Contents

1 (Growtl	n of Self-Organized Quantum Dots	
JS.	Lee		1
1.1	Introd	uction	1
1.2	Fabric	ation Techniques of Quantum Dots	1
	1.2.1	Quantum Dot Fabrication by Lithographic Techniques	1
	1.2.2	Self-Organized Quantum Dot Fabrication	8
1.3	Orderi	ng of Three-Dimensional Islands	13
	1.3.1	Structural Characterization of Quantum Dots	13
	1.3.2	Ordering of Quantum Dot Position	20
1.4	Real-T	Time Monitoring	
	of Self	-Organized Quantum Dot Formation	34
	1.4.1	Reflection High-Energy Electron Diffraction	
		in Molecular Beam Epitaxy	35
	1.4.2	Optical in situ Measurement	
		in Metal-Organic Vapor-Phase Epitaxy	40
Refe	rences		55
2 I	Excito	nic Structures and Optical Properties	
of Q	uantu	m Dots	
Tosh	ihide T	akagahara	59
2.1	Introd	uction	59
2.2	Quant	um and Dielectric Confinement Effect	60
2.3	Nonlo	cal Response Theory of Radiative Decay Rate	
	of Exc	itons in Quantum Dots:	
	Size D	ependence and Temperature Dependence	70
	2.3.1	Formulation	70
	2.3.2	Size Dependence of Excitonic Radiative Decay Rate	77
	2.3.3	Effect of Homogeneous Broadening	
		on Excitonic Radiative Decay Rate	79
2.4	Electro	on–Hole Exchange Interaction	
	in Deg	renerate Valence Band Structures	81
	2.4.1	Formulation	82
	2.4.2	Exciton Doublet Structures	88
	2.4.3	Polarization Characteristics of Exciton Doublets	92

XII	Contents
	0 0 110 0 1100

2.5	Enhancement of Excitonic Optical Nonlinearity
	in Quantum Dot Arrays
	2.5.1 Exciton Band Structure in Quantum Dot Arrays
	2.5.2 Excitonic Optical Nonlinearity of Quantum Dot Arrays 102
	2.5.3 Tolerance Limits for the Fluctuation
	of Structure Parameters of the Quantum Dot Array 105
	2.5.4 The Polariton Effect and Photonic Band Structures 106
2.6	Summary
Appe	endix A. Expression of Depolarization Field
Appe	endix B. Depolarization Field in the Presence
	of a Background Dielectric Constant 108
Appe	endix C. Vector Spherical Harmonics 109
Appe	endix D. Parameter Related
	to the Electron–Hole Exchange Energies
Refei	rences
3 E	Electron–Phonon Interactions
in Se	emiconductor Quantum Dots
Tosh	ihide Takagahara 115
3.1	Introduction
3.2	Energy Spectra of Acoustic Phonon Modes
-	in Spherical Nanocrystals
	3.2.1 The Case of the Stress-Free Boundary Condition 117
	3.2.2 The Case of Smooth Contact Between a Quantum Dot
	and the Surrounding Medium
33	Derivation of the Electron–Acoustic-Phonon Interactions 123
3.4	Derivation of Electron–Polar-Ontical-Phonon Interaction
0.1	in Quantum Dots 126
3 5	A Formal Theory on the Exciton–Phonon System
0.0	Within the Franck-Condon Approximation 135
36	Luminosconce Stokes Shift and Huang-Blue Factor 138
3.0 3.7	Summerv 1/1
J.1 App	andix A Strain Tonsor Components
npp	in Congral Orthogonal Curvilingar Coordinates 141
App	andix B Vector Spherical Harmonics 144
Rofo	rendrix D. Vector Spherical Harmonics
neiei	ences
4 N	Vicro-Imaging and Single Dot Spectroscopy
of Se	elf-Assembled Quantum Dots
Mits	uru Sugisaki
4 1	Introduction 140
4.1	Here to Cot Access to a Simula One to D t
4.2	How to Get Access to a Single Quantum Dot
4.3	Observation Energy Dependence and Optical Anisotropy 157
	4.3.1 Mechanism for Optical Anisotropy 159
	4.3.2 Optical Anisotropy of Individual Quantum Dots 163

Contents	XIII
0011001100	

4.4	Many	Carrier Effects	. 165
	4.4.1	State Filling Effects Studied by Micro-Imaging	. 166
	4.4.2	Multiexciton States	. 168
	4.4.3	Biexciton Binding Energy	. 171
4.5	Tempe	erature Dependence	. 172
	4.5.1	Band Gap Energy Shift	. 172
	4.5.2	Thermal Activation	. 174
	4.5.3	Study of Thermal Activation	
		by Micro-Photoluminescence Images	. 174
4.6	Fluore	scence Intermittency	. 178
	4.6.1	Micro-Photoluminescence Images of Blinking Dots	. 179
	4.6.2	Random Telegraph Signals in Various Systems	. 180
	4.6.3	Random or Correlated?	. 181
	4.6.4	Excitation Power Dependence	. 183
	4.6.5	Origin of Fluorescence Intermittency	
		in InP Self-Assembled Dots	. 186
	4.6.6	Experimental Verification of the Model	. 187
4.7	Some	Other Interesting Phenomena	. 194
	4.7.1	External Electric Field Effects	. 194
	4.7.2	Magnetic Micro-Photoluminescence Spectra	. 196
	4.7.3	Fine Splitting by Anisotropic Strain	. 198
	4.7.4	Time Domain and Nonlinear Measurements	. 201
4.8	Summ	ary	. 202
Refe	rences		. 203
- 1	.		
5 1	Persist	ent Spectral Hole Burning	
in S	emicor	iductor Quantum Dots	200
Yasu	laki Ma	sumoto	. 209
5.1	Introd	uction	. 209
5.2	Precur	sor and Discovery	
	of the	Persistent Spectral Hole-Burning Phenomenon	. 211
5.3	Persist	ent Spectral Hole Burning, Hole Filling,	
	and T	heir Mechanism	. 212
5.4	Lumin	escence Hole Burning and Charged Exciton Complexes	. 223
5.5	Photos	stimulated Luminescence, Luminescence Blinking,	
	and Sr	pectral Diffusion	. 229
5.6	Applic	ation of Persistent Spectral Hole Burning	
	to Site	-Selective Spectroscopy	. 234
5.7	Summ	ary	. 240
Refe	rences	~ · · · · · · · · · · · · · · · · · · ·	. 241

XIV Contents

in Self-Assembled Quantum Dots Ivan V. Ignatiev and Igor E. Kozin	6 I	Dynamics of Carrier Relaxation	
Ivan V. Ignatiev and Igor E. Közin 245 6.1 Introduction 245 6.2 Experimental Details 253 6.3 Photoluminescence Spectra 253 6.4 Physical Mechanisms 262 6.4 Physical Mechanisms 262 6.4 Physical Mechanisms 262 6.4 Model of Selective Photoluminescence Quenching 265 6.5 Kinetics 273 6.6 Acoustic Phonon Resonances 273 6.7 Auger-Like Processes 280 6.8 Conclusion 288 References 290 7 Resonant Two-Photon Spectroscopy of Quantum Dots Alexander Baranov 295 7.1 Introduction 295 7.1 Introduction 295 7.2 Electronic Structure of CdS(Se) Quantum Dots 297 7.2.1 Two-Photon Absorption Techniques 298 7.2.2 The Line-Narrowing Technique 300 7.2.3 Analysis of RHRS and RSHS Excitation Spectra 301 7.3 Energy Structure of Low-Energy Confined Excitons 303 7.4.1 <t< th=""><th>in S</th><th>elf-Assembled Quantum Dots</th><th>0.45</th></t<>	in S	elf-Assembled Quantum Dots	0.45
6.1 Introduction 245 6.2 Experimental Details 253 6.3 Photoluminescence Spectra 253 6.4 Physical Mechanisms 262 6.4.1 Model of Selective Photoluminescence Quenching 265 6.5 Kinetics 269 6.6 Acoustic Phonon Resonances 273 6.7 Auger-Like Processes 280 7.8 References 290 7 Resonant Two-Photon Spectroscopy of Quantum Dots 295 7.1 Introduction 295 7.2 Electronic Structure of CdS(Se) Quantum Dots 297 7.2.1 Two-Photon Absorption Techniques 298 7.2.2 The Line-Narrowing Technique 300 7.2.3 Analysis of RHRS and RSHS Excitation Spectra 301 7.4 Exciton-Phonon Interaction in CuBr and CuCl Quantum Dots 303 7.4 Exciton-LO-Phonon Interaction 313 7.4.2 CuCl Quantum Dots: 301 7.4.2 CuCl Quantum Dots: Size Dependence 310 7.4.3 CuCl Quantum Dots: Size Dependence 313 <td>Ivan</td> <td>V. Ignatiev and Igor E. Kozin</td> <td>245</td>	Ivan	V. Ignatiev and Igor E. Kozin	245
6.2 Experimental Details 253 6.3 Photoluminescence Spectra 253 6.4 Physical Mechanisms 262 6.4.1 Model of Selective Photoluminescence Quenching 265 6.5 Kinetics 269 6.6 Acoustic Phonon Resonances 273 6.7 Auger-Like Processes 280 6.8 Conclusion 288 References 290 7 Resonant Two-Photon Spectroscopy of Quantum Dots Alexander Baranov 295 7.1 Introduction 295 7.2 Electronic Structure of CdS(Se) Quantum Dots 296 7.2.2 The Line-Narrowing Technique 300 7.2.3 Analysis of RHRS and RSHS Excitation Spectra 301 7.3 Energy Structure of Low-Energy Confined Excitons 303 7.4 Exciton-Phonon Interaction in CuBr and CuCl Quantum Dots 303 7.4.1 CuBr Quantum Dots: 303 7.4.2 CuCl Quantum Dots: Size Dependence 310 7.4.2 CuCl Quantum Dots: Size Dependence 313 7.4.3 CuCl Quantum D	6.1	Introduction	245
6.3 Photoluminescence Spectra in External Electric Field 256 6.4 Physical Mechanisms 262 6.4.1 Model of Selective Photoluminescence Quenching 265 6.5 Kinetics 269 6.6 Acoustic Phonon Resonances 273 6.7 Auger-Like Processes 280 6.8 Conclusion 288 References 290 7 Resonant Two-Photon Spectroscopy of Quantum Dots Alexander Baranov 295 7.1 Introduction 295 7.2 Electronic Structure of CdS(Se) Quantum Dots 297 7.2.2 The Line-Narrowing Technique 300 7.2.3 Analysis of RHRS and RSHS Excitation Spectra 301 7.3 Energy Structure of Low-Energy Confined Excitons 303 7.4 Exciton-Phonon Interaction in CuBr and CuCl Quantum Dots 303 7.4.1 CuBr Quantum Dots: 303 7.4.2 CuCl Quantum Dots: 303 7.4 Exciton-LO-Phonon Interaction 313 7.4.3 CuCl Quantum Dots: Size Dependence 310 7.4.3	6.2	Experimental Details	253
in External Electric Field	6.3	Photoluminescence Spectra	
6.4 Physical Mechanisms 262 6.4.1 Model of Selective Photoluminescence Quenching 265 6.5 Kinetics 269 6.6 Acoustic Phonon Resonances 273 6.7 Auger-Like Processes 280 6.8 Conclusion 288 References 290 7 Resonant Two-Photon Spectroscopy of Quantum Dots Alexander Baranov 295 7.1 Introduction 295 7.2 Electronic Structure of CdS(Se) Quantum Dots 297 7.2.2 The Line-Narrowing Technique 290 7.3 Energy Structure of Low-Energy Confined Excitans 301 7.4 Exciton-Phonon Interaction in CuBr and CuCl Quantum Dots 303 7.4 Exciton-Phonon Interaction in CuBr and CuCl Quantum Dots 310 7.4.1 CuBr Quantum Dots: 310 7.4.2 CuCl Quantum Dots: 313 7.4.3 CuCl Quantum Dots: 316 7.5 Determination of the Orientation of CuCl Nanocrystals 316 7.4 Single Nanocrystal Luminescence by Two-Photon Excitation 322		in External Electric Field	256
6.4.1 Model of Selective Photoluminescence Quenching 265 6.5 Kinetics 269 6.6 Acoustic Phonon Resonances 273 6.7 Auger-Like Processes 280 6.8 Conclusion 288 References 290 7 Resonant Two-Photon Spectroscopy of Quantum Dots Alexander Baranov 295 7.1 Introduction 295 7.2 Electronic Structure of CdS(Se) Quantum Dots 297 7.2.1 Two-Photon Absorption Techniques 298 7.2.2 The Line-Narrowing Technique 300 7.3 Energy Structure of Low-Energy Confined Excitans 301 7.3 Energy Structure of Low-Energy Confined Excitons 303 7.4 Exciton-Phonon Interaction in CuBr and CuCl Quantum Dots 310 7.4.1 CuBr Quantum Dots: 310 7.4.2 CuCl Quantum Dots: 312 7.4.3 CuCl Quantum Dots: 313 7.4.4 CuCl Quantum Dots: 316 7.5.5 Determination of the Orientation of CuCl Nanocrystals 316 7.5 D	6.4	Physical Mechanisms	262
6.5 Kinetics 269 6.6 Acoustic Phonon Resonances 273 6.7 Auger-Like Processes 280 6.8 Conclusion 288 References 290 7 Resonant Two-Photon Spectroscopy of Quantum Dots Alexander Baranov 295 7.1 Introduction 295 7.2 Electronic Structure of CdS(Se) Quantum Dots 297 7.2.1 Two-Photon Absorption Techniques 298 7.2.2 The Line-Narrowing Technique 300 7.2.3 Analysis of RHRS and RSHS Excitation Spectra 301 7.3 Energy Structure of Low-Energy Confined Excitons 303 7.4 Exciton-Phonon Interaction in CuBr and CuCl Quantum Dots 310 7.4.1 CuBr Quantum Dots: 310 7.4.2 CuCl Quantum Dots: Size Dependence 313 7.4.3 CuCl Quantum Dots: Softening of LO Phonons 316 7.5 Determination of the Orientation of CuCl Nanocrystals 322 in a NaCl Matrix 320 322 7.4 Single Nanocrystal Luminescence by Two-Photon Excitation 321		6.4.1 Model of Selective Photoluminescence Quenching	265
6.6 Acoustic Phonon Resonances 273 6.7 Auger-Like Processes 280 6.8 Conclusion 288 References 290 7 Resonant Two-Photon Spectroscopy of Quantum Dots Alexander Baranov 295 7.1 Introduction 295 7.2 Electronic Structure of CdS(Se) Quantum Dots 297 7.2.1 Two-Photon Absorption Techniques 298 7.2.2 The Line-Narrowing Technique 300 7.2.3 Analysis of RHRS and RSHS Excitation Spectra 301 7.3 Energy Structure of Low-Energy Confined Excitons 303 7.4 Exciton-Phonon Interaction in CuBr and CuCl Quantum Dots 310 7.4.1 CuBr Quantum Dots: 310 7.4.2 CuCl Quantum Dots: Size Dependence 310 7.4.3 CuCl Quantum Dots: Softening of LO Phonons 316 7.5 Determination of the Orientation of CuCl Nanocrystals 320 7.6 Single Nanocrystal Luminescence by Two-Photon Excitation 321 7.7 Conclusion 322 8 Homogeneous Width of Confined Excitons	6.5	Kinetics	269
6.7 Auger-Like Processes 280 6.8 Conclusion 288 References 290 7 Resonant Two-Photon Spectroscopy of Quantum Dots Alexander Baranov 295 7.1 Introduction 295 7.2 Electronic Structure of CdS(Se) Quantum Dots 297 7.2.1 Two-Photon Absorption Techniques 298 7.2.2 The Line-Narrowing Technique 300 7.2.3 Analysis of RHRS and RSHS Excitation Spectra 301 7.3 Energy Structure of Low-Energy Confined Excitons 303 7.4 Exciton-Phonon Interaction in CuBr and CuCl Quantum Dots 310 7.4.1 CuBr Quantum Dots: 310 7.4.2 CuCl Quantum Dots: Size Dependence of the Exciton-LO-Phonon Interaction 313 7.4.2 CuCl Quantum Dots: Size Dependence 310 7.4.3 CuCl Quantum Dots: Size Dependence 310 7.4.3 CuCl Quantum Dots: Softening of LO Phonons 316 7.5 Determination of the Orientation of CuCl Nanocrystals 320 7.6 Single Nanocrystal Luminescence by Two-Photon Excitatio	6.6	Acoustic Phonon Resonances	273
6.8 Conclusion 288 References 290 7 Resonant Two-Photon Spectroscopy of Quantum Dots Alexander Baranov 295 7.1 Introduction 295 7.2 Electronic Structure of CdS(Se) Quantum Dots 297 7.2.1 Two-Photon Absorption Techniques 298 7.2.2 The Line-Narrowing Technique 300 7.3 Energy Structure of Low-Energy Confined Excitons 301 7.3 Energy Structure of Low-Energy Confined Excitons 303 7.4 Exciton-Phonon Interaction in CuBr and CuCl Quantum Dots 310 7.4.1 CuBr Quantum Dots: 310 7.4.2 CuCl Quantum Dots: 310 7.4.3 CuCl Quantum Dots: 310 7.4.3 CuCl Quantum Dots: 313 7.4.3 CuCl Quantum Dots: 316 7.5 Determination of the Orientation of CuCl Nanocrystals 320 7.6 Single Nanocrystal Luminescence by Two-Photon Excitation 321 7.7 Conclusion 322 8 Homogeneous Width of Confined Excitons 322 8<	6.7	Auger-Like Processes	280
References 290 7 Resonant Two-Photon Spectroscopy of Quantum Dots 295 Alexander Baranov 295 7.1 Introduction 295 7.2 Electronic Structure of CdS(Se) Quantum Dots 297 7.2.1 Two-Photon Absorption Techniques 298 7.2.2 The Line-Narrowing Technique 300 7.2.3 Analysis of RHRS and RSHS Excitation Spectra 301 7.3 Energy Structure of Low-Energy Confined Excitons 303 7.4 Exciton-Phonon Interaction in CuBr and CuCl Quantum Dots 310 7.4.1 CuBr Quantum Dots: 310 7.4.2 CuCl Quantum Dots: Size Dependence 310 7.4.3 CuCl Quantum Dots: Softening of LO Phonons 313 7.4.3 CuCl Quantum Dots: Softening of LO Phonons 316 7.5 Determination of the Orientation of CuCl Nanocrystals 322 8 Homogeneous Width of Confined Excitons 322 8 Homogeneous Width of Confined Excitons 322 8 1 Dutatum Dots – Experimental 325	6.8	Conclusion	288
7 Resonant Two-Photon Spectroscopy of Quantum Dots Alexander Baranov 295 7.1 Introduction 295 7.2 Electronic Structure of CdS(Se) Quantum Dots 297 7.2.1 Two-Photon Absorption Techniques 298 7.2.2 The Line-Narrowing Technique 300 7.2.3 Analysis of RHRS and RSHS Excitation Spectra 301 7.3 Energy Structure of Low-Energy Confined Excitons 303 7.4 Exciton-Phonon Interaction in CuBr and CuCl Quantum Dots 310 7.4.1 CuBr Quantum Dots: 310 7.4.2 CuCl Quantum Dots: 310 7.4.3 CuCl Quantum Dots: 310 7.4.4.3 CuCl Quantum Dots: Size Dependence 310 7.4.3 CuCl Quantum Dots: Softening of LO Phonons 316 7.5 Determination of the Orientation of CuCl Nanocrystals 320 7.6 Single Nanocrystal Luminescence by Two-Photon Excitation 321 7.7 Conclusion 322 8 Homogeneous Width of Confined Excitons 322 8 Homogeneous Width of Confined Excitons 325	Refe	rences	290
Alexander Baranov 295 7.1 Introduction 295 7.2 Electronic Structure of CdS(Se) Quantum Dots 297 7.2.1 Two-Photon Absorption Techniques 298 7.2.2 The Line-Narrowing Technique 300 7.2.3 Analysis of RHRS and RSHS Excitation Spectra 301 7.3 Energy Structure of Low-Energy Confined Excitons 303 7.4 Exciton-Phonon Interaction in CuBr and CuCl Quantum Dots 310 7.4.1 CuBr Quantum Dots: 303 7.4.2 CuCl Quantum Dots: 310 7.4.3 CuCl Quantum Dots: 310 7.4.4.3 CuCl Quantum Dots: Size Dependence 313 7.4.3 CuCl Quantum Dots: Softening of LO Phonons 316 7.5 Determination of the Orientation of CuCl Nanocrystals 320 7.6 Single Nanocrystal Luminescence by Two-Photon Excitation 321 7.7 Conclusion 322 8 Homogeneous Width of Confined Excitons 322 8 Homogeneous Width of Confined Excitons 325 9.1 Intro ducting 325	7 1	Resonant Two-Photon Spectroscopy of Quantum Dots	
7.1 Introduction 295 7.2 Electronic Structure of CdS(Se) Quantum Dots 297 7.2.1 Two-Photon Absorption Techniques 298 7.2.2 The Line-Narrowing Technique 300 7.3 Energy Structure of Low-Energy Confined Excitons 301 7.3 Energy Structure of Low-Energy Confined Excitons 303 7.4 Exciton-Phonon Interaction in CuBr and CuCl Quantum Dots 310 7.4.1 CuBr Quantum Dots: 310 7.4.2 CuCl Quantum Dots: 310 7.4.2 CuCl Quantum Dots: 310 7.4.3 CuCl Quantum Dots: 310 7.4.4 CuCl Quantum Dots: Size Dependence 313 7.4.3 CuCl Quantum Dots: Softening of LO Phonons 316 7.5 Determination of the Orientation of CuCl Nanocrystals 320 7.6 Single Nanocrystal Luminescence by Two-Photon Excitation 321 7.7 Conclusion 322 8 Homogeneous Width of Confined Excitons 322 8 Homogeneous Width of Confined Excitons 325 9.1 Intreduction 325 <	Alex	ander Baranov	295
7.1 Introduction 295 7.2 Electronic Structure of CdS(Se) Quantum Dots 297 7.2.1 Two-Photon Absorption Techniques 298 7.2.2 The Line-Narrowing Technique 300 7.2.3 Analysis of RHRS and RSHS Excitation Spectra 301 7.3 Energy Structure of Low-Energy Confined Excitons 303 7.4 Exciton-Phonon Interaction in CuBr and CuCl Quantum Dots 310 7.4.1 CuBr Quantum Dots: 310 7.4.2 CuCl Quantum Dots: 310 7.4.3 CuCl Quantum Dots: Size Dependence 310 7.4.3 CuCl Quantum Dots: Softening of LO Phonons 316 7.4.3 CuCl Quantum Dots: Softening of LO Phonons 316 7.5 Determination of the Orientation of CuCl Nanocrystals 320 7.6 Single Nanocrystal Luminescence by Two-Photon Excitation 321 7.7 Conclusion 322 8 Homogeneous Width of Confined Excitons 322 8 Homogeneous Width of Confined Excitons 325 9.1 Introduction 325	THOA		200
7.2 Electronic Structure of CdS(Se) Quantum Dots 297 7.2.1 Two-Photon Absorption Techniques 298 7.2.2 The Line-Narrowing Technique 300 7.2.3 Analysis of RHRS and RSHS Excitation Spectra 301 7.3 Energy Structure of Low-Energy Confined Excitons 303 7.4 Exciton-Phonon Interaction in CuBr and CuCl Quantum Dots 303 7.4 Exciton-Phonon Interaction in CuBr and CuCl Quantum Dots 310 7.4.1 CuBr Quantum Dots: 310 7.4.2 CuCl Quantum Dots: Size Dependence 310 7.4.3 CuCl Quantum Dots: Softening of LO Phonons 316 7.4.3 CuCl Quantum Dots: Softening of LO Phonons 316 7.5 Determination of the Orientation of CuCl Nanocrystals 320 7.6 Single Nanocrystal Luminescence by Two-Photon Excitation 322 8 Homogeneous Width of Confined Excitons 322 8 Homogeneous Width of Confined Excitons 325 9.1 Introduction 325	7.1	Introduction	295
7.2.1 Two-Photon Absorption Techniques 298 7.2.2 The Line-Narrowing Technique 300 7.2.3 Analysis of RHRS and RSHS Excitation Spectra 301 7.3 Energy Structure of Low-Energy Confined Excitons 303 7.4 Exciton-Phonon Interaction in CuBr and CuCl Quantum Dots 303 7.4 Exciton-Phonon Interaction in CuBr and CuCl Quantum Dots 310 7.4.1 CuBr Quantum Dots: 310 7.4.2 CuCl Quantum Dots: Size Dependence 310 7.4.3 CuCl Quantum Dots: Softening of LO Phonons 316 7.4.3 CuCl Quantum Dots: Softening of LO Phonons 316 7.5 Determination of the Orientation of CuCl Nanocrystals 320 7.6 Single Nanocrystal Luminescence by Two-Photon Excitation 322 8 Homogeneous Width of Confined Excitons 322 8 Homogeneous Width of Confined Excitons 325 9.1 Intro duction 325	7.2	Electronic Structure of CdS(Se) Quantum Dots	297
7.2.2 The Line-Narrowing Technique 300 7.2.3 Analysis of RHRS and RSHS Excitation Spectra 301 7.3 Energy Structure of Low-Energy Confined Excitons 303 7.4 Exciton-Phonon Interaction in CuBr and CuCl Quantum Dots 303 7.4 Exciton-Phonon Interaction in CuBr and CuCl Quantum Dots 310 7.4.1 CuBr Quantum Dots: Coupled Exciton-LO-Phonon States 310 7.4.2 CuCl Quantum Dots: Size Dependence 313 7.4.3 CuCl Quantum Dots: Softening of LO Phonons 316 7.5 Determination of the Orientation of CuCl Nanocrystals 320 7.6 Single Nanocrystal Luminescence by Two-Photon Excitation 321 7.7 Conclusion 322 8 Homogeneous Width of Confined Excitons 322 8 Homogeneous Width of Confined Excitons 325 9.1 Leter duction 325		7.2.1 Two-Photon Absorption Techniques	298
7.2.3 Analysis of RHRS and RSHS Excitation Spectra		7.2.2 The Line-Narrowing Technique	300
7.3 Energy Structure of Low-Energy Confined Excitons in CuCl Quantum Dots 303 7.4 Exciton–Phonon Interaction in CuBr and CuCl Quantum Dots 310 7.4.1 CuBr Quantum Dots: Coupled Exciton–LO-Phonon States 310 7.4.2 CuCl Quantum Dots: Size Dependence of the Exciton–LO-Phonon Interaction 313 7.4.3 CuCl Quantum Dots: Softening of LO Phonons in the Presence of an Exciton 316 7.5 Determination of the Orientation of CuCl Nanocrystals in a NaCl Matrix 320 7.6 Single Nanocrystal Luminescence by Two-Photon Excitation 321 7.7 Conclusion 322 8 Homogeneous Width of Confined Excitons in Quantum Dots – Experimental 325 8 I Interchection 325		7.2.3 Analysis of RHRS and RSHS Excitation Spectra	301
 in CuCl Quantum Dots	7.3	Energy Structure of Low-Energy Confined Excitons	
7.4 Exciton-Phonon Interaction in CuBr and CuCl Quantum Dots		in CuCl Quantum Dots	303
7.4.1 CuBr Quantum Dots: Coupled Exciton–LO-Phonon States 310 7.4.2 CuCl Quantum Dots: Size Dependence of the Exciton–LO-Phonon Interaction 313 7.4.3 CuCl Quantum Dots: Softening of LO Phonons in the Presence of an Exciton 316 7.5 Determination of the Orientation of CuCl Nanocrystals in a NaCl Matrix 320 7.6 Single Nanocrystal Luminescence by Two-Photon Excitation 321 7.7 Conclusion 322 8 Homogeneous Width of Confined Excitons in Quantum Dots – Experimental 325 9.1 Intercharting 325	7.4	Exciton–Phonon Interaction in CuBr and CuCl Quantum Dots	310
Coupled Exciton-LO-Phonon States 310 7.4.2 CuCl Quantum Dots: Size Dependence of the Exciton-LO-Phonon Interaction 313 7.4.3 CuCl Quantum Dots: Softening of LO Phonons in the Presence of an Exciton 316 7.5 Determination of the Orientation of CuCl Nanocrystals in a NaCl Matrix 320 7.6 Single Nanocrystal Luminescence by Two-Photon Excitation 321 7.7 Conclusion 322 8 Homogeneous Width of Confined Excitons in Quantum Dots – Experimental 325 8.1 Interclustion 325		7.4.1 CuBr Quantum Dots:	
7.4.2 CuCl Quantum Dots: Size Dependence of the Exciton-LO-Phonon Interaction 313 7.4.3 CuCl Quantum Dots: Softening of LO Phonons in the Presence of an Exciton 316 7.5 Determination of the Orientation of CuCl Nanocrystals in a NaCl Matrix 320 7.6 Single Nanocrystal Luminescence by Two-Photon Excitation 321 7.7 Conclusion 322 8 Homogeneous Width of Confined Excitons in Quantum Dots – Experimental 325 8.1 Interclustion 325		Coupled Exciton–LO-Phonon States	310
of the Exciton-LO-Phonon Interaction 313 7.4.3 CuCl Quantum Dots: Softening of LO Phonons in the Presence of an Exciton 316 7.5 Determination of the Orientation of CuCl Nanocrystals in a NaCl Matrix 320 7.6 Single Nanocrystal Luminescence by Two-Photon Excitation 321 7.7 Conclusion 322 References 322 8 Homogeneous Width of Confined Excitons in Quantum Dots - Experimental 325 9.1 Interclustion 325		7.4.2 CuCl Quantum Dots: Size Dependence	010
7.4.3 CuCl Quantum Dots: Softening of LO Phonons in the Presence of an Exciton		of the Exciton–LO-Phonon Interaction	313
7.5 Determination of the Orientation of CuCl Nanocrystals in a NaCl Matrix 320 7.6 Single Nanocrystal Luminescence by Two-Photon Excitation 321 7.7 Conclusion 322 References 322 8 Homogeneous Width of Confined Excitons in Quantum Dots - Experimental Yasuaki Masumoto 325 9.1 Interclustion 325		in the Dreamer of an Erection	916
7.5 Determination of the Orientation of CuCl Nanocrystals in a NaCl Matrix 320 7.6 Single Nanocrystal Luminescence by Two-Photon Excitation 321 7.7 Conclusion 322 References 322 8 Homogeneous Width of Confined Excitons in Quantum Dots – Experimental 325 8.1 Interclustion 325	75	Determination of the Orientation of CuCl Nancountrals	510
7.6 Single Nanocrystal Luminescence by Two-Photon Excitation 321 7.7 Conclusion 322 References 322 8 Homogeneous Width of Confined Excitons in Quantum Dots – Experimental 325 9.1 Interclustion 325	1.5	in a NaCl Matrix	220
7.0 Single Nanocrystal Eulinnescence by Two-Thoton Excitation	76	Single Nancarustal Luminesconce by Two Photon Excitation	320
8 Homogeneous Width of Confined Excitons in Quantum Dots – Experimental Yasuaki Masumoto 225 22	7.0	Conclusion	041 200
8 Homogeneous Width of Confined Excitons in Quantum Dots – Experimental Yasuaki Masumoto 225 9 1 Interventering	1.1 Pofe		344 200
8 Homogeneous Width of Confined Excitons in Quantum Dots – Experimental Yasuaki Masumoto 325 9.1 Interclustion 325	nele	Tences	344
in Quantum Dots – Experimental Yasuaki Masumoto	8 I	Homogeneous Width of Confined Excitons	
Yasuaki Masumoto	in Q	Quantum Dots – Experimental	
9.1 Interaduation 201	Yasu	aki Masumoto	325
\mathbf{X} Introduction 375	81	Introduction	30¤
8.2 Spectral Hole Burning and Fluorescence Line Narrowing 396	8.2	Spectral Hole Burning and Fluorescence Line Narrowing	320 326
8.3 Single Quantum Dot Spectroscopy 327	8.3	Single Quantum Dot Spectroscopy	327
8.4 Photon Echo	8.4	Photon Echo	329

	Contents	XV
8.5	Accumulated Photon Echo	330 330
	Application to Quantum Dots 8.5.3 Accumulated Photon Echo Signal	333
	and the Homogeneous Width of CuCl Quantum Dots 8.5.4 Accumulated Photon Echo Signal	334
	and the Homogeneous Width of CdSe Quantum Dots 8.5.5 Lowest-Temperature Accumulated Photon Echo Signal	339
	and Homogeneous Width8.5.6 Summary of the Accumulated Photon Echo	342
	of Quantum Dots	346
8.6	Coherency Measurements	346
Refe	rences	349
9] in Se	Theory of Exciton Dephasing emiconductor Quantum Dots	
Tosh	ihide Takagahara	353
9.1	Introduction	353
9.2	Green Function Formalism of Exciton Dephasing Rate	354
9.3	Exciton–Phonon Interactions	361
9.4	Excitons in Anisotropic Quantum Disks	362
9.5	Temperature–Dependence of the Exciton Dephasing Bate	365
9.6	Elementary Processes of Exciton Pure Dephasing	369
0.7	Mechanisms of Population Decay of Excitons	370
9.1	0.7.1 Phonon Assisted Population Belayation	370
	9.7.1 Filonon-Assisted Fogulation Relaxation	370 971
0.0	9.7.2 Phonon-Assisted Exciton Migration	371
9.8	Correlation Between Temperature Dependence	
	of Exciton Dephasing Rate	~
0.0	and Strength of Quantum Confinement	377
$9.9 \\ 9.10$	Polarization Relaxation of Excitons Photoluminescence Spectrum	378
	under Selective Excitation	383
9.11	Summary and Discussion	386
Refe	rences	387
10 and	Excitonic Optical Nonlinearity Weakly Correlated Exciton-Pair States	
Selva	kumar V. Nair and Toshihide Takagahara	389
10.1	Introduction	389
10.2	Exciton States	390
	10.2.1 Formulation	391
	10.2.2 Configuration Interaction in a Truncated Basis $\ldots \ldots \ldots$	393

10.2.3 Variational Approach
10.2.4 Kayanuma's Correlated Basis Set
10.3 Biexciton States
10.3.1 Variational Approach
10.3.2 Exciton-Exciton Product State Basis
10.3.3 Electron–Hole Exchange Interaction
10.4 Exciton and Biexciton Energy Levels: The Case of CuCl 402
10.5 Transition Dipole Moments
10.5.1 Formulation
10.5.2 Results for CuCl 409
10.6 Weakly Correlated Exciton Pair States
10.7 Nonlinear Optical Properties 415
10.7.1 Size Dependence
of the Third-Order Nonlinear Susceptibility
10.7.2 Excited State Absorption from the Exciton Ground State 421
10.7.3 Experimental Observation
of the Weakly Correlated Exciton Pair States
10.7.4 Recent Progress in Nonlinear Nano-Optics
10.8 Summary and Conclusions
Appendix A. Two-Particle States with $L = 1, 2, \dots, 430$
Appendix B. Electron–Hole Exchange Interaction
Keierences
11 Coulomb Effects in the Optical Spectra
11 Coulomb Effects in the Optical Spectra of Highly Excited Semiconductor Quantum Dots
11Coulomb Effects in the Optical Spectraof Highly Excited Semiconductor Quantum DotsSelvakumar V. Nair439
11 Coulomb Effects in the Optical Spectra of Highly Excited Semiconductor Quantum Dots Selvakumar V. Nair 11 L. Line Lettice
11 Coulomb Effects in the Optical Spectra of Highly Excited Semiconductor Quantum Dots Selvakumar V. Nair 439 11.1 Introduction 439 11.2 Log ID 449
11 Coulomb Effects in the Optical Spectra of Highly Excited Semiconductor Quantum Dots Selvakumar V. Nair 439 11.1 Introduction 439 11.2 Local Density Approximation for Electrons and Holes 440
11 Coulomb Effects in the Optical Spectra of Highly Excited Semiconductor Quantum Dots Selvakumar V. Nair 439 11.1 Introduction 439 11.2 Local Density Approximation for Electrons and Holes 440 11.3 Application of the Local Density Approximation 441
11 Coulomb Effects in the Optical Spectra of Highly Excited Semiconductor Quantum Dots Selvakumar V. Nair 439 11.1 Introduction 439 11.2 Local Density Approximation for Electrons and Holes 440 11.3 Application of the Local Density Approximation to Quantum Dots 441
11 Coulomb Effects in the Optical Spectra of Highly Excited Semiconductor Quantum Dots Selvakumar V. Nair 439 11.1 Introduction 439 11.2 Local Density Approximation for Electrons and Holes 440 11.3 Application of the Local Density Approximation to Quantum Dots 441 11.3.1 Spherical Approximation 442 11.3.2 Coline Local Density Approximation 442
11 Coulomb Effects in the Optical Spectra of Highly Excited Semiconductor Quantum Dots Selvakumar V. Nair 439 11.1 Introduction 439 11.2 Local Density Approximation for Electrons and Holes 440 11.3 Application of the Local Density Approximation 441 11.3.1 Spherical Approximation 442 11.3.2 Cylindrical Quantum Dots 446
11 Coulomb Effects in the Optical Spectra of Highly Excited Semiconductor Quantum Dots Selvakumar V. Nair 439 11.1 Introduction 439 11.2 Local Density Approximation for Electrons and Holes 440 11.3 Application of the Local Density Approximation 441 11.3.1 Spherical Approximation 442 11.3.2 Cylindrical Quantum Dots 446 11.4 Beyond the Local Density Approximation: 440
11 Coulomb Effects in the Optical Spectra of Highly Excited Semiconductor Quantum Dots Selvakumar V. Nair 439 11.1 Introduction 439 11.2 Local Density Approximation for Electrons and Holes 440 11.3 Application of the Local Density Approximation 441 11.3.1 Spherical Approximation 442 11.3.2 Cylindrical Quantum Dots 446 11.4 Beyond the Local Density Approximation: 5 Spectral Broadening and Relaxation by Coulomb Scattering 448
11 Coulomb Effects in the Optical Spectra of Highly Excited Semiconductor Quantum Dots Selvakumar V. Nair 439 11.1 Introduction 439 11.2 Local Density Approximation for Electrons and Holes 440 11.3 Application of the Local Density Approximation to Quantum Dots 441 11.3.1 Spherical Approximation 442 11.3.2 Cylindrical Quantum Dots 446 11.4 Beyond the Local Density Approximation: Spectral Broadening and Relaxation by Coulomb Scattering 448 11.5 Spin Fine Structure of a Few Exciton Spectra: 440
11 Coulomb Effects in the Optical Spectra of Highly Excited Semiconductor Quantum Dots Selvakumar V. Nair 439 11.1 Introduction 439 11.2 Local Density Approximation for Electrons and Holes 440 11.3 Application of the Local Density Approximation 441 11.3.1 Spherical Approximation 442 11.3.2 Cylindrical Quantum Dots 446 11.4 Beyond the Local Density Approximation: 5pectral Broadening and Relaxation by Coulomb Scattering 448 11.5 Spin Fine Structure of a Few Exciton Spectra: 450 11.6 Configuration Interaction Approach 450
11 Coulomb Effects in the Optical Spectra of Highly Excited Semiconductor Quantum Dots Selvakumar V. Nair 439 11.1 Introduction 439 11.2 Local Density Approximation for Electrons and Holes 440 11.3 Application of the Local Density Approximation 441 11.3.1 Spherical Approximation 442 11.3.2 Cylindrical Quantum Dots 446 11.4 Beyond the Local Density Approximation: 5 Spectral Broadening and Relaxation by Coulomb Scattering 448 11.5 Spin Fine Structure of a Few Exciton Spectra: 450 11.6 Conclusions 454
11 Coulomb Effects in the Optical Spectra of Highly Excited Semiconductor Quantum Dots Selvakumar V. Nair 439 11.1 Introduction 439 11.2 Local Density Approximation for Electrons and Holes 440 11.3 Application of the Local Density Approximation 441 11.3.1 Spherical Approximation 442 11.3.2 Cylindrical Quantum Dots 446 11.4 Beyond the Local Density Approximation: 5 Spectral Broadening and Relaxation by Coulomb Scattering 448 11.5 Spin Fine Structure of a Few Exciton Spectra: 450 11.6 Conclusions 454 References 455
11 Coulomb Effects in the Optical Spectra of Highly Excited Semiconductor Quantum Dots Selvakumar V. Nair 439 11.1 Introduction 439 11.2 Local Density Approximation for Electrons and Holes 440 11.3 Application of the Local Density Approximation 441 11.3.1 Spherical Approximation 442 11.3.2 Cylindrical Quantum Dots 446 11.4 Beyond the Local Density Approximation: 5 Spectral Broadening and Relaxation by Coulomb Scattering 448 11.5 Spin Fine Structure of a Few Exciton Spectra: 450 11.6 Conclusions 450 11.6 Conclusions 455 12 Device Applications of Quantum Dots 455
11 Coulomb Effects in the Optical Spectra of Highly Excited Semiconductor Quantum Dots Selvakumar V. Nair 439 11.1 Introduction 439 11.2 Local Density Approximation for Electrons and Holes 440 11.3 Application of the Local Density Approximation 441 11.3.1 Spherical Approximation 442 11.3.2 Cylindrical Quantum Dots 446 11.4 Beyond the Local Density Approximation: 5 Spectral Broadening and Relaxation by Coulomb Scattering 448 11.5 Spin Fine Structure of a Few Exciton Spectra: 450 11.6 Conclusions 455 12 Device Applications of Quantum Dots 457
11 Coulomb Effects in the Optical Spectra of Highly Excited Semiconductor Quantum Dots Selvakumar V. Nair 439 11.1 Introduction 439 11.2 Local Density Approximation for Electrons and Holes 440 11.3 Application of the Local Density Approximation 441 11.3.1 Spherical Approximation 442 11.3.2 Cylindrical Quantum Dots 446 11.4 Beyond the Local Density Approximation: 448 11.5 Spin Fine Structure of a Few Exciton Spectra: 448 11.6 Conclusions 450 11.6 Conclusions 455 12 Device Applications of Quantum Dots 457
11 Coulomb Effects in the Optical Spectra of Highly Excited Semiconductor Quantum Dots Selvakumar V. Nair 439 11.1 Introduction 439 11.2 Local Density Approximation for Electrons and Holes 440 11.3 Application of the Local Density Approximation 441 11.3.1 Spherical Approximation 442 11.3.2 Cylindrical Quantum Dots 446 11.4 Beyond the Local Density Approximation: 5 Spectral Broadening and Relaxation by Coulomb Scattering 448 11.5 Spin Fine Structure of a Few Exciton Spectra: 450 11.6 Conclusions 455 12 Device Applications of Quantum Dots 457 12.1 Improvements of Characteristics in Quantum Dot Devices 457
11 Coulomb Effects in the Optical Spectra of Highly Excited Semiconductor Quantum Dots Selvakumar V. Nair 439 11.1 Introduction 439 11.2 Local Density Approximation for Electrons and Holes 440 11.3 Application of the Local Density Approximation 441 11.3.1 Spherical Approximation 442 11.3.2 Cylindrical Quantum Dots 446 11.4 Beyond the Local Density Approximation: 5 Spectral Broadening and Relaxation by Coulomb Scattering 448 11.5 Spin Fine Structure of a Few Exciton Spectra: 450 11.6 Conclusions 455 12 Device Applications of Quantum Dots 457 12.1 Improvements of Characteristics in Quantum Dot Devices 457 12.1 Thermal Broadening in Bulk 457
11 Coulomb Effects in the Optical Spectra of Highly Excited Semiconductor Quantum Dots Selvakumar V. Nair 439 11.1 Introduction 439 11.2 Local Density Approximation for Electrons and Holes 440 11.3 Application of the Local Density Approximation 440 11.3.1 Spherical Approximation 442 11.3.2 Cylindrical Quantum Dots 446 11.4 Beyond the Local Density Approximation: 446 11.4 Beyond the Local Density Approximation: 446 11.5 Spin Fine Structure of a Few Exciton Spectra: 448 11.6 Conclusions 450 11.6 Conclusions 457 12 Device Applications of Quantum Dots 457 12.1 Improvements of Characteristics in Quantum Dot Devices 457 12.1.1 Thermal Broadening in Bulk and Quantum Well Semiconductors 457

XVI Contents

	12.1.3	Other Characteristic Changes of Quantum Dots
		for Device Applications
	12.1.4	Required Quantum Dots Dimensions
		for Device Applications
	12.1.5	Required Characteristics
		for Quantum Dot Optical Devices
	12.1.6	Advantages of Self-Assembled Quantum Dots 463
12.2	Optica	l Devices with Quantum Dots
	12.2.1	Quantum Dot Lasers
		with Improved Temperature Characteristics
	12.2.2	Lasing Wavelength Control in Quantum Dot Lasers 465
	12.2.3	Reduction of Threshold Current Density
		in Quantum Dot Lasers
	12.2.4	Vertical-Cavity Surface-Emitting Lasers
		with Quantum Dots
	12.2.5	Miscellaneous Improvements in Quantum Dot Lasers 474
	12.2.6	Other Optical Devices
12.3	Future	of Quantum Dot Devices
	12.3.1	Ideal Quantum Dot Structures for Device Applications $\ldots~477$
	12.3.2	Ultimate Device Performances with Quantum Dots 478
References		
Index		