Contents

1.	Mole	cular N	Multiphoton Excitation. By C.D. Cantrell (With 1 Figure)	1
	1.1	Introd	uctory Comments	1
	Refe	rences		7
2.	Sing	le-Infra	ared-Frequency Studies of Multiple-Photon Excitation	
	and	Dissoci	ation of Polyatomic Molecules	
	By J	.L. Lym	an, G.P. Quigley, and O.P. Judd (With 34 Figures)	9
	2.1	0vervi	ew	9
	2.2	Data-R	eduction Techniques	11
		2.2.1	Absorption of Nonuniform Optical Beams by Nonlinear Media	11
		2.2.2	Data-Analysis Techniques for Reaction Studies	16
	2.3	A Quan	titative Comparison of Absorption Data	19
		2.3.1	Interaction Dynamics and Normalization Concepts	20
			a) General Considerations for MPE Processes	20
			b) Molecular Absorption in a Coupled Two-Level Approximation $\cdot\cdot$	22
			c) Generalization to the MPE Processes	24
		2.3.2	MPA in Different Polyatomic Molecules	25
			a) Absorption Characteristics of Polyatomic Molecules	26
			b) Calculation of <f> for SF₆</f>	29
			c) Discussion	30
	2.4	A Revi	ew of MPA Data for SF ₆	32
		2.4.1	Low-Fluence Spectral Absorption	33
		2.4.2	Fluence-Dependent Absorption at 300 K	35
		2.4.3	Fluence-Dependent Absorption at 140 K	40
		2.4.4	Effect of Optical Pulse Duration and Shape in MPA Processes	41
		2.4.5	Discussion and Summary	43
	2.5	A Comp	arison of Multiple-Photon Dissociation Data	44
		2.5.1	Experimental Techniques	45
		2.5.2	Molecular Properties	48
		2.5.3	Comparison of MPD Results	51
		2.5.4	Concluding Remarks	61
	2.6		ional Effects in Multiple-Photon Absorption and	
		Dissoc	iation Processes	61

		2.6.1	Collisional Effects on Multiple-Photon Absorption	• • • • • • • • • • • • • • • • • • • •	63
		2.6.2	The Influence of Collisions on Multiple-Photon		
			Dissociation		68
		2.6.3	Collisional Effects in Laser-Induced Isomerization		
			Reactions and Molecular Elimination		69
		2.6.4	Secondary Processes		70
	2.7	Conclu	sion		7 1
	2.8	Append	lix		72
	Refe	rences			87
3.	Mole	cular-B	leam Studies of Laser-Induced Multiphoton Dissociati	on	
	By A	.S. Suc	tbø, P.A. Schulz, Y.R. Shen, and Y.T. Lee		
			gures)		95
	3.1	0yervi	ew		95
	3.2		· · · · · · · · · · · · · · · · · · ·		97
	3.3	Experi	mental Arrangement		106
	3.4		imental Results		108
	3.5		ssion		113
	3.6	Conclu	uding Remarks		120
	Refe				121
4.	Two	-Freque	ency Technique for Multiple-Photon Dissociation and		
٠.			pe Separation. By R.V. Ambartzumian (With 20 Figur	es)	123
	4.1		round		123
	4.2		Concepts of Two-IR-Frequency Dissociation		125
	4.3		tivity of Dissociation		126
		4.3.1			126
		4.3.2			129
	4.4		tigation of Multiple-Photon Excitation		133
		4.4.1	Evaluation of q and & in OsO4		134
	4.5	Intera	action of the Nonresonant Pulse with Excited Molecul		138
		4.5.1			138
		4.5.2	Effects of Variation of ω_2		141
	4.6		uding Remarks		146
	Refe	erences			146
5.	Exci	tation	Spectrum of SF ₆ Irradiated by an Intense IR Laser	Field	
·			impiev, N.V. Karlov, E.M. Khokhlov, S.M. Nikiforov,		
			orov, B.G. Sartakov, and A.L. Shtarkov (With 7 Figur	res)	149
	5.1		round		149
	5 2	170	xperiment		150

5.3	Experimental Results and Discussion	153
5.4	Conclusion	157
Refe	erences	157
	A CONTRACTOR OF THE CONTRACTOR	
	er-Induced Decomposition of Polyatomic Molecules:	
	omparison of Theory with Experiment	450
	1.F. Goodman, J. Stone, and E. Thiele (With 23 Figures)	
6.1	Brief Historical Review	
6.2	Models of a Complex Polyatomic Molecule	
	6.2.1 Laser Excitation Mechanisms	
	6.2.2 The Heat Bath Feedback Model	
	6.2.3 Some Microscopic Aspects of the Theory	
6.3	Comparisons of Theory with Experiment	
	6.3.1 Laser-Induced Decomposition of CF ₂ HC1	169
	a) Coherent Discrete Level Pumping and Interface with	
	Rate Equation	
	b) Rotational Hole Filling by Collisions	
	c) A Comparison of Theory with Experiment for CF ₂ HCl	
	d) Additional Theoretical Predictions	174
	6.3.2 Laser Intensity Versus Fluence Effects	
	6.3.3 High-Pressure Fallof Reaction Rate	177
	6.3.4 Laser-Induced Decomposition of SF ₆	
	a) Discrete Level Spectrum and Tuning Curve	179
	b) Dissociation Yield and Cross Section	182
	6.3.5 Laser Frequency Effects	183
6.4	Effects Specific to Laser Excitation	185
	6.4.1 Energy Distribution: Laser Versus Thermal	186
	6.4.2 Microstate Formalism	190
	a) Intramolecular Relaxation	190
	b) Relation to RRK and RRKM Theory	191
	c) A Discriminating Reaction Mechanism	193
	d) Multiple Reaction Channels and Product Selectivity	196
6.5	Phase Coherence and the Transition to Incoherent Pumping	198
	6.5.1 The Generalized Master Equation	200
	6.5.2 Fermi Golden Rule Considerations	
	6.5.3 Coherent Quasi-Continuum Pumping - an Open Question	208
6.6	Addendum	209
	References	211

6.

7.	A Method of Laser Isotope Separation Using Adiabatic Inversion					
	By G	.L. Peterson and C.D. Cantrell (With 6 Figures)	215			
	7.1	Background	215			
	7.2	Physical Principles	216			
	7.3	Multilevel Systems Under the Adiabatic Approximation	218			
	7.4	Conditions for Effective Adiabatic Laser Isotope Separation	219			
	Refe	rences	221			
8.	Thre	e-Level Superfluorescence. By F.P. Mattar, P.R. Berman, A.W. Matos,				
	Y. C	laude, C. Goutier, and C.M. Bowden (With 30 Figures)	223			
	8.1	Background	224			
	8.2	Pump Dynamics Effects in Three-Level Superfluorescence	231			
	8.3	Semiclassical Equations of Motion and Computational Method \dots	233			
	8.4	Deterministic Effects of Pump Dynamics in the Nonlinear Regime				
		of Superfluorescence	240			
	8.5	Conclusions Concerning Deterministic Three-Level Superfluorescence \ldots	262			
	8.6	Quantum Initiation: Calculational Results and Delay-Time Statistics \cdot	262			
	8.7	Conclusions Concerning Effects of Resonance Diffraction and				
		Copropagation on Superfluorescence Evolution	266			
	8.8	The Role of Dispersion	268			
	8.9	Conclusions Concerning the Mutual Influence of Dispersion and				
		Diffraction on the Superfluorescence Buildup	276			
	8.10	Final Conclusion	276			
	Refe	rences	277			
	Addi	tional References	28			
C 1-		and an	200			