

CONTENTS

Preface	v
-------------------	---

Chapter 1. Multiphoton Processes in Frequency-Modulated System

E. Hanamura

1. Introduction	2
2. Frequency Modulation Model	4
2.1 Model of Gaussian-Markoffian Frequency Modulation	6
2.2 Absorption Spectrum	8
3. Optical Nutation	12
3.1 Single-Time Green Function	12
3.2 Density Matrix under Pump Field	15
3.3 Physical Results	22
4. Photon Echo — Effect of Spatial Fluctuation	28
5. Free Induction Decay	39
5.1 Double-Time Electronic Operator	42
5.2 Physical Discussion	48
6. Emission Spectrum under Strong Pumping	55
7. Absorption Spectrum under Strong Pumping	64
8. Raman Scattering and Luminescence in Three-Level System	71
9. Summary	76
References	78

Chapter 2. Photodesorption by Resonant Infrared Laser-Adsorbate Coupling

A Review of the Theoretical Approaches

P. Piercy, Z. W. Gortel and H. J. Kreuzer

1. Introduction	105
2. Basic Photodesorption Model Theory	115
2.1 Introduction	115
2.2 The Hamiltonian of the Gas-Solid-Laser System	119
2.2.1 The static Hamiltonian	120

2.2.2 The electromagnetic field	123
2.2.3 Dynamics of the solid-phonons	131
2.2.4 The complete Hamiltonian	133
2.3 The Master Equation	135
2.4 The Transition Rates in Lowest Order Perturbation Theory	140
2.4.1 Laser-assisted vibrational transitions	142
2.4.2 The phonon interaction	144
2.4.3 Tunneling to the gas phase	149
2.5 Solution of the Master Equation.	151
2.5.1 Direct numerical solution	151
2.5.2 Approximate analytic solution.	154
2.6 Early Results — Theory and Experiment.	166
2.6.1 Theoretical model results	166
2.6.2 Comparison with experimental results	172
3. Resonant Heating	174
3.1 Introduction.	174
3.2 Theory	177
3.2.1 Exact solution	180
3.2.2 Approximate solution	186
3.3 Comparison with Experiments; Discussion	194
4. Lateral Vibrational Energy Transfer	205
4.1 Introduction.	205
4.2 Resonant V-V Transitions.	212
4.3 Macroscopic Energy Flow.	215
4.4 Microscopic V-V Transfer in Photodesorption	219
4.4.1 Harmonic vibrational mode	225
4.4.2 Anharmonic vibrational mode	226
4.5 Discussion	230
5. Coherent Two-Quanta Processes	232
5.1 Introduction.	232
5.2 Formulation of the Transition Rate Calculation	234
5.2.1 Transition rate formula.	235
5.2.2 Description of the reservoirs with Thermo Field Dynamics	238
5.2.3 Perturbation expansion of the transition rates.	243
5.2.4 Bound state resonances in the two-quanta transition rates.	257
5.3 Coherent Two-Phonon Transitions.	260
5.4 Coherent Two-Photon Transitions	271
5.5 Coherent One-Photon plus One-Phonon Transitions	276
5.6 Effect of Coherent Two-Quanta Processes on Desorption Kinetics	279

6. Theory of the Photodesorption Line Shape	286
6.1 Introduction.	286
6.2 Homogeneous Adsorbate	287
6.3 Effect of Vibrational Anharmonicity.	291
6.4 Inhomogeneous Adsorbate	297
6.5 Discussion	302
7. Discussion and Outlook	305
7.1 Selective Photodesorption versus Resonant Heating	305
7.2 Conclusions	312
References	317
Appendix A — Normal Modes of the Radiation Field	324
Appendix B — Morse Potential Matrix Elements	326
Appendix C — V-V Coupling Rate Matrix Elements	328
Appendix D — Diagram Rules and Derivation of (5.38)	330
Appendix E — Components of Equation (5.73)	335