Contents

1.	intr	oduction	1
2.	Mol 2.1	ecular Energy States	5
		and the Hamiltonian Operator	5
	2.2	Molecular Electronic States	6
	2.3	Molecular Vibrational Levels	10
	2.4	Molecular Rotational Levels	14
	2.5	Molecular Vibration-Rotational Levels	18
	2.6	Coupling of Molecular Rotation and Electronic Motion	19
	2.7	Perturbations	22
	2.8	Quadrupole Hyperfine Structure of Molecules	27
	2.9	Magnetic Dipole Hyperfine Structure in Molecules	29
	2.10	Isotopic Energy-Level Shifts	31
	2.11	Molecular Rydberg States	33
3.	Lin	ear Molecular Spectroscopy	42
	3.1	Infrared Pure-Rotational Spectra	42
	3.2	Infrared Vibrational Spectra	43
		Infrared Vibration-Rotational Spectra	44
		Vibrational Band Systems of Diatomic Molecules	48
	3.5	Rotational Spectra of Electronic Bands	
	200	of Diatomic Molecules	50
	3.6	Electric Quadrupole and Magnetic Dipole Hyperfine	
		Spectra of Molecules	51
	3.7	The Goals for Experimental Studies of Molecular	
		Spectroscopy	52
	3.8	Advances of Molecular Spectroscopy	
		Through Linear Interaction of Molecules with Lasers	53
2		and Oles and the control of the cont	
4.		ctral Characteristics of Molecular Two-Photon	<i>-</i>
	4.1	nsitions	02
	4.1		
	4.2	Two-Photon Transitions	0.3
	4.2	Excitation Probability of a Two-Photon Transition	64
	4.3	with One Near-Resonant Enhancing Level	oc
	4.3	Coarse Structure of Near-Resonantly Enhanced	7
		Molecular Two-Photon Absorption Spectra	14

	4.4	Fine Structure of Near-Resonantly Enhanced	
		Molecular Two-Photon Transitions	83
	4.5	Line Shapes and Higher-Order Corrections	
		for Near-Resonant Two-Photon Transitions	
		in Three-Level Systems	89
	4.6	Molecular Two-Photon Transitions Enhanced	
		by Mixing Levels	93
	4.7	Semiclassical Theory for a Two-Photon Transition	
			101
	4.8	Coherent Effects on the Line Shape of a Near-Resonant	-0-
			104
5	Mo	lecular Nonlinear Uncoupling Spectra	
J.			108
	5.1		
	3.1		109
			111
			117
	<i>5</i> 3		121
	5.2		123
	5.3		129
	5.4		137
	5.5	Applications of Nonlinear High-Resolution Laser	
			141
		5.5.1 Accurate Measurements of the Properties of Perturbed	
			141
			147
			152
			157
		5.5.5 General Features of the Applications of Doppler-Free	
		Spectroscopy to the Study of Molecular Spectra	
		and Molecular Structure	159
6.	Mo	lecular Nonlinear Coupling Spectral Effects	161
	6.1		161
	6.2		162
	6.3	The state of the s	165
	6.4		103
	0.1		169
	6.5	Stimulated and Coherent Radiation by Hybrid Excitation	10)
	0.5		174
			174
		6.5.2 Unequal-Frequency Two-Step Hybrid	1/4
			176
			177
		6.5.4 Four-Wave Mixing Following Two-Step Hybrid	1//
			101
			181 184
		0.5.5 Profession Comsional Energy-Transfer Processes .	104

Based on Molecular Electronic Transitions 6.7 Lasers Based on Molecular Photodissociation 6.8 Optically Pumped Far-Infrared Lasers Based on Pure Rotational Molecular Transitions 6.9 Optically Pumped Mid-Infrared Laser Based on Molecular Vibration-Rotational Transitions 6.10 Applications of Coherent Transient Spectroscopy in the Measurement of Molecular Parameters 6.10.1 Determination of Dipole Moments for Molecular Transitions 6.10.2 Lifetime of the Upper Level of a Molecular Transition 6.10.3 Determination of Transition Relaxation 6.10.4 The Fine or Hyperfine Splitting of Molecular-Spectral Lines 6.10.5 Study of Weak Intramolecular Coupling 6.10.6 Measurement of Ultrafast Inter- or Intra-Molecular Processes by Novel Transient Techniques 7. Simplification and Identification of Molecular Spectra 7.1 Laser-Induced Fluorescence 7.1.1 Formation and Classification of Resonant Fluorescence 7.1.2 Measurements and Applications 7.2 Population Labelling 7.3 Polarization Labelling 7.4 Two-Step Polarization Labelling	188 197 201 204 206 207 207 208 208 209 210 212 212 217
6.8 Optically Pumped Far-Infrared Lasers Based on Pure Rotational Molecular Transitions 6.9 Optically Pumped Mid-Infrared Laser Based on Molecular Vibration-Rotational Transitions 6.10 Applications of Coherent Transient Spectroscopy in the Measurement of Molecular Parameters 6.10.1 Determination of Dipole Moments for Molecular Transitions 6.10.2 Lifetime of the Upper Level of a Molecular Transition 6.10.3 Determination of Transition Relaxation 6.10.4 The Fine or Hyperfine Splitting of Molecular-Spectral Lines 6.10.5 Study of Weak Intramolecular Coupling 6.10.6 Measurement of Ultrafast Inter- or Intra-Molecular Processes by Novel Transient Techniques 7.1 Laser-Induced Fluorescence 7.1.1 Formation and Classification of Resonant Fluorescence 7.1.2 Measurements and Applications 7.3 Population Labelling 7.4 Two-Step Polarization Labelling	201 204 206 207 207 208 208 209 210 212 212
Based on Pure Rotational Molecular Transitions 6.9 Optically Pumped Mid-Infrared Laser Based on Molecular Vibration-Rotational Transitions 6.10 Applications of Coherent Transient Spectroscopy in the Measurement of Molecular Parameters 6.10.1 Determination of Dipole Moments for Molecular Transitions 6.10.2 Lifetime of the Upper Level of a Molecular Transition 6.10.3 Determination of Transition Relaxation 6.10.4 The Fine or Hyperfine Splitting of Molecular-Spectral Lines 6.10.5 Study of Weak Intramolecular Coupling 6.10.6 Measurement of Ultrafast Inter- or Intra-Molecular Processes by Novel Transient Techniques 7. Simplification and Identification of Molecular Spectra 7.1 Laser-Induced Fluorescence 7.1.1 Formation and Classification of Resonant Fluorescence 7.1.2 Measurements and Applications 7.3 Population Labelling 7.4 Two-Step Polarization Labelling	204 206 207 207 208 208 209 210 212 212
6.9 Optically Pumped Mid-Infrared Laser Based on Molecular Vibration-Rotational Transitions 6.10 Applications of Coherent Transient Spectroscopy in the Measurement of Molecular Parameters 6.10.1 Determination of Dipole Moments for Molecular Transitions 6.10.2 Lifetime of the Upper Level of a Molecular Transition 6.10.3 Determination of Transition Relaxation 6.10.4 The Fine or Hyperfine Splitting of Molecular-Spectral Lines 6.10.5 Study of Weak Intramolecular Coupling 6.10.6 Measurement of Ultrafast Inter- or Intra-Molecular Processes by Novel Transient Techniques 7.1 Laser-Induced Fluorescence 7.1.1 Formation and Classification of Resonant Fluorescence 7.1.2 Measurements and Applications 7.3 Population Labelling 7.4 Two-Step Polarization Labelling	204 206 207 207 208 208 209 210 212 212
Based on Molecular Vibration-Rotational Transitions 6.10 Applications of Coherent Transient Spectroscopy in the Measurement of Molecular Parameters 6.10.1 Determination of Dipole Moments for Molecular Transitions 6.10.2 Lifetime of the Upper Level of a Molecular Transition 6.10.3 Determination of Transition Relaxation 6.10.4 The Fine or Hyperfine Splitting of Molecular-Spectral Lines 6.10.5 Study of Weak Intramolecular Coupling 6.10.6 Measurement of Ultrafast Inter- or Intra-Molecular Processes by Novel Transient Techniques 7. Simplification and Identification of Molecular Spectra 7.1 Laser-Induced Fluorescence 7.1.1 Formation and Classification of Resonant Fluorescence 7.1.2 Measurements and Applications 7.3 Population Labelling 7.4 Two-Step Polarization Labelling	206 207 207 208 208 209 210 212 212
 6.10 Applications of Coherent Transient Spectroscopy in the Measurement of Molecular Parameters 6.10.1 Determination of Dipole Moments for Molecular Transitions 6.10.2 Lifetime of the Upper Level of a Molecular Transition 6.10.3 Determination of Transition Relaxation 6.10.4 The Fine or Hyperfine Splitting of Molecular-Spectral Lines 6.10.5 Study of Weak Intramolecular Coupling 6.10.6 Measurement of Ultrafast Inter- or Intra-Molecular Processes by Novel Transient Techniques 7. Simplification and Identification of Molecular Spectra 7.1 Laser-Induced Fluorescence 7.1.1 Formation and Classification of Resonant Fluorescence 7.1.2 Measurements and Applications 7.3 Population Labelling 7.4 Two-Step Polarization Labelling 	206 207 207 208 208 209 210 212 212
in the Measurement of Molecular Parameters 6.10.1 Determination of Dipole Moments for Molecular Transitions 6.10.2 Lifetime of the Upper Level of a Molecular Transition 6.10.3 Determination of Transition Relaxation 6.10.4 The Fine or Hyperfine Splitting of Molecular-Spectral Lines 6.10.5 Study of Weak Intramolecular Coupling 6.10.6 Measurement of Ultrafast Inter- or Intra-Molecular Processes by Novel Transient Techniques 7.1 Laser-Induced Fluorescence 7.1.1 Formation and Classification of Resonant Fluorescence 7.1.2 Measurements and Applications 7.3 Population Labelling 7.4 Two-Step Polarization Labelling	206 207 207 208 208 209 210 212 212
6.10.1 Determination of Dipole Moments for Molecular Transitions 6.10.2 Lifetime of the Upper Level of a Molecular Transition 6.10.3 Determination of Transition Relaxation 6.10.4 The Fine or Hyperfine Splitting of Molecular-Spectral Lines 6.10.5 Study of Weak Intramolecular Coupling 6.10.6 Measurement of Ultrafast Inter- or Intra-Molecular Processes by Novel Transient Techniques 7.1 Laser-Induced Fluorescence 7.1.1 Formation and Classification of Resonant Fluorescence 7.1.2 Measurements and Applications 7.2 Population Labelling 7.3 Polarization Labelling 7.4 Two-Step Polarization Labelling	206 207 207 208 208 209 210 212 212
for Molecular Transitions 6.10.2 Lifetime of the Upper Level of a Molecular Transition 6.10.3 Determination of Transition Relaxation 6.10.4 The Fine or Hyperfine Splitting of Molecular-Spectral Lines 6.10.5 Study of Weak Intramolecular Coupling 6.10.6 Measurement of Ultrafast Inter- or Intra-Molecular Processes by Novel Transient Techniques 7.1 Laser-Induced Fluorescence 7.1.1 Formation and Classification of Resonant Fluorescence 7.1.2 Measurements and Applications 7.2 Population Labelling 7.3 Polarization Labelling 7.4 Two-Step Polarization Labelling	207 207 208 208 209 210 212 212
6.10.2 Lifetime of the Upper Level of a Molecular Transition 6.10.3 Determination of Transition Relaxation 6.10.4 The Fine or Hyperfine Splitting of Molecular-Spectral Lines 6.10.5 Study of Weak Intramolecular Coupling 6.10.6 Measurement of Ultrafast Inter- or Intra-Molecular Processes by Novel Transient Techniques 7.1 Laser-Induced Fluorescence 7.1.1 Formation and Classification of Resonant Fluorescence 7.1.2 Measurements and Applications 7.2 Population Labelling 7.3 Polarization Labelling 7.4 Two-Step Polarization Labelling	207 207 208 208 209 210 212 212
6.10.3 Determination of Transition Relaxation 6.10.4 The Fine or Hyperfine Splitting of Molecular-Spectral Lines 6.10.5 Study of Weak Intramolecular Coupling 6.10.6 Measurement of Ultrafast Inter- or Intra-Molecular Processes by Novel Transient Techniques 7. Simplification and Identification of Molecular Spectra 7.1 Laser-Induced Fluorescence 7.1.1 Formation and Classification of Resonant Fluorescence 7.1.2 Measurements and Applications 7.2 Population Labelling 7.3 Polarization Labelling 7.4 Two-Step Polarization Labelling	207 208 208 209 210 212 212
6.10.4 The Fine or Hyperfine Splitting of Molecular-Spectral Lines 6.10.5 Study of Weak Intramolecular Coupling 6.10.6 Measurement of Ultrafast Inter- or Intra-Molecular Processes by Novel Transient Techniques 7. Simplification and Identification of Molecular Spectra 7.1 Laser-Induced Fluorescence 7.1.1 Formation and Classification of Resonant Fluorescence 7.1.2 Measurements and Applications 7.2 Population Labelling 7.3 Polarization Labelling 7.4 Two-Step Polarization Labelling	208 208 209 210 212 212
of Molecular-Spectral Lines 6.10.5 Study of Weak Intramolecular Coupling 6.10.6 Measurement of Ultrafast Inter- or Intra-Molecular Processes by Novel Transient Techniques 7. Simplification and Identification of Molecular Spectra 7.1 Laser-Induced Fluorescence 7.1.1 Formation and Classification of Resonant Fluorescence 7.1.2 Measurements and Applications 7.2 Population Labelling 7.3 Polarization Labelling 7.4 Two-Step Polarization Labelling	208 209 210 212 212
6.10.5 Study of Weak Intramolecular Coupling 6.10.6 Measurement of Ultrafast Inter- or Intra-Molecular Processes by Novel Transient Techniques 7. Simplification and Identification of Molecular Spectra 7.1 Laser-Induced Fluorescence 7.1.1 Formation and Classification of Resonant Fluorescence 7.1.2 Measurements and Applications 7.2 Population Labelling 7.3 Polarization Labelling 7.4 Two-Step Polarization Labelling	208 209 210 212 212
6.10.6 Measurement of Ultrafast Inter- or Intra-Molecular Processes by Novel Transient Techniques 7. Simplification and Identification of Molecular Spectra 7.1 Laser-Induced Fluorescence 7.1.1 Formation and Classification of Resonant Fluorescence 7.1.2 Measurements and Applications 7.2 Population Labelling 7.3 Polarization Labelling 7.4 Two-Step Polarization Labelling	209 210 212 212
Processes by Novel Transient Techniques 7. Simplification and Identification of Molecular Spectra 7.1 Laser-Induced Fluorescence 7.1.1 Formation and Classification of Resonant Fluorescence 7.1.2 Measurements and Applications 7.2 Population Labelling 7.3 Polarization Labelling 7.4 Two-Step Polarization Labelling	210 212 212
7. Simplification and Identification of Molecular Spectra 7.1 Laser-Induced Fluorescence 7.1.1 Formation and Classification of Resonant Fluorescence 7.1.2 Measurements and Applications 7.2 Population Labelling 7.3 Polarization Labelling 7.4 Two-Step Polarization Labelling	210 212 212
 7.1 Laser-Induced Fluorescence 7.1.1 Formation and Classification of Resonant Fluorescence 7.1.2 Measurements and Applications 7.2 Population Labelling 7.3 Polarization Labelling 7.4 Two-Step Polarization Labelling 	212 212
 7.1 Laser-Induced Fluorescence 7.1.1 Formation and Classification of Resonant Fluorescence 7.1.2 Measurements and Applications 7.2 Population Labelling 7.3 Polarization Labelling 7.4 Two-Step Polarization Labelling 	212
 7.1.1 Formation and Classification of Resonant Fluorescence 7.1.2 Measurements and Applications 7.2 Population Labelling 7.3 Polarization Labelling 7.4 Two-Step Polarization Labelling 	
7.2 Population Labelling	217
7.2 Population Labelling	41/
7.3 Polarization Labelling	218
7.4 Two-Step Polarization Labelling	221
	232
7.5 Modulated Polarization Two-Photon Spectroscopy	236
7.6 Molecular Energy Level Information	
Provided by Selective Simplified Molecular Spectra	241
7.7 Comprehensive Identification of Equal-Frequency	
Molecular Two-Photon Transitions	245
7.7.1 Approximate Numerical Calculations for Predicting	
Observable Molecular Two-Photon Absorption	
Frequencies	245
7.7.2 Experimental Methods	247
7.7.3 Examination of the Population Paths of a Fine Level	
for a Two-Photon Transition	252
References	
	255