

Contents

1	Introduction	1
1.1	Monolayer Transition Metal Dichalcogenides (TMDs)	2
1.1.1	Electronic Band Structure	2
1.1.2	Optical Selection Rules	5
1.1.3	Excitons	7
1.2	Time-Resolved Spectroscopy	8
1.2.1	Coherent Light-Matter Interactions	9
1.2.2	Quasi-equilibrium Dynamics	9
	References	10
2	Time-Resolved Absorption Spectroscopy	13
2.1	Experimental Setup	14
2.1.1	Overview	14
2.1.2	Laser Amplifier	14
2.1.3	Optical Parametric Amplifier	16
2.1.4	White Light Continuum	17
2.2	Data Analysis	20
2.2.1	Kramers-Kronig Analysis	21
2.2.2	Maxwell's Equations for Monolayer Sample	22
	References	25
3	Intervalley Biexcitons in Monolayer MoS₂	27
3.1	Intervalley Biexcitons	28
3.2	Experimental Methods	29
3.3	Experimental Results and Discussions	30
3.3.1	Intravalley and Intervalley Scattering	31
3.3.2	Signature of Intervalley Biexcitons	32
3.4	Time-Resolved Cooling Process	33
3.5	Conclusions	35
	References	35

4	Valley-Selective Optical Stark Effect in Monolayer WS_2	37
4.1	Optical Stark Effect	38
4.1.1	Semiclassical Description	40
4.1.2	Quantum-Mechanical Description	42
4.2	Experimental Methods	43
4.3	Observation of the Optical Stark Effect	44
4.4	Valley Selectivity	45
4.5	Fluence and Detuning Dependences	46
4.6	Proposal: Valley-Specific Floquet Topological Phase in TMDs	48
4.7	Supplementary	49
4.7.1	Time-Resolved Spectra	49
4.7.2	Polarization-Resolved Spectra	51
4.7.3	Obtaining Energy Shift	51
4.7.4	Comparison from Semiclassical Theory	55
	References	55
5	Intervalley Biexcitonic Optical Stark Effect in Monolayer WS_2	59
5.1	Blue-Detuned Optical Stark Effect	60
5.2	Experimental Methods	61
5.3	Experimental Results and Data Analysis	61
5.4	Intervalley Biexcitonic Optical Stark Effect	64
5.4.1	Four-Level Jaynes-Cummings Model	66
5.5	Perspective: Zeeman-Type Optical Stark Effect	68
5.6	Supplementary	68
5.6.1	Coherent and Incoherent Optical Signals	68
5.6.2	Time-Trace Fitting Decomposition Analysis	70
5.6.3	Possible Effects Under Red-Detuned Pumping	72
5.6.4	Fitting Analysis	73
	References	74
6	Large, Valley-Exclusive Bloch-Siegert Shift in Monolayer WS_2	77
6.1	Bloch-Siegert Shift	77
6.1.1	Semiclassical Description	79
6.1.2	Quantum-Mechanical Description	82
6.2	Experimental Methods	84
6.3	Observation of the Bloch-Siegert Shift	85
6.4	Fluence and Detuning Dependences	87
6.5	Valley-Exclusive Optical Stark Shift and Bloch-Siegert Shift	87
6.6	Conclusions	90
	References	91

7	Lennard-Jones-Like Potential of 2D Excitons in Monolayer WS₂	93
7.1	Many-Body Interactions in 2D TMDs	94
7.2	Experimental Methods	95
7.3	Optical Signature of Many-Body Effects	95
7.3.1	Exciton Redshift-Blueshift Crossover	96
7.3.2	At Low Density: Plasma Contribution	98
7.3.3	At High Density: Exciton Contribution	99
7.4	Lennard-Jones-Like Potential as an Effective Model	99
7.5	Chronological Signature of Interactions in Time-Resolved Spectra	101
7.6	Summary	104
7.7	Supplementary	104
7.7.1	Microscopic Many-Body Computation	104
7.7.2	Exciton-Exciton Annihilation Effect	107
7.7.3	Heat Capacity and Estimated Temperature	108
	References	112
8	XUV-Based Time-Resolved ARPES	115
8.1	Building a High-Resolution XUV Light Source for TR-ARPES	115
8.1.1	Overview	115
8.1.2	XUV Light Source	117
8.1.3	XUV Monochromator	119
8.1.4	XUV Diagnostic Chamber	122
8.2	Measuring TMDs Using 30 eV XUV TR-ARPES	123
8.2.1	WSe ₂ Semiconductors	124
8.2.2	WTe ₂ Semimetal	126
	References	128

Coherent Light-Matter Interactions in Monolayer
Transition-Metal Dichalcogenides

Sie, E.J.

2018, XVII, 129 p. 83 illus., 82 illus. in color., Hardcover

ISBN: 978-3-319-69553-2