .	41
3.3.1.	Charge-transfer excitations . . . . .	42
3.3.2.	Excited states with a multiple-excitation character . . . . .	43
3.3.3.	From the oligomer to the polymer limit . . . . .	44
4.	Synopsis . . . . .	46
	Acknowledgments . . . . .	47
	References . . . . .	47

### **Chapter 3. The Molecular Dynamics Method: An Introduction**

*Claudio Zannoni*

1.	Introduction . . . . .	53
2.	Equations of Motion . . . . .	54
3.	Force Fields . . . . .	55
4.	Integration of the Equations of Motion . . . . .	63
5.	Performing the Simulation . . . . .	67
5.1.	Boundary Conditions . . . . .	67
5.2.	Equilibration . . . . .	68
5.3.	Constant Temperature, Constant Pressure MD . . . . .	69
6.	Calculation of Observable Properties . . . . .	70
6.1.	Thermodynamic Quantities . . . . .	72
6.1.1.	Density . . . . .	72
6.1.2.	Heat capacity . . . . .	72
6.2.	Atomistic Level Observables . . . . .	74
6.2.1.	X-ray scattering . . . . .	74
6.2.2.	NMR . . . . .	74
6.2.3.	Transport properties for organic electronics . . . . .	75
6.3.	Molecular Level Observables . . . . .	76
6.3.1.	The singlet distribution and its expansion . . . . .	76
6.3.2.	Orthogonal expansion of singlet distributions . . . . .	77
6.3.3.	Crystals . . . . .	78
6.3.4.	Positional ordered: Smectics . . . . .	79
6.3.5.	Orientational order . . . . .	79
6.4.	Pair Distributions . . . . .	80

7.	Dynamic Properties . . . . .	83
7.1.	Linear Response . . . . .	84
7.2.	Theory of Dielectric Response . . . . .	86
	References . . . . .	86

## Chapter 4. Photophysical Properties of Molecular Aggregates “101”

*Frank C. Spano and David Beljonne*

1.	Introduction . . . . .	93
2.	Excitons in Assemblies of Rigid Molecules . . . . .	94
3.	The Nature of Electronic Coupling . . . . .	100
3.1.	Coulombic Through-Space Coupling . . . . .	100
3.2.	Coulombic Coupling . . . . .	101
3.3.	Point Dipole–Point Dipole Coupling . . . . .	102
3.4.	Coupling Derived from Atomic Transition Charge Densities . . . . .	103
3.5.	Short Range Coupling Based on Wave Function Overlap . . . . .	105
4.	Excitons in Assemblies of Non-rigid Molecules . . . . .	106
4.1.	Vibronic Spectral Signatures of J- and H-aggregates . . . . .	111
5.	Static Disorder and Thermal Effects . . . . .	117
5.1.	Static Disorder . . . . .	117
5.2.	Thermal Effects . . . . .	120
6.	Exciton Coherence . . . . .	121
7.	Conclusions and Outlook . . . . .	124
	Acknowledgments . . . . .	126
	References . . . . .	126

## Chapter 5. Introduction to the Theory of Metal/Organic Interfaces

131

*Georg Heimel*

1.	Introduction . . . . .	131
2.	The Image-Charge Effect . . . . .	133
2.1.	Electrostatic Fundamentals . . . . .	133
2.2.	Narrowing of the Transport Gap . . . . .	134
3.	Level Broadening . . . . .	135
4.	The Push-back, Pillow, or Cushion Effect . . . . .	139
4.1.	Wave Function Overlap and Normalization . . . . .	139
4.2.	Interface Dipole Layers . . . . .	143
5.	Induced Density of Interface States . . . . .	145
6.	Surface Modification and Contamination . . . . .	149
6.1.	Molecular Monolayers . . . . .	150
6.2.	Energy-level Bending and Fermi-level Pinning . . . . .	152

7. Summary . . . . .	154
Acknowledgments . . . . .	155
References . . . . .	155
<b>Chapter 6. Experimental Characterization of Interfaces of Relevance to Organic Electronics</b>	159
<i>Gabriel Man, James Endres, Xin Lin and Antoine Kahn</i>	
1. Introduction . . . . .	159
2. Initial Considerations on Organic Surfaces and Interfaces . . . . .	161
3. Experimental Determination of Organic Surface and Interface Electronic Structures . . . . .	164
3.1. Surfaces . . . . .	164
3.1.1. Occupied states, ionization energy, and work function . . . . .	164
3.1.2. Unoccupied states and electron affinity . . . . .	172
3.1.3. Other methods used to measure electronic properties of organic surfaces . . . . .	175
3.2. Interfaces . . . . .	176
3.2.1. Full interface characterization, and energy level alignment . . . . .	176
3.2.2. Inorganic–organic interfaces . . . . .	178
3.2.3. Organic–organic interfaces . . . . .	183
4. Summary . . . . .	186
Acknowledgments . . . . .	187
References . . . . .	187
<b>Chapter 7. Charge Transport in Crystalline Organic Semiconductors</b>	193
<i>Yuan Li, Veaceslav Coropceanu and Jean-Luc Brédas</i>	
1. Introduction . . . . .	193
2. Charge-Transport Microscopic Parameters . . . . .	194
2.1. Electronic Coupling . . . . .	195
2.2. Site Energy . . . . .	200
2.3. Electron–Phonon Couplings . . . . .	202
2.4. Reorganization Energy and Local Electron–Phonon Coupling . . . . .	203
2.5. Non-local Electron–Phonon Coupling . . . . .	208
3. Charge-Transport Mechanisms and Charge-Carrier Mobility . . . . .	217
4. Conclusions . . . . .	224
Acknowledgments . . . . .	225
References . . . . .	225

<b>Chapter 8. Experimental Characterization of Charge and Exciton Transport in Organic Semiconductors</b>	<b>231</b>
<i>Wei Xie, S. Matthew Menke, C. Daniel Frisbie and Russell J. Holmes</i>	
1. Introduction . . . . .	231
2. Charge Transport . . . . .	232
2.1. Electrical Conduction in Organic Semiconductors . . . . .	232
2.1.1. Electrical conductivity and resistivity . . . . .	232
2.1.2. Carrier mobility and charge density . . . . .	233
2.1.3. Conductivity and mobility tensors . . . . .	235
2.2. Measurement of Electrical Conductivity . . . . .	236
2.2.1. Ohmic vs. Schottky contacts . . . . .	237
2.2.2. The four-terminal method . . . . .	238
2.2.3. The van der Pauw technique . . . . .	240
2.2.4. The transmission line method (TLM) . . . . .	242
2.3. Measurement of Carrier Mobility . . . . .	242
2.3.1. TOF method . . . . .	242
2.3.2. SCLC method . . . . .	244
2.3.3. Hall effect method . . . . .	246
2.3.4. FET method . . . . .	249
2.3.4.1. Linear and saturation mobility . . . . .	253
2.4. Charge Transport Models . . . . .	254
2.4.1. Band vs. band-like transport . . . . .	254
2.4.2. Hopping transport . . . . .	257
2.4.3. Band transport plus traps: Multiple trapping and release (MTR) . . . . .	258
2.5. Case Studies . . . . .	259
2.5.1. Band-like transport in rubrene single crystals . . . . .	259
2.5.2. MTR in pentacene polycrystalline thin films . . . . .	262
2.5.3. Variable range hopping transport in doped conducting polymers . . . . .	264
3. Measurements of Exciton Transport in Organic Semiconductors . . . . .	266
3.1. Definition of Exciton Diffusion Length and Diffusivity . . . . .	266
3.2. Mechanisms for Excited State Energy Transfer . . . . .	268
3.3. Disorder and Sub-diffusive Motion . . . . .	269
3.4. Techniques to Measure the Exciton Diffusion Length . . . . .	271
3.4.1. Spectroscopic techniques . . . . .	271
3.4.2. Charge carrier techniques . . . . .	277
3.4.3. Imaging exciton diffusion . . . . .	279
3.4.4. Comparison of $L_D$ measurement techniques . . . . .	279

3.5. Exciton Transport Case Studies . . . . .	279
3.5.1. Rubrene . . . . .	279
3.5.2. PTCDA . . . . .	282
3.5.3. SubPc . . . . .	284
4. Conclusions . . . . .	285
Acknowledgments . . . . .	286
References . . . . .	286
<b>Chapter 9. Impact of Organic Semiconductor Microstructure on Transport: Basic Concepts</b>	<b>293</b>
<i>Alberto Salleo</i>	
1. Introduction . . . . .	293
2. Types of Disorder in Organic Semiconductors . . . . .	296
2.1. Modulation of Crystalline Order: Paracrystallinity . . . . .	297
2.2. Local Defects . . . . .	299
2.3. One-Dimensional Extended Defects: Dislocations . . . . .	299
2.4. Two-dimensional Defects: Grain-Boundaries . . . . .	302
3. Microstructure and Transport in Organic Semiconductors . . . . .	304
3.1. Nanoscale Defects and Transport . . . . .	305
3.2. Effect of One-Dimensional Defects on Charge Transport . . . . .	309
3.3. Effect of Grain-Boundaries on Transport . . . . .	310
3.4. Small Molecules and Polymers: A Summary of the Microstructural Effects on Transport and the Limiting Factors . . . . .	317
4. Conclusion . . . . .	318
References . . . . .	318
<b>Chapter 10. Nonlinear Optics</b>	<b>325</b>
<i>André Persoons</i>	
1. Introduction . . . . .	325
2. The Maxwell Equations and Linear Optics . . . . .	326
3. Permittivity, or Dielectric Constant, and Polarizability . . . . .	329
3.1. Intermezzo: Notes on Tensor Algebra . . . . .	332
4. Nonlinear Optics, the Formalism . . . . .	337
5. Nonlinear Optical Processes and Phenomena . . . . .	341
5.1. Second Harmonic Generation . . . . .	341
5.2. Sum Frequency and Difference Frequency Generation (SFG and DFG), Phase Matching . . . . .	342
5.3. Third-Order Processes . . . . .	345
5.4. Nonlinear Refractive Index . . . . .	348
6. Molecular Hyperpolarizabilities . . . . .	349

<i>Contents</i>	xi
7. Measurement Techniques: The EFISHG Method . . . . .	352
8. Measurement Techniques: HRS . . . . .	356
9. Electro-optics and the Pockels Effect . . . . .	368
10. Nonlinear Magneto-optics and Faraday Rotation . . . . .	371
11. Third-Order Processes . . . . .	376
12. The Photorefractive Effect . . . . .	384
13. Z-Scan . . . . .	385
Further Reading . . . . .	389
<b>Chapter 11. Understanding the Relationships Among Molecular Structure, Excited-State Properties, and Polarizabilities of <math>\pi</math>-Conjugated Chromophores</b>	393 <i>Rebecca L. Giesecking, Chad Risko, Seth R. Marder, and Jean-Luc Brédas</i>
1. Energy and Molecular Polarizability: Defining the Derivative Relationships . . . . .	393
2. SOS Expressions for Molecular Polarizabilities . . . . .	397
3. Evolution of the Polarizabilities of Model Organic Molecules . . . . .	399
3.1. Molecular Orbitals (MOs) of $\pi$ -Conjugated Systems . . . . .	399
3.2. Excited-State Properties of $\pi$ -Conjugated Systems . . . . .	405
4. The SOS Expression Applied to Molecular Systems . . . . .	408
4.1. The SOS expression for $\alpha_{xx}$ . . . . .	408
4.2. The SOS expression for $\beta_{xxx}$ . . . . .	410
4.3. The SOS Expression for $\gamma_{xxxx}$ . . . . .	412
5. Conclusions . . . . .	417
Acknowledgments . . . . .	417
References . . . . .	417
<b>Index</b>	421

## CONTENTS

<b>Chapter 1. Conducting Polymers: Redox States in Conjugated Systems</b>	<b>1</b>
<i>James F. Ponder Jr. and John R. Reynolds</i>	
1. Setting the Stage . . . . .	1
2. Summary and Perspective . . . . .	15
Acknowledgments . . . . .	15
References . . . . .	15
<b>Chapter 2. OFETs: Basic Concepts and Material Designs</b>	<b>19</b>
<i>Wen-Ya Lee, Jiaoguo Mei and Zhenan Bao</i>	
1. Introduction . . . . .	19
2. Operation and Characterizations of Organic Field-Effect Transistors . . . . .	20
2.1. Current-voltage Characteristics . . . . .	21
2.1.1. Mobility, on/off and threshold voltage . . . . .	22
2.1.2. Cautions in mobility estimation . . . . .	24
2.2. Device Architecture . . . . .	27
3. Organic Semiconductor Layer . . . . .	29
3.1. <i>P</i> -type, <i>N</i> -type and Ambipolar Organic Semiconductors . . . . .	32
3.2. Case Studies of Tuning Electronic Properties in Organic Semiconductors through Molecular Design . . . . .	43
3.3. Tuning Molecular Packing in Organic Semiconductors through Processing . . . . .	45
4. Gate Dielectric . . . . .	49
4.1. Characterization of Dielectric Materials . . . . .	50
4.1.1. Dielectric constant and capacitance evaluation . . . . .	50
4.1.2. Dielectric thickness and roughness . . . . .	51
4.1.3. Interface Engineering: Surface modification . . . . .	53
4.2. Representative Dielectric Materials . . . . .	55
4.2.1. Inorganic materials . . . . .	55
4.2.2. Polymeric dielectrics . . . . .	56

4.2.3. Electrolyte gate dielectrics . . . . .	60
4.2.4. Self-assembled monolayer (SAM) and multilayer dielectric . . . . .	62
4.2.5. Organic–inorganic hybrid dielectrics . . . . .	63
5. Patterning Methods for OFETs . . . . .	65
5.1. Shadow Mask Patterning . . . . .	65
5.2. Photolithography . . . . .	65
5.3. Screen Printing . . . . .	66
5.4. Inkjet Printing . . . . .	66
5.5. Soft Lithography . . . . .	69
5.5.1. Microcontact printing ( $\mu$ CP) . . . . .	69
5.5.2. Micromolding in capillaries (MIMIC) . . . . .	71
6. Summary . . . . .	72
References . . . . .	73

**Chapter 3. Engineering Applications of OFETs in Flexible and Stretchable Electronics** 85

*Martin Kaltenbrunner and Takao Someya*

1. Introduction . . . . .	85
2. Flexible Organic Transistors, Circuits, and Devices . . . . .	86
3. Materials and Methods for Ultraflexible and Lightweight Electronic Circuits . . . . .	91
4. Stretchable Organic Circuits and Devices . . . . .	102
5. Outlook . . . . .	109
References . . . . .	112

**Chapter 4. Organic Photovoltaics: Physical Concepts Behind Device Operation** 115

*Bernard Kippelen*

1. Introduction . . . . .	115
2. Review of Semiconductor Physics . . . . .	119
2.1. Classical Thermodynamics . . . . .	119
2.2. Statistical Thermodynamics . . . . .	121
2.3. Thermal Equilibrium . . . . .	122
2.4. Fermi Level Energy and Semantic Pitfalls . . . . .	122
2.5. Fermi–Dirac Distribution Function . . . . .	122
2.6. Carrier Densities . . . . .	123
2.7. Non-Degenerate Semiconductors . . . . .	124
2.8. Intrinsic and Doped Semiconductors . . . . .	125
2.9. Non-Equilibrium Conditions and Quasi-Fermi Level Energies . . . . .	126
2.10. Quasi-Fermi Level Energy and Electrochemical Potential . . . . .	127

2.11. Carrier Recombination and Generation . . . . .	130
2.12. Carrier Transport . . . . .	132
2.12.1. Charge mobility and drift current density . . . . .	133
2.12.2. Diffusion coefficient and diffusion current density . . . . .	134
2.12.3. Einstein relations . . . . .	135
2.12.4. Total charge current . . . . .	136
2.12.5. Continuity equation and surface recombination velocity . . . . .	138
3. Physics of Solar Cells . . . . .	139
3.1. Photocurrent, Photovoltage, and Power Conversion Efficiency . . . . .	139
3.2. Design of Organic Solar Cells . . . . .	143
3.2.1. Design of the absorber . . . . .	143
3.2.2. Charge-collecting layers and electrodes . . . . .	144
3.3. Ideal Shockley Diode Equation . . . . .	148
3.4. Equivalent Circuit and Parasitic Resistances . . . . .	150
3.4.1. Equivalent-circuit analysis applied to organic solar cells . . . . .	153
Acknowledgments . . . . .	156
References . . . . .	157

**Chapter 5. Organic Semiconductors: Manipulation and Control of the Microstructure of Active Layers**      159  
*Neil D. Treat, Paul Westacott and Natalie Stingelin*

1. Introduction . . . . .	159
2. Phase Behavior of Single and Multicomponent Functional Organic Systems . . . . .	161
2.1. One-Component Systems . . . . .	161
2.1.1. Crystalline domains (long-range order) . . . . .	162
2.1.2. Liquid crystallinity . . . . .	166
2.1.3. Local order and aggregation (short-range order) . . . . .	168
2.1.4. Disorder and amorphous phases . . . . .	170
2.2. Two-Components Systems . . . . .	171
2.2.1. Co-crystal formation and solid solutions . . . . .	171
2.2.2. Eutectic systems . . . . .	173
2.2.3. Multi-phase systems: Case study — Polymer: Fullerene blends . . . . .	175
3. Solidification . . . . .	177
3.1. Liquid–Liquid Phase Separation and Spinodal Decomposition . . . . .	177
3.2. Crystallization . . . . .	179
3.2.1. Nucleation and growth . . . . .	183
3.3. Vitrification . . . . .	184
4. Microstructure Development in the Solid State . . . . .	187
4.1. Annealing (Tempering) . . . . .	187

5. Conclusion . . . . .	187
Acknowledgments . . . . .	188
References . . . . .	188
<b>Chapter 6. Organic Light Emitting Devices</b>	<b>195</b>
<i>Rasha Hamze, Peter I. Djurovich and Mark E. Thompson</i>	
1. Organic Light Emitting Diodes . . . . .	195
1.1. Basic Steps in Electroluminescence . . . . .	196
1.2. Emissive Dopants for Increased OLED Efficiency . . . . .	198
1.3. OLED Efficiency and Chromaticity: Units . . . . .	199
1.3.1. Carrier recombination and spin . . . . .	201
1.3.2. Phosphorescent materials as emitters in OLEDs . . . . .	202
2. Introduction to SOC . . . . .	203
2.1. External and Internal Heavy Atom Effects . . . . .	204
2.2. SOC in Organo-Transition Metallic Complexes . . . . .	204
2.2.1. Formalisms for SOC between MLCT states . . . . .	206
2.2.2. SOC in terms of perturbation theory . . . . .	207
2.3. SOC Dependence on Coordination Geometry . . . . .	208
2.3.1. Pseudo-octahedral and square planar complexes . . . . .	208
2.3.2. Tetrahedral complexes . . . . .	208
2.4. Collecting Triplet States through Thermally Assisted Delayed Fluorescence . . . . .	209
3. Cyclometallated Complexes for OLEDs . . . . .	211
3.1. Synthesis of Cyclometallated IR Complexes . . . . .	214
3.2. Tuning Emission Energy in Cyclometallated Complexes . . . . .	216
3.3. Blue Luminescent Cyclometallated Complexes . . . . .	218
3.4. Phosphorescent OLEDs . . . . .	222
3.5. TADF-Based OLEDs . . . . .	227
3.6. White OLEDs . . . . .	228
References . . . . .	229
<b>Chapter 7. Polymer Light Emitting Diodes</b>	<b>243</b>
<i>S. Matthew Menke, Richard H. Friend and Dan Credgington</i>	
1. History and Principles . . . . .	243
2. Device Structure . . . . .	244
3. Device Physics . . . . .	246
3.1. Polymer Luminescence . . . . .	247
3.2. Polarons and Excitons . . . . .	248
3.3. Color Tuning . . . . .	248
3.4. White Light Emission . . . . .	250

4.	Quantum Efficiency . . . . .	251
4.1.	Electron–Hole Capture . . . . .	251
4.2.	Luminescence Efficiency . . . . .	256
4.3.	Outcoupling Efficiency . . . . .	257
5.	Spin . . . . .	259
5.1.	Intersystem Crossing . . . . .	260
5.2.	Phosphorescence . . . . .	261
5.3.	Thermally Activated Reverse Intersystem Crossing . . . . .	263
5.4.	Triplet Fusion . . . . .	267
5.5.	Spin-Dependant Electron–Hole Capture . . . . .	271
6.	Summary . . . . .	272
	References . . . . .	272

**Chapter 8. Thermoelectric Properties of Conducting Polymers**      277*Xavier Crispin*

1.	Introduction . . . . .	277
2.	Thermoelectric Energy Conversion . . . . .	279
3.	Thermoelectric Materials . . . . .	283
4.	Doping . . . . .	284
5.	Electronic Structure . . . . .	287
6.	Power Factor . . . . .	289
7.	Thermal Conductivity . . . . .	292
8.	Organic TEGs . . . . .	293
9.	Conclusion . . . . .	295
	Acknowledgments . . . . .	295
	References . . . . .	295

**Chapter 9. Effects of Magnetic Fields in Organic Devices: Basic Concepts**      299*Eitan Ehrenfreund and Z. Valy Vardeny*

1.	Introduction . . . . .	299
2.	A Brief Introduction To Organic Electronic and Spintronic Devices . . .	300
2.1.	Organic Light Emitting Diodes . . . . .	300
2.2.	Organic Photovoltaic Cell . . . . .	301
2.3.	Bipolar Organic Spin-Valve Device: Spin Polarized Light Emitting Diode . . . . .	302
3.	Magnetic Field Effects in Organic Diodes; Hyperfine Interaction in Polaron Pairs . . . . .	304
4.	Magnetic Field Effects in Organic Photovoltaic Cells; the $\Delta g$ Mechanism . . . . .	307
5.	Unipolar Organic Spin-Valves . . . . .	309

6. Bipolar Organic Spin-Valves in the Space Charge Limited Current Regime . . . . .	311
7. Conclusions and Outlook . . . . .	316
Acknowledgments . . . . .	317
References . . . . .	317
<b>Chapter 10. Organic Conducting Polymer Actuators</b>	321
<i>W. Zheng, G.M. Spinks and G.G. Wallace</i>	
1. Introduction . . . . .	321
2. Preparation and Performance of Polypyrrole Based Actuators . . . . .	323
3. Preparation and Performance of PANi-Based Actuators . . . . .	327
4. Preparation and Performance of PTh-Based Actuators . . . . .	330
5. Enhancement of CPs Actuator Performance . . . . .	332
5.1. Increased Strain and Speed . . . . .	334
5.2. Enhanced Cycle Life . . . . .	335
5.3. Enhanced Mechanical Work Output . . . . .	336
6. Microscale Conducting Polymer Actuators . . . . .	337
7. Conclusions . . . . .	337
Acknowledgments . . . . .	338
References . . . . .	338
<b>Chapter 11. Organic Bioelectronics for Interfacing with the Brain</b>	345
<i>Christophe Bernard, Daniel T. Simon and George G. Malliaras</i>	
1. Introduction . . . . .	345
2. The Human Brain . . . . .	347
3. Interfacing Electronics with Electrolytes . . . . .	351
4. Organic Devices that Use Ions as Part of their Operation . . . . .	354
4.1. Conducting Polymer Electrodes . . . . .	354
4.2. Organic Electrochemical Transistors . . . . .	358
4.3. Organic Electronic Ion Pumps . . . . .	361
5. Outlook . . . . .	365
Acknowledgments . . . . .	366
References . . . . .	366
<b>Chapter 12. NLO: Electro-Optic Applications</b>	369
<i>Larry Dalton, Matthias Lauermann and Christian Koos</i>	
1. Introduction . . . . .	369
2. Simple stripline Devices . . . . .	373
3. Resonant Devices . . . . .	377

4.	Prisms and Spatial Light Modulators . . . . .	382
5.	OEO Materials: Evolution and Issues . . . . .	382
6.	All-Organic Devices . . . . .	383
7.	SOH Devices . . . . .	384
7.1.	Silicon Photonics and SOH Integration . . . . .	384
7.2.	SOH Device Concepts . . . . .	385
7.3.	EO Materials for SOH Devices . . . . .	386
7.4.	Demonstration of SOH Devices . . . . .	387
8.	Conclusions and Outlook . . . . .	390
	Acknowledgments . . . . .	390
	References . . . . .	391

## Chapter 13. Two-Photon Absorption: Concepts, Molecular Materials and Applications

397

*Joel M. Hales, San-Hui Chi, Vincent W. Chen  
and Joseph W. Perry*

1.	Introduction . . . . .	397
2.	Fundamental Concepts of 2PA . . . . .	400
2.1.	Beam Attenuation due to 2PA . . . . .	401
2.2.	Probability of Excitation via 2PA . . . . .	405
2.3.	Microscopic and Macroscopic Parameters . . . . .	406
3.	2PA Properties of Conjugated Molecules . . . . .	408
3.1.	Dipolar Molecules: Two-State Model . . . . .	410
3.2.	Quadrupolar Molecules: Three-State Model . . . . .	414
3.3.	Octupolar Molecules . . . . .	418
4.	Applications of 2PA . . . . .	420
4.1.	Two-Photon Excitation Fluorescence Spectroscopy . . . . .	421
4.2.	2PFM . . . . .	422
4.3.	Two-Photon Photodynamic Therapy . . . . .	424
4.4.	3D Microfabrication . . . . .	425
4.5.	OPL . . . . .	428
4.6.	Other Applications . . . . .	433
5.	Summary and Outlook . . . . .	434
6.	List of Abbreviations Used . . . . .	435
7.	List of Symbols Used . . . . .	436
8.	Appendices . . . . .	437
8.1.	Appendix A . . . . .	437
8.2.	Appendix B . . . . .	438
	References . . . . .	439

## Index

443