

# CONTENTS

## I. LASER RAMAN SPECTROSCOPY OF CRYSTALS

THE CURRENT STATE AND ISSUE OF RAMAN POLARITON LIGHT SCATTERING SPECTROSCOPY .....	1
A.N. Penin, Yu.N. Polivanov	
Bibliography .....	10
THE INTENSITY OF RAMAN POLARITON LIGHT SCATTERING AND THE NONLINEAR SUSCEPTIBILITY DISPERSION IN CERTAIN ANISOTROPIC POLYATOMIC CRYSTALS .....	15
O.A. Doil'nitsyna, Yu.N. Polivanov	
1. Experimental technique .....	16
2. Lithium iodate .....	17
3. Potassium niobate .....	24
4. Potassium bromate .....	29
Bibliography .....	33
MANIFESTATION OF ANHARMONISM EFFECTS IN POLARITON AND OPTICAL PHONON RAMAN SCATTERING SPECTRA .....	37
Yu.N. Polivanov	
1. Select theoretical data .....	39
2. Experimental results .....	44
2.1. Lithium iodate .....	44
2.2. Ammonium chloride .....	47
2.3. Lithium formiate .....	47
2.4. Iodic acid .....	51
Bibliography .....	58
THE SPECTRA OF RAMAN LIGHT SCATTERING BY SOFT-MODE POLARITONS .....	63
Sh. Atabaev, S.N. Poluektov	
Introduction .....	63
1. The spectral distribution of Raman light scattering inten- sity by soft-modes .....	64
2. Relations describing the intensity distribution of Raman light scattering by polaritons associated with damped phonons	65
3. The $k$ - and $\omega$ -spectra of Raman light scattering by polaritons	

associated with resonant soft-modes .....	66
4. The spectra of Raman light scattering by polaritons associated with relaxational soft-modes .....	72
5. Experimental investigation of the spectra of Raman light scattering by polaritons associated with the soft-mode of a lithium tantalate crystal .....	74
Bibliography .....	76

RESONANCE HYPER-RAMAN LIGHT SCATTERING IN CdS CRYSTAL .. 79

R.Sh. Sayakhov

Introduction .....	79
1. Experimental technique for recording hyper-Raman .....	80
2. Measurement of hyper-Raman scattering cross-sections .....	81
3. The influence of two-photon absorption on the hyper-Raman scattering process .....	83
4. Resonance hyper-Raman scattering .....	85
Bibliography .....	87

**II. COHERENT ANTI-STOKES RAMAN LIGHT  
SCATTERING IN MOLECULAR GASES**

High-Resolution CARS- [Coherent anti-Stokes Raman Scattering] Spectroscopy of Molecular Gases .....	91
--	----

S.Yu. Volkov, D.N. Kozlov, A.M. Prokhorov  
V.V. Smirnov, V.I. Fabelinskiy

Chapter I - CARS as a Spectroscopy Technique For Application to Gases .....	91
Chapter II - Experimental Equipment For CARS-Spectroscopy of Gases .....	99
1. The design principles for the CARS-spectrometer scheme .....	99
2. Pulsed CARS-spectrometer .....	100
2.1. Biharmonic laser pump system .....	100
2.2. Optical scheme .....	101
2.3. Data recording and display system .....	101
3. High-resolution IR-CARS-spectrometer .....	102
3.1. The principle of operation of the IR-CARS-spectrometer	103
3.2. The laser pump system .....	103
3.3. The frequency measurement and spectral monitoring system .....	105
3.4. A parametric difference frequency oscillator .....	105
3.5. The data control, collection and processing system ...	105
4. A precision laser analyzer-frequency meter .....	106

5.	A narrow band tunable infrared oscillator .....	110
5.1.	Difference frequency generation as a method of obtaining tunable infrared emission .....	110
5.2.	Difference frequency generation in an $\text{LiIO}_3$ crystal ..	110
6.	A pulsed wideband tunable amplifier based on organic dyes ..	112
Chapter III - High-Resolution CARS- and IR-Spectroscopy of Spherical Top Molecules $\text{CD}_4$ , $\text{CH}_4$ , $\text{SiH}_4$ , $\text{GeH}_4$ , $\text{SF}_6$ .....		115
1.	The effective vibrational-rotational Hamiltonian and structure of the rotational levels in the $\nu_1(a_1)$ and $\nu_3(f_2)$ vibrational states of tetrahedral molecules .....	116
2.	The rotational structure of the CARS-spectra of the $Q_{01}$ -branch and the structure of the $\nu_1$ vibrational state in the $\text{CD}_4$ and $\text{CH}_4$ molecules .....	123
3.	The rotational structure of the CARS-spectra of the $Q_{01}$ -branch and the IR-spectra of the $Q_{03}$ -branch and the structure of the $\nu_1$ and $\nu_3$ vibrational states in the $^{28}\text{SiH}_4$ and $^{74}\text{GeH}_4$ molecules .....	124
4.	The rotational structure of the CARS-spectra of the $Q_{01}$ -branch of the $\nu_1$ vibration of the $^{32}\text{SF}_6$ and $^{34}\text{SF}_6$ octahedral molecules .....	130
Chapter IV - The High-Resolution CARS-spectroscopy of the Line Contour .....		132
1.	The features of the behavior of the Q-branch spectral contour of linear and spherical molecules in gases with variable densities and the relation to the relaxational mechanisms .....	133
1.1.	Introductory commentary .....	133
1.2.	The contour of a separate rotational line in the resolved CARS-spectra of the Q-branch .....	134
1.3.	The behavior of the Q-branch contour with overlapping rotational components in the high density range .....	136
2.	Broadening of separate lines in the resolved spectra of the Q-branch of $D_2$ molecules .....	139
3.	An investigation of the contour of the Q-branch of the spectra for the $\text{N}_2$ and $\text{C}_2\text{H}_2$ linear molecules .....	141
4.	An investigation of the Q-branch contour of the $\nu_1$ vibration in the $\text{CH}_4$ and $\text{SiH}_4$ spherical top spectra .....	145
Bibliography .....		150
LOCAL NONINVASIVE DIAGNOSTICS OF GASEOUS MEDIA .....		165
S.I. Valyanskiy, K.A. Vereshchagin, A.Yu. Volkov		
A.A. Ilyukhin, P.P. Pashinin, V.V. Smirnov		
V.I. Fabelinskiy		

Chapter I - CARS-Investigation of the Population Distribution Functions and Kinetics of the Vibrational States of Nitrogen Molecules .....	165
1. Temperature measurements and the spectroscopy of discharge- excited vibrational-rotational states in nitrogen .....	166
2. An investigation of the kinetics of the vibrational distri- bution function of pulsed discharge-excited nitrogen .....	168
2.1. Experimental set-up .....	169
2.2. Determining the rotational (and translational) gas temperature in pulsed discharge .....	169
2.3. Experimental investigations of the vibrational kinetics of nitrogen .....	170
2.4. Numerical modeling of the $\nu$ - $\nu$ -process .....	171
3. Measuring the $\nu$ - $\nu$ -exchange rate constant in nitrogen under biharmonic excitation .....	175
3.1. Experimental technique .....	175
3.2. Numerical modeling of the $\nu$ - $\nu$ -exchange and diffusion of vibrationally-excited molecules .....	178
Chapter II - The Application of the CARS Method For Practical Local Noninvasive Measurement of Gas Media .....	181
1. The diagnostic capabilities of the CARS method .....	181
2. An analysis of the CARS-spectra of the Q-branch of nitrogen for temperature and density diagnostics .....	182
3. Measurement of density and temperature distributions in a supersonic nitrogen flow .....	183
4. Measuring gas temperature in a flow by the intensity distri- bution of the lines in the O- and S-branches of CARS-spectra	185
5. Temperature measurements in an air flow heated by plasm- tron electric arc discharge .....	186
Bibliography .....	187
THE CARS-SPECTROSCOPY OF SF <sub>6</sub> MOLECULES EXCITED IN A POWERFUL IR-LASER FIELD .....	191
S.S. Alimpiev, A.A. Mokhnatyuk, S.M. Nikiforov, P.P. Pashinin B.G. Sartakov, V.V. Smirnov, V.I. Fabelinskiy	
Introduction .....	191
1. Experimental set-up and measurement technique .....	192
2. The initial distribution function of excited molecules .....	195
3. Investigation resonance $\nu$ - $\nu$ -exchange and interisotope vibrational exchange of excited SF <sub>6</sub> molecules .....	198

4. Vibrational energy exchange with molecules excited to the quasicontinuum range .....	203
5. Determining the energy boundary of loss of modal selectivity of SF <sub>6</sub> molecular excitation .....	204
Bibliography .....	208
SUBJECT INDEX.....	209