

Contents

1	Introduction: Electron and Photon Systems	
H. Nejo and H. Hori	1	
1.1	Size Considerations Concerning Electromagnetic Fields	3
1.2	Comparison of Plasmon and Exciton	4
1.3	Coupling of Light with Plasmons	5
1.4	Photon Emission from STM	5
1.5	Photon Emission from a Molecule Under an STM Tip	9
1.6	Single Polariton Probe	11
1.7	Coulomb Blockade and Connection to a Nonequilibrium Reservoir	13
1.8	Structure of the Book	15
References	15	
2	STM-Induced Photon Emission from Single Molecules	
Z.-C. Dong, T. Ohgi, D. Fujita, H. Nejo, S. Yokoyama, T. Terui, S. Mashiko, T. Okamoto	21	
2.1	Past and Present	23
2.1.1	Semiconductor Surfaces	23
2.1.2	Metal Surfaces	24
2.1.3	Mechanisms	25
2.1.4	Factors Affecting STM-induced Photon Emission and Modes of Measurements	27
2.1.5	The Debate on Mechanisms	28
2.1.6	Molecules on Surfaces	29
2.2	STM-Induced Light Emission from Porphyrin Molecules	32
2.2.1	Experimental Setup	32
2.2.2	Porphyrin Molecules	33
2.2.3	Tunneling Transport Through Molecules and STM Imaging	35
2.2.4	STM-Induced Photon Emission	38
2.2.5	Intensity and Quantum Efficiency	39
2.2.6	Optical Spectra	41
2.3	Conclusion	44
References	45	

VIII Contents

3 Photon Counting Methods in STM and SMS	
A.G. Vitukhnovsky, I.S. Osad'ko	49
3.1 Introduction	49
3.2 Light Emission from STM	50
3.3 Role of Photon Counting in Time-Resolved Spectroscopy	53
3.4 Ensembles and Single-Molecule Spectroscopy Studies	54
3.5 Importance of Ensembles for Applications. Novel Ideas with Nanosize Ensembles.....	55
3.6 Nanoensembles	56
3.6.1 Regular Dendrimers	56
3.6.2 Hyperbranched Polymers.....	57
3.6.3 J-aggregates	58
3.7 Time-Correlated Single Photon Counting (TCSPC) Method	58
3.8 Application of Photon Counting to Kinetic Measurements on Nanoensembles	60
3.8.1 Dendrimer Rotation and Viscosity.....	60
3.8.2 J-aggregate Radiative Lifetime and Structure	63
3.9 Photon Counting in SMS	64
3.10 Intermittency in Single-Molecule Fluorescence	67
3.11 Spectroscopy of Single Molecule with Triplet Level. Photon Bunching	69
3.12 Two-Photon Correlators	72
3.13 Full Two-Photon Correlator for Single Molecule with Triplet Level	75
3.14 Tunneling Dynamics of Polymers Probed by SMS	77
3.15 Relation Between One- and Two-Photon Counting Methods in a More Complex Case	80
3.16 Local Dynamics of Individual Molecules in Polymers	82
3.17 Conclusions	84
3.17.1 Fast Photon Counting vs. CW Measurements	84
3.17.2 Combination of TCSPC and NSOM: New Advantages ...	85
3.17.3 Kinetics of Energy Transfer Between the NSOM Tip and Single Molecules/Ensembles on Surfaces	85
References	86
4 Scanning Tunneling Spectroscopy and Electronic Properties of Single Fullerene Molecules	
J.G. Hou, B. Li	91
4.1 Scanning Tunneling Spectroscopy	92
4.2 Electronic Structures of Fullerene Molecules	94
4.3 Scanning Tunneling Spectroscopy of Single C ₆₀ Molecules Adsorbed on a Si Surface	97
4.3.1 C ₆₀ Molecules on an Si(111)-(7×7) Surface	97
4.3.2 C ₆₀ Molecules on an Si(100)-(2×1) Surface	105

4.4	Electronic Transport Properties of a Single Fullerene Molecule	107
4.4.1	Electrical Resistance of a Single C ₆₀ Molecule Determined by STM	108
4.4.2	Resonant Tunneling of Single C ₆₀ Molecules in a Double Barrier Tunneling Junction (DBTJ)	111
4.4.3	Molecular Device Using a Single C ₆₀ Molecule as the Functional Unit	114
4.5	Conclusion	117
	References	117
5 Interaction Between Nonrelativistic Electrons and Optical Evanescent Waves		
J. Bae, R. Ishikawa, K. Mizuno		121
5.1	Microgap Interaction Circuits	122
5.1.1	Energy and Momentum Conservation	122
5.1.2	Transition Rates of Electrons	124
5.2	Metal Microslit	127
5.2.1	Near-Field Distributions	127
5.2.2	Optimum Slit Width	129
5.2.3	Phase Matching Condition	131
5.2.4	Interaction Space	131
5.3	Modulation with Infrared Evanescent Waves	133
5.3.1	Experimental Setup	133
5.3.2	Electron Energy Spectrum	134
5.3.3	Laser Field Dependence	136
5.3.4	Electron Velocity Dependence	138
5.4	Interaction with Visible Light	139
5.5	Conclusion	141
	References	141
6 The Tunneling Time Problem Revisited		
N. Yamada		143
6.1	What is the Issue?	143
6.2	Well-known Approaches	146
6.2.1	Notation	146
6.2.2	Following the Motion of a Wave Packet	148
6.2.3	Velocity Under the Barrier	149
6.2.4	The Larmor Clock	150
6.2.5	Time-Modulated Barriers	152
6.2.6	Dwell Time and its Decomposition	154
6.2.7	Feynman Paths Approach	157
6.2.8	Systematic Projector Approach	159
6.2.9	Weak Measurement Approach	161
6.2.10	Bohm Trajectory Approach	163

X Contents

6.3	Tunneling Times as Interfering Alternatives:	
	A Feynman Paths Approach	165
6.3.1	Lesson from Two-Slit Experiments	165
6.3.2	Tunneling Time Probability Distribution is not Definable.	167
6.3.3	Relation to Gell-Mann and Hartle's Formulation of Quantum Mechanics	168
6.3.4	$\tau_d = P_t \tau_t + P_r \tau_r$ Is Doubtful	168
6.3.5	Range of Tunneling Times.....	169
6.3.6	An Example: The Monochromatic Case	171
6.4	Conclusion	173
	References	174
	Index	177