

Contents

1	Introduction	1
	Hassan Raza	
1.1	Overview	1
1.2	Book Summary	7
1.3	Outlook	10
	References.....	11

Part I Metrology and Synthesis

2	Raman Spectroscopy: Characterization of Edges, Defects, and the Fermi Energy of Graphene and sp^2 Carbons	15
	M.S. Dresselhaus, A. Jorio, L.G. Cançado, G. Dresselhaus, and R. Saito	
2.1	Introduction to the Resonance Raman Spectra of Graphene.....	15
2.1.1	The Raman Spectra of sp^2 Carbons	16
2.1.2	Edge Structure of Graphene	18
2.1.3	The Multiple-Resonance Raman Scattering Process ...	18
2.1.4	Concept of the Kohn Anomaly	21
2.1.5	Introduction to Near-Field Raman Spectroscopy	22
2.2	Characterization of Defects.....	22
2.2.1	Point Defects Induced by Ion Bombardment	23
2.2.2	Model for the D-Band Activated Region	24
2.2.3	Line Defects at the Edges of Nanographene.....	26
2.3	Characterization of Edges	29
2.3.1	Overview of Graphene Edges	29
2.3.2	The Characterization of Graphene Edges from Their <i>D</i> -Band Scattering	30
2.3.3	Mode assignments of the Raman Spectra of Graphene Nanoribbons	34
2.3.4	Polarization Dependence of the Raman Intensity	38

2.4	The Fermi Energy Dependence: The Kohn Anomaly	40
2.4.1	Effect of Gate Doping on the <i>G</i> -Band of Single-Layer Graphene	40
2.4.2	Effect of Gate Doping on the <i>G</i> Band of Double-Layer Graphene	42
2.5	Near-Field Raman Spectroscopy	44
2.5.1	The Spatial Resolution in Optical Microscopes	45
2.5.2	The Principle of TERS	45
2.5.3	Mechanism of Near-Field Enhancement	46
2.5.4	Application to Carbon Nanotubes	47
2.6	Summary and Perspective	49
	References	53
3	Scanning Tunneling Microscopy and Spectroscopy of Graphene	57
	Guohong Li and Eva Y. Andrei	
3.1	Introduction	57
3.2	STM/STS Techniques	58
3.3	Sample Preparation	61
3.4	Hallmarks of Graphene in STM/STS	61
3.5	Line Shape of Landau Levels	66
3.6	Electron-phonon Coupling	67
3.7	Coupling Between Graphene Layers	69
3.8	Twist Between Graphene Layers	71
3.8.1	Appearance of Moiré Pattern	72
3.8.2	Saddle Point Van Hove Singularities	73
3.8.3	Single Layer-like Behavior and Velocity Renormalization	73
3.9	Graphene on SiO ₂	77
3.9.1	Three Types of Corrugations	77
3.9.2	Scanning Tunneling Spectroscopy	79
3.9.3	Quantum Interference and Fermi Velocity	79
3.9.4	Trapped Charges in SiO ₂	80
3.10	Edges, Defects and Magnetism	81
3.11	SPM-based Nano-lithography	82
3.11.1	Signs of Invasiveness of an STM Tip	83
3.11.2	Folding Graphene Layers	83
3.11.3	Cutting Graphene Layers	84
3.11.4	Surface Modification	85
3.12	Summary and Perspectives	87
	References	88
4	The Electronic Properties of Adsorbates on Graphene	93
	Eli Rotenberg	
4.1	Introduction: What Are Adsorbates on Graphene Good for?	93
4.2	Angle-Resolved Photoemission Spectroscopy	96
4.2.1	Introduction	96

4.2.2	Band Structure Determination of Graphene	96
4.2.3	Self-energy Determination	99
4.3	The “Zoology” of Adsorbates	102
4.3.1	Adsorption of Nontransition-Metal Atoms	103
4.3.2	Adsorption of Transition Metal Atoms	107
4.4	Adsorbate–Graphene Interactions: General Symmetry Considerations	110
4.5	Hydrogen on Graphene As a Prototype Adsorbate System	112
4.5.1	Introduction	112
4.5.2	Hydrogen on Graphene: Experimental Evidence for Anderson Localization	114
4.6	Potassium on Graphene: The Coulomb Interaction in Graphene, Revealed	118
4.6.1	K Adsorption on Epitaxial Graphene on SiC(0001) ...	118
4.6.2	K Adsorption on Quasi-free-Standing Epitaxial Graphene on SiC(0001)	120
4.7	Calcium Adsorption: Superconducting Instability of Graphene	124
4.8	Conclusions and Outlook	128
	References	129
5	Epitaxial Graphene on SiC(0001)	135
	Thomas Seyller	
5.1	Introduction	135
5.2	Silicon Carbide and Its Polar Surfaces	137
5.3	Growth of Epitaxial Graphene on SiC(0001) in Ultra-High Vacuum	138
5.4	The $(6\sqrt{3} \times 6\sqrt{3})R30^\circ$ Reconstruction	140
5.5	Electronic Structure of Monolayer and Bilayer Graphene at the K-point	143
5.6	State-of-the Art Graphene Growth in Argon Atmosphere	146
5.7	Transport Properties of Graphene on SiC(0001)	149
5.8	Engineering the Interface Between Graphene and SiC(0001) by Hydrogen Intercalation	152
5.9	Conclusion	155
	References	155
6	Magneto-Transport on Epitaxial Graphene	161
	Peide D. Ye, Michael Capano, Tian Shen, Yanqing Wu, and Michael L. Bolen	
6.1	Introduction	161
6.2	Epitaxial Graphene Synthesis	163
6.3	Dielectric Integration on Epitaxial Graphene	168
6.4	Top-Gate Graphene Field-Effect Transistors	169
6.5	Half-Integer Quantum Hall-Effect in Epitaxial Graphene	172
6.6	Ballistic and Coherent Transport on Epitaxial Graphene	178

6.7	Spin Transport on Epitaxial Graphene	183
6.8	Summary	185
	References	185
7	Epitaxial Graphene on Metals	189
	Yuriy Dedkov, Karsten Horn, Alexei Preobrajenski, and Mikhail Fonin	
7.1	Introduction	189
7.2	Methods of Graphene Preparation on Metal Surfaces.....	193
7.3	Experimental Methods	194
7.4	Graphene on Lattice-Matched 3 <i>d</i> -Metal Surfaces	197
	7.4.1 Atomic Structure of Graphene Layer on Ni(111) and Co(0001)	198
	7.4.2 Electronic Structure of Graphene on Lattice-Matched Surfaces	200
	7.4.3 Magnetism of Graphene on the Ni(111) Surface	206
7.5	Graphene on Lattice-Mismatched 4 <i>d</i> , 5 <i>d</i> -Metal Surfaces	209
	7.5.1 Structure of Graphene on Ir(111), Ru(0001), and Rh(111)	210
	7.5.2 Electronic Structure of Graphene on Lattice-Mismatched Surfaces	214
7.6	Hybrid Structures on the Basis of Graphene Layers on Metal Surfaces	218
	7.6.1 Intercalation-like Systems	219
	7.6.2 Growth of Noble Metal Clusters on Graphene Moirè	222
	7.6.3 Growth of Magnetic Metal Clusters on Graphene Moirè	225
	7.6.4 Chemical Functionalization of Graphene on Transition Metal Surfaces	226
7.7	Conclusions and Outlook	228
	References	230

Part II Electronic-structure and Transport Properties

8	Electronic Properties of Monolayer and Bilayer Graphene	237
	Edward McCann	
8.1	Introduction	237
8.2	The Crystal Structure of Monolayer Graphene	238
	8.2.1 The Real Space Structure	238
	8.2.2 The Reciprocal Lattice of Graphene	239
	8.2.3 The Atomic Orbitals of Graphene	239
8.3	The Tight-Binding Model	240
8.4	The Tight-Binding Model of Monolayer Graphene	242
	8.4.1 Diagonal Matrix Elements	242
	8.4.2 Off-Diagonal Matrix Elements	244

8.4.3	The Low-Energy Electronic Bands of Monolayer Graphene	246
8.5	Massless Chiral Quasiparticles in Monolayer Graphene	248
8.5.1	The Dirac-Like Hamiltonian	248
8.5.2	Pseudospin and Chirality in Graphene	249
8.6	The Tight-Binding Model of Bilayer Graphene	251
8.7	Massive Chiral Quasiparticles in Bilayer Graphene	254
8.7.1	The Low-Energy Bands of Bilayer Graphene	254
8.7.2	The Two-Component Hamiltonian of Bilayer Graphene	255
8.7.3	Pseudospin and Chirality in Bilayer Graphene	256
8.8	The Integer Quantum Hall Effect in Graphene	258
8.8.1	The Landau Level Spectrum of Monolayer Graphene	258
8.8.2	The Integer Quantum Hall Effect in Monolayer Graphene	260
8.8.3	The Landau Level Spectrum of Bilayer Graphene	261
8.8.4	The Integer Quantum Hall Effect in Bilayer Graphene	262
8.9	Trigonal Warping in Graphene	263
8.9.1	Trigonal Warping in Monolayer Graphene	263
8.9.2	Trigonal Warping and Lifshitz Transition in Bilayer Graphene	264
8.10	Tuneable Band Gap in Bilayer Graphene	266
8.10.1	Asymmetry Gap in the Band Structure of Bilayer Graphene	266
8.10.2	Self-Consistent Model of Screening in Bilayer Graphene	268
8.11	Summary	272
	References	273
9	Electronic Properties of Graphene Nanoribbons	277
	Katsunori Wakabayashi	
9.1	Introduction	277
9.2	Electronic States of Graphene	279
9.2.1	Tight-Binding Model and Edge States	281
9.2.2	Massless Dirac Equation	284
9.2.3	Edge Boundary Condition and Intervalley Scattering	286
9.3	Electronic Transport Properties	287
9.3.1	One-Way Excess Channel System	288
9.3.2	Model of Impurity Potential	291
9.3.3	Perfectly Conducting Channel: Absence of Anderson Localization	291

9.4	Universality Class	293
9.4.1	Graphene Nanoribbons with Generic Edge Structures	294
9.5	Transport Properties Through Graphene Nanojunction	296
9.6	Summary	297
	References	298
10	Mesoscopics in Graphene: Dirac Points in Periodic Geometries	301
	H.A. Fertig and L. Brey	
10.1	Graphene Ribbons	303
10.1.1	Hamiltonian	303
10.1.2	Zigzag Nanoribbons	304
10.1.3	Armchair Nanoribbons	307
10.2	Graphene Quantum Rings	310
10.2.1	Chirality in Armchair Nanoribbons	311
10.2.2	Phase Jumps at Corner Junctions	312
10.2.3	Numerical Results	314
10.3	Graphene in a Periodic Potential	317
10.3.1	Counting Dirac Points	317
10.3.2	Numerical Solutions of the Dirac Equation	320
10.3.3	Conductivity	320
10.4	Conclusion	322
	References	322
11	Electronic Properties of Multilayer Graphene	325
	Hongki Min	
11.1	Introduction	325
11.1.1	Stacking Arrangements	326
11.1.2	π -Orbital Continuum Model	327
11.2	Energy Band Structure	327
11.2.1	Preliminaries	327
11.2.2	Monolayer Graphene	328
11.2.3	AA Stacking	329
11.2.4	AB Stacking	331
11.2.5	ABC Stacking	333
11.2.6	Arbitrary Stacking	334
11.3	Landau-Level Spectrum	336
11.3.1	Preliminaries	336
11.3.2	AA Stacking	336
11.3.3	AB Stacking	337
11.3.4	ABC Stacking	339
11.3.5	Arbitrary Stacking	339
11.4	Low-Energy Effective Theory	341
11.4.1	Introduction	341
11.4.2	Pseudospin Hamiltonian	341

11.4.3	Stacking Diagrams	342
11.4.4	Partitioning Rules	342
11.4.5	Degenerate State Perturbation Theory	344
11.4.6	Limitations of the Minimal Model	347
11.4.7	Effects of the Consecutive Stacking	347
11.5	Applications	348
11.5.1	Quantum Hall Conductivity	348
11.5.2	Optical Conductivity	350
11.5.3	Electrical Conductivity	351
11.6	Conclusions	354
	References	355
12	Graphene Carrier Transport Theory	357
	Shaffique Adam	
12.1	Introduction	357
12.2	Graphene Boltzmann Transport	360
12.2.1	Screening: Random Phase Approximation (RPA).....	362
12.2.2	Coulomb Scatterers	365
12.2.3	Gaussian White Noise Disorder	366
12.2.4	Yukawa Potential	367
12.2.5	Gaussian Correlated Impurities	367
12.2.6	Midgap States	368
12.3	Transport at Low Carrier Density	369
12.3.1	Self-Consistent Approximation	371
12.3.2	Effective Medium Theory	377
12.3.3	Magneto-Transport and Temperature Dependence of the Minimum Conductivity	381
12.3.4	Quantum to Classical Crossover	383
12.3.5	Summary of Theoretical Predictions for Coulomb Impurities.....	386
12.4	Comparison with Experiments	387
12.4.1	Magnetotransport: Dependence of σ_{xx} and σ_{xy} on Carrier Density	387
12.4.2	Dependence of σ_{\min} and Mobility on Impurity Concentration	389
12.4.3	Dependence of σ_{\min} and Mobility on Dielectric Environment	389
12.5	Conclusion	391
	References	392
13	Exploring Quantum Transport in Graphene Ribbons with Lattice Defects and Adsorbates	395
	George Kirczenow and Sjarhei Ihnatsenka	
13.1	Landauer Theory of Transport	397
13.2	Subband Structure and Transport in Ideal Ribbons	399
13.3	Quantized Ballistic Conductance	402

13.4	Electron Transport in Graphene Ribbons	403
13.5	Discovery of Quantized Conductance in Strongly Disordered Graphene Ribbons	404
13.6	The Roles of Different Classes of Defects	405
13.7	Tight Binding Model of Ribbons with Edge Disorder, Interior Vacancies, and Long-Ranged Potentials	406
13.8	Numerical Simulations of Quantum Transport	406
13.8.1	Disorder-Induced Conductance Suppression, Fluctuations and Destruction of the Ballistic Quantized Conductance Plateaus	408
13.8.2	Conductance Dips at the Edges of Ribbon Subbands	410
13.8.3	The Role of Temperature	411
13.8.4	From Ballistic Transport to Anderson Localization	412
13.8.5	The Quantized Conductance in Disordered Ribbons: Theory vs. Experiment	414
13.9	Adsorbates on Graphene and Dirac Point Resonances	416
13.9.1	Tight Binding Hamiltonian for Adsorbates on Graphene	417
13.9.2	Effective Hamiltonian for Adsorbates on Graphene ...	419
13.9.3	The T-matrix Formalism	420
13.9.4	Dirac Point Scattering Resonances due to H, F, and O Atoms and OH Molecules Adsorbed on Graphene	421
13.10	Electron Quantum Transport in Graphene Ribbons with Adsorbates	423
13.10.1	Building Efficient Tight-Binding Models	423
13.10.2	Results of Numerical Simulations of Quantum Transport in Ribbons with Adsorbates	426
13.11	Summary	431
	References	431
14	Graphene Oxide: Synthesis, Characterization, Electronic Structure, and Applications	435
	Derek A. Stewart and K. Andre Mkhoyan	
14.1	Introduction	436
14.2	Understanding Bulk Graphite Oxide and Graphene Oxide Monolayers	437
14.3	Fabrication of Graphite Oxide and Graphene Oxide	439
14.3.1	Traditional Approaches to Fabricate Graphite Oxide	440
14.3.2	New Fabrication Techniques for Graphite Oxide and Graphene Oxide	441

14.4 Characterization Approaches 444

14.4.1 Optical Microscopy 444

14.4.2 Scanning Transmission Electron Microscopy 445

14.4.3 Electron Energy Loss Spectroscopy 447

14.4.4 Atomic Force Microscopy 448

14.4.5 X-ray Photoelectron Spectroscopy 449

14.4.6 Raman Spectroscopy of Graphene Oxide
and Reduced Graphene 451

14.5 Insight from Simulations 452

14.5.1 Using Epoxy Groups to Unzip Graphene 452

14.5.2 Graphene Oxide Electronic Structure 454

14.5.3 Electron Mobility and Transport 455

14.6 Applications for Graphene Oxide 457

14.6.1 Graphene Oxide Electronics 457

14.6.2 Sensors 458

14.6.3 Carbon-Based Magnetism 458

14.7 Future Perspectives 459

References 460

Part III From Physics and Chemistry of Graphene to Device Applications

15 Graphene *pn* Junction: Electronic Transport and Devices 467

Tony Low

15.1 Introduction 467

15.2 Transport in the Absence of a Magnetic Field 469

15.2.1 Dirac Equation, Pseudospin, and Chirality 470

15.2.2 Abrupt *pn* Junction and Analogy with Optics 472

15.2.3 Tunneling for Dirac and Schrödinger Fermions 474

15.2.4 Quantum Transport Modeling 477

15.2.5 Experiments: Asymmetry and odd Resistances 479

15.3 Transport in the Presence of Magnetic Fields 482

15.3.1 Weak Magnetic Field Regime 482

15.3.2 Edge States, Snake States, and Valley Isospin 485

15.3.3 Quantum Hall Regime: The Ballistic Case 487

15.3.4 Experiments: Ballistic to Ohmic Transition 490

15.4 Transport in the Presence of Strain-Induced
Pseudo-Magnetic Fields 494

15.4.1 Strain-Induced Pseudo-Magnetic Field 494

15.4.2 Edge States and Transport Gap 497

15.4.3 Magnetic and Electric Snake States 501

15.5 Discussions 503

15.5.1 Devices: Current Status and Outlook 503

15.5.2 Conclusions 505

References 505

16	Electronic Structure of Bilayer Graphene Nanoribbon and Its Device Application: A Computational Study	509
	Kai-Tak Lam and Gengchiao Liang	
16.1	Introduction	509
16.2	Methodology	511
16.3	Electronic Structure of Monolayer Graphene Nanoribbon	512
16.3.1	Armchair Edges	512
16.3.2	Zigzag Edges	513
16.3.3	Dopant Effect	514
16.4	Electronic Structure of Bilayer Graphene Nanoribbon	516
16.4.1	Armchair Edges	517
16.4.2	Zigzag Edges with Dopants	518
16.4.3	Interlayer Distance	518
16.5	Bilayer Graphene Nanoribbon Device	519
16.6	Bilayer ZGNR NEM Switch	521
16.7	Conclusion	524
	References	525
17	Field-Modulation Devices in Graphene Nanostructures	529
	Hassan Raza	
17.1	Introduction	529
17.2	Electronic Structure	530
17.3	Theoretical Framework: Extended Hückel Theory	533
17.4	Bilayer Graphene	535
17.4.1	\bar{A} – B stacking	536
17.4.2	Strain Engineering	536
17.4.3	Misalignment	538
17.5	Armchair Graphene Nanoribbons	538
17.5.1	Pristine Edges	539
17.5.2	Periodic edge roughness effects	543
17.6	Zigzag Graphene Nanoribbons with Periodic Edge Roughness	546
17.7	Novel Applications	550
17.8	Conclusions	551
	References	552
18	Graphene Nanoribbons: From Chemistry to Circuits	555
	F. Tseng, D. Unluer, M.R. Stan, and A.W. Ghosh	
18.1	The Innermost Circle: The Atomistic View	556
18.1.1	Flatland: A Romance in Two Dimensions	557
18.1.2	Whither Metallicity?	558
18.1.3	Edge Chemistry: Benzene or Graphene?	559
18.1.4	Whither Chirality?	561
18.2	The Next Circle: Two Terminal Mobilities and I–Vs	563
18.2.1	Current–Voltage Characteristics (I–Vs)	563

18.2.2	Low Bias Mobility-Bandgap Tradeoffs: Asymptotic Band Constraints	566
18.3	The Third Level: Active Three-Terminal Electronics	569
18.3.1	Wide–Narrow–Wide: All Graphene Devices	569
18.3.2	Solving Quantum Transport and Electrostatic Equations	570
18.3.3	Improved Electrostatics in 2-D	571
18.3.4	Three-Terminal I–Vs	574
18.3.5	Pinning vs. Quasi-Ohmic Contacts	575
18.4	The Penultimate Circle: GNR Circuits.....	576
18.4.1	Geometry of An All Graphene Circuit.....	577
18.4.2	Compact Model Equations	579
18.4.3	Digital Circuits.....	579
18.4.4	How ‘Good’ is a Graphene-based Invertor?	580
18.4.5	Physical Domain Issues: Monolithic Device-Interconnect Structures	583
18.5	Conclusions	583
	References.....	585
	Index	587