

## Contents

**Preface** *xiii*

**List of Abbreviations** *xv*

- 1 Preparation of 2D Materials** *1*  
*Yue Tang and Hua Xu*
- 1.1 Mechanical Exfoliation of 2D Materials *2*
  - 1.2 Liquid-Phase Exfoliation of 2D Materials *4*
  - 1.3 Chemical Vapor Deposition Growth of 2D Materials *6*
  - 1.4 CVD Growth of Wafer-Scale Single Crystal 2D Materials *8*
  - 1.5 Thickness Control in CVD Growth of 2D Materials *10*
  - 1.6 Phase Control in CVD Growth of 2D Materials *12*
  - 1.7 Summary and Prospect *14*
  - References *15*
- 2 An Introduction to the Nonlinear Optical Properties of 2D Materials** *21*  
*Bolong Wang and Hao-Li Zhang*
- 2.1 Introduction *21*
  - 2.2 Nonlinear Optics of 2D Materials *22*
    - 2.2.1 SHG, THG, and HHG Setups *24*
    - 2.2.2 Four-Wave Mixing *26*
    - 2.2.3 Z-Scan Techniques *27*
    - 2.2.4 Nonlinear Optical Imaging *29*
    - 2.2.5 Pump-Probe Techniques *32*
  - 2.3 Application of 2D Nonlinear Materials *35*
    - 2.3.1 Optical Limiting *35*
    - 2.3.2 Q-Switched and Mode-Locked Lasers *38*
    - 2.3.3 Optical Switch and Modulation *40*
    - 2.3.4 Other Nonlinear Optical Phenomena *41*
  - 2.4 Prospect *43*
    - 2.4.1 Precise Fabrication and Functionalization of 2D Materials *43*
    - 2.4.2 High Resolution Spatiotemporal Characterizing Techniques *44*

- 2.4.3 New Physics of 2D Materials 44
- 2.4.4 Ever-Lasting Expansion of Applications 44
- Acknowledgment 45
- References 45

### **3 Modulation and Enhancement of Optical Nonlinearity in 2D Materials 55**

*Xinglin Wen and Qihua Xiong*

- 3.1 Introduction 55
- 3.2 Nonlinear Optics in 2D Materials 57
  - 3.2.1 Basics of Nonlinear Optics 57
  - 3.2.2 Parametric Nonlinear Optics 58
  - 3.2.3 Nonparametric Nonlinear Optics 59
- 3.3 Nonlinearity Modulation in 2D Materials 60
  - 3.3.1 Structural Engineering 60
    - 3.3.1.1 Layer Number 60
    - 3.3.1.2 Twisted Angle 62
    - 3.3.1.3 Stacking Order 64
    - 3.3.1.4 Edge States 65
  - 3.3.2 Optical Modulation 66
    - 3.3.2.1 Excitonic Effects Enhanced Nonlinearity 66
    - 3.3.2.2 Valley-Dependent Nonlinearity 69
    - 3.3.2.3 Optical Cavity-Enhanced Nonlinearity 71
    - 3.3.2.4 All-Optical Modulation 73
  - 3.3.3 Electrical Modulation 75
  - 3.3.4 Magnetic Modulation 78
  - 3.3.5 Perspectives 78
- Author Contributions 78
- Notes 79
- Acknowledgment 79
- References 79

### **4 Characterizing the Nonlinear Optical Properties of 2D Materials by Double 4f Nonlinear Imaging System with Phase Object and Four-Wave-Mixing Microscopy 87**

*Zhongguo Li and Yinglin Song*

- 4.1 Introduction 87
- 4.2 Principle of NLO Measurement Technique 88
  - 4.2.1 Nonlinear Optical Interaction in 2D Materials 88
    - 4.2.2 Typical NLO Measurement Method in 2D Material Research 89
      - 4.2.2.1 Self-Phase Modulation (SPM) 89
        - 4.2.2.2 Z-Scan 89
        - 4.2.2.3 Third-Harmonic Generation (THG) 90
        - 4.2.2.4 Two-Photon-Induced Photoluminescence (TPL) 90

4.3	Characterizing NLO Response of 2D Materials via Double 4f Nonlinear Imaging System with Phase Object (NIT-PO) Microscopy	91
4.4	Characterizing NLO Response of 2D Materials via Four-Wave-Mixing Microscopy	95
4.5	Outlook and Perspective	97
	References	98
<b>5</b>	<b>Ultrafast Carrier Dynamics in Emerging 2D Materials</b>	<b>103</b>
	<i>Jiawei Huang, Ningning Dong, and Jun Wang</i>	
5.1	Introduction	103
5.2	Ultrafast Time-Resolved Spectroscopy	104
5.2.1	Principle of Pump-Probe Measurement	104
5.2.2	Experiment for Ultrafast Spectroscopy	107
5.2.3	Fundamental Mechanism of Recombination Dynamics	107
5.3	Ultrafast Optics in van der Waals 2D Materials	109
5.3.1	Graphene	109
5.3.2	Transition Metal Dichalcogenides (TMDC)	109
5.3.3	Black Phosphorus	115
5.4	Ultrafast Optics in Emerging Quasi-2D Materials	118
5.4.1	2D Perovskite	118
5.4.2	Non-Layered 2D Platinum Sulfide	120
5.5	Perspectives on Ultrafast Optics for 2D Material	124
	Author Contributions	125
	Notes	125
	Acknowledgment	125
	References	125
<b>6</b>	<b>Transient Terahertz Spectroscopy for 2D Materials</b>	<b>131</b>
	<i>Jingyin Xu, Kai Zhang, Hong Li, and Tianwu Wang</i>	
6.1	Introduction	131
6.2	Generation and Detection of THz Radiation	132
6.2.1	Generation of THz Radiation Based on Nonlinear Optical Processes	132
6.2.1.1	Photoconductive Antennas (PCAs)	133
6.2.1.2	Electro-optic (EO) Crystals	135
6.2.1.3	Air Plasma	136
6.2.1.4	THz Emission of 2D Semiconductor Materials	140
6.2.2	Typical THz Time-Domain Spectroscopy Setup	141
6.2.2.1	Time-Domain THz Spectroscopy Under Pressure	143
6.2.3	Photoinduced Changes of the Dielectric Function in 2D via THz Spectroscopy	147
6.3	Nanoscale THz Scanning Probe Microscopy of 2D Materials	149
6.3.1	Scanning Near-Field THz Microscopy for Polariton in 2D Materials	150
6.3.2	THz-STM	151
6.4	Perspectives	154
6.4.1	Transient THz Spectroscopy	154

6.4.2	High-Resolution Spatial and Temporal THz Spectroscopies	155
	Acknowledgment	155
	References	155
<b>7</b>	<b>Graphene Glass for Nonlinear Optics</b>	<b>163</b>
	<i>Qi Xiao, Jingyu Sun, and Hao-Li Zhang</i>	
7.1	Light Absorption of Graphene	163
7.2	Nonlinear Optical Properties of Graphene	164
7.2.1	Third-Order Nonlinear Optical Properties of Graphene	164
7.2.2	Saturable Absorption of Graphene	165
7.2.3	High-Order Harmonic Generation of Graphene	165
7.3	Nonlinear Optical Properties of Graphene Glass	167
7.3.1	Graphene-Doped Glass	167
7.3.2	Graphene Glass Fabricated by Spin Coating	167
7.3.3	Graphene Glass Fabricated by Transfer Method	168
7.3.4	Directly Growth Graphene on Glass	168
7.3.4.1	Graphene Growth on High-Temperature-Resistant Glass	168
7.3.4.2	Graphene Growth on Molten Glass	171
7.3.4.3	Metal-Catalyst-Assisted Graphene Growth	173
7.3.4.4	Plasma-Enhanced Low-Temperature Graphene Growth	176
7.4	Perspectives	179
	Acknowledgment	180
	References	180
<b>8</b>	<b>2D Materials for Nonlinear Optical Limiting</b>	<b>185</b>
	<i>Wen Shang, Bolong Wang, and Qiang Wang</i>	
8.1	Introduction	185
8.2	Nonlinear Optical Limiting Mechanism	186
8.2.1	Reverse Saturable Absorption	186
8.2.2	Two-Photon/Multiphoton Absorption	187
8.2.3	Free Carrier Absorption	188
8.2.4	Nonlinear Refraction	188
8.2.5	Nonlinear Scattering	189
8.3	2D Materials for Optical Limiting	189
8.3.1	Graphene and Analogs	190
8.3.1.1	Graphene	190
8.3.1.2	Transition Metal Dichalcogenides	194
8.3.1.3	Black Phosphorus	196
8.3.1.4	Hexagonal Boron Nitride (h-BNS)	197
8.3.2	Novel 2D Materials	197
8.3.2.1	Antimonene	197
8.3.2.2	Tellurene	198
8.3.2.3	2D Perovskites	200
8.3.2.4	2D Metal–Organic Frameworks (2D-MOFs)	201
8.3.3	Materials with New Structures and Mechanism	203

8.3.3.1	Nonlinear Photonic Metamaterials	203
8.3.3.2	Plasmonic Effect-Enhanced Nonlinearity	205
8.3.4	Multifunctional Optical Limiting Devices	206
8.4	Conclusions and Prospects	209
	Acknowledgments	210
	References	210
<b>9</b>	<b>The Saturable Absorbers Based on 2D Materials</b>	<b>221</b>
	<i>Xin-Hai Yan, Lei Zhang, and Kai-Ge Zhou</i>	
9.1	Introduction	221
9.2	The Fundamentals in the 2D Materials-Based Saturable Absorbers	222
9.2.1	SA Theory	222
9.2.2	Slow- and Fast-Saturable Absorber	223
9.2.2.1	Slow-Saturable Absorber	224
9.2.2.2	Fast-Saturable Absorber	225
9.3	The Family of 2D Material-Based Saturable Absorbers	225
9.3.1	Mono-Elemental 2D Materials	226
9.3.1.1	Group IVA-Elemental 2D Materials	226
9.3.1.2	Group VA-Elemental 2D Materials	230
9.3.2	Dual-Elemental 2D Materials	234
9.3.2.1	TMDs	234
9.3.2.2	Topological Insulators (TIs)	235
9.3.3	Multi-Elemental 2D Materials	237
9.3.3.1	MOFs	237
9.3.3.2	MXenes	237
9.3.3.3	2D Perovskites	239
9.4	Applications	239
9.4.1	Fabrication of SA Device Based on 2D Materials	239
9.4.1.1	Scotch Tape Exfoliation	240
9.4.1.2	CVD, PVD	240
9.4.1.3	LPE	240
9.4.2	Q-Switched Laser	242
9.4.2.1	Q-Switched Fiber Laser	243
9.4.3	Mode-Locked Laser	246
9.5	Perspectives and Outlook	248
	Acknowledgment	249
	References	249
<b>10</b>	<b>Second-Harmonic and Third-Harmonic Generations in 2D Layered Materials</b>	<b>257</b>
	<i>Xudong Jin and Min Zhao</i>	
10.1	Introduction	257
10.1.1	Fundamentals of SHG and THG	258
10.1.2	2DLM-Based SHG	259
10.1.3	Graphene	260

10.1.4	Transition Metal Dichalcogenides	262
10.1.5	Other 2DLMs	266
10.1.6	2DLM-Based THG	268
10.1.6.1	Graphene	268
10.1.7	Transition Metal Dichalcogenides	268
10.1.7.1	Other 2DLMs	270
10.2	Conclusions and Outlook	274
	References	274
<b>11</b>	<b>2D Perovskites for Nanolasers</b>	<b>281</b>
	<i>Wei Yuan and Chuanjiang Qin</i>	
11.1	Introduction	281
11.2	Laser Formation Principle and Performance Parameters	283
11.2.1	Basic Optical Physical Process	283
11.2.2	Laser Formation Condition	283
11.2.3	Performance Parameters	284
11.2.4	Optical Cavities	285
11.3	The Application of 2D Perovskites in Nanolasers	286
11.3.1	Microplates	287
11.3.2	Microrods	287
11.3.3	Microcrystals	291
11.3.4	Nanowires	291
11.3.5	Microrings	293
11.3.6	Thin Films	294
11.3.7	Thin Flakes	298
11.4	Prospect	299
	References	300
<b>12</b>	<b>2D Materials for Space Use</b>	<b>303</b>
	<i>Shuyan Wang and Qiang Wang</i>	
12.1	Introduction	303
12.2	Space Radiation and Two-Dimensional Materials	305
12.2.1	Space Radiation Environment	305
12.2.2	2D Materials Under Irradiation	307
12.2.2.1	Electron Irradiation	307
12.2.2.2	Ion Irradiation	307
12.2.2.3	Proton Irradiation	308
12.2.2.4	$\gamma$ -Ray Irradiation	308
12.3	2D Materials for Space Use	309
12.3.1	Emerging Nonlinear Optical Applications	309
12.3.1.1	Laser Protection and Mode Locking	310
12.3.1.2	Other Nonlinear Phenomena	313
12.3.2	Space X-ray Detection and Imaging	314
12.3.3	Perovskite Solar Cells (PSCs)	316
12.3.4	2D Single-Photon Sources	317

12.3.5	Integrated Optoelectronic Platform	318
12.3.5.1	Field-Effect Transistor and Photodetectors	318
12.3.5.2	Sensors	320
12.3.6	Other Applications: Lubricants	320
12.4	Perspectives	321
	Acknowledgment	322
	References	322
	<b>Index</b>	333

