Saturation of infrared absorption in sulfur hexafluoride near 10.5 μ m at the P12, P14, P16, P18 and P20 of emission CO₂ laser lines

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This paper presents the results of the investigations of absorption in SF_6 for several emission lines of CO_2 laser. The results obtained are compared with those reported from other laboratories. Making use of four-level model of absorbers the characteristic absorption parameters of SF_6 have been calculated.

1. Introduction

Sulfur hexafluoride has found many applications in investigations of such optical phenomena as: self-induced transparency [1], photon echo [2], infrared double resonance [3], fluorescence [4], nutation effect [5], passive mode locking [6], transmission of infrared pulses [7], third harmonic generation at 10.6 μ m [8], passive Q-switching [9], saturation absorption [10]. The last two optical phenomena are used to produce short light pulses [11], in saturation laser spectroscopy [12], and frequency stabilization of CO_2/SF_6 lasers [13]. In these experiments the knowledge of the transmission properties of the absorbing medium is necessary. These properties were investigated for ν_3 band of SF_6 which well coincides with ν_6 emission band of CO_2 laser [14–16].

In this paper results of investigation of absorption in SF_6 for several emission lines of a cw CO₂ laser are given.

2. Energy levels system of SF.

The SF₆ molecules have the fundamental vibrations of the following symmetries: A_{1u}-absorption band ($v_1 - 12.90 \mu$ m), E_g-absorption band ($v_2 - 15.52 \mu$ m), F_{1u}-absorption bands ($v_3 - 10.55 \mu$ m or $v_4 - 16.26 \mu$ m), F_{2g}-absorption band ($v_5 - 19.08 \mu$ m), F_{2u}-absorption band ($v_6 - 27.54 \mu$ m) [17, 18]. These symmetries and their components give a complex infrared spectrum which had been observed as being continuous [19] until laser spectroscopy was applied [20].

The strongest absorbing transitions occur between v_0 and v_3 , and v_6 and $v_3 + v_6$ bands as result of thermal population [21]. According to McDowell et al. [22] 30.1% of SF₆ molecules are in the vibrational ground state, 17.2% are excited to v_6 band, 7.4% to v_5 band and 6.5% to 2 v_6 band at 300 K. The remaining 38.8% of molecules are excited to other vibrational-rotational states. In addition there are fast thermal V-V transitions from v_3 to v_6 with an order of magnitude of 10^{-6} s [2]. These transitions are very effective since v_3 energy level lies near J47 rotational level of v_6 band. The J47 rotational level positioned in the maximum of energy distribution function of rotational levels of v_3 band [16]. Hence, four levels v_0 , v_3 , v_6 and $v_3 + v_6$ are the most important in absorption processes (see Fig. 1).



Fig. 1. Four-level of SF_6 as a saturable absorber

3. Four-level system

Absorbing processes of electromagnetic radiation in such absorber as SF_6 is well described by four-level model of saturable absorbers presented by HERCHER [23]. The intensity I of radiation as a function of depth x in optically thick absorbing medium is given by

$$\frac{dI}{dx} = -I\varkappa \tag{1}$$

where \varkappa – absorption coefficient.

Assuming fast transmitions 3-2 and 4-2 (Fig. 1), what is true in the case of SF₆, the steady-state solution of rate equations for the population of the energy levels in four-level model shows that the coefficient \varkappa depends on the intensity *I* of radiation. Therefore, according to Hercher, we may write

$$\frac{dI}{dx} = -I\left[\frac{\alpha_0 + \beta_0(I/I_s)}{1 + I/I_s}\right]$$
(2)

where: a_0 and β_0 — small-signal absorption coefficient for the transitions 1-3 and 2-4, respectively, I_s — saturation intensity. The absorber transition $T = I/I_0$ for boundary condition $I = I_0$ at x = 0 may be found from Eq. (2)

$$\ln T - \ln T_0 = (\gamma - 1) \ln \left[\frac{\gamma + I_0 / I_s}{\gamma + T (I_0 / I_s)} \right]$$
(3)

where

$$\gamma = \alpha_0 / \beta_0 \tag{4}$$

is the measure of hot transitions 2-4 participation in the absorption process,

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and

$$T_{0} = \exp[-(a_{0} + \beta_{0})x]$$
(5)

is small-signal transmission $(I \leq I_s)$.

The parameters $a_0 + \beta_0$, γ and I_s describe the response of saturable absorber excited by external radiation. The parameter $a_0 + \beta_0$ may be calculated from the expression (5), if T_0 is measured. The first-order approximation of series expansion of (5) in powers of γI_s (for $I \ge I_s$) gives

$$T \approx T_0^{1/\gamma} = T_{\max}.$$
 (6)

The parameter γ may be calculated from the expression (6), if T_{\max} is known from the experiment. For a low intensity ($I \leq I_s$) a series expansion of (5) in powers of I_0/I_s gives this time

$$\ln T - \ln T_0 = \frac{(\gamma - 1)(1 - T)I_0}{\gamma I_s}.$$
 (7)

If transmission T at different input radiation intensity I_0 is known from the measurements, the saturation intensity I_s may be easily calculated from (7).

4. Experiment

The home-made low power cw CO_2 laser (output power $P_{max} \approx 2$ W) was used in the experiment [24]. The selection of emission lines was realized by means of reflecting diffraction grating (153 lines per mm). The current stabilizer was connected with a discharge laser tube from the cathode sides in order to control discharge current (Fig. 2). The piezoceramic driver and dc amplifier give the



Fig. 2. Experimental set-up

possibility of fixing the laser frequency at the centre of emission line. The absorption cell, 30 cm long and 2.5 cm in diameter, filled with SF₆ at a pressure of a 100 m Torr was ended with NaCl Brewster-angle windows. The ν_3 band of SF₆ was investigated with CO₂ laser until proper lines were selected (Fig. 3). The results of experiment were similar to those in [25]. Five emission lines, i.e., P12, P14, P16, P18 and P20 coinciding with ν_3 band centre of SF₆, were selected for the measurements of the parameters I_s , $a_0 + \beta_0$ and γ . The measurements of transmission characteristics $T = f(I_0)$ were realized with two noncooled CdHgTe detectors [26] which were calibrated by means of a bolometer. From the characteristics $T = f(I_0)$ it follows that the absorber investigated does not reach the 100 % transmission value (Fig. 4), as it is in four-level saturable absorber model [23]. In that case the expressions (5)-(7) were used to calculate the absorption parameters $a_0 + \beta_0$, γ and I_s , respectively.

The results obtained and those given by other authors are shown in the Table.



Fig. 3. Transmittance of CO₂ laser radiation for the emission lines of P branch of 10.4 μ m band passing through the absorption cell at different pressures of SF₆



Fig. 4. Transmittance of CO_2 laser radiation vs. intensity of input radiation I_0 for different emission lines

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Emission line of CO ₂ laser	Present work	References	
		[15]	[16]
	$a_0 + \beta_0$		
	$m^{-1}Pa^{-1}(cm^{-1}Torr^{-1})$		
P12	0.09 (0.12)	_	_
P14	0.17 (0.23	-	_
P16	0.51 (0.68)	0.98 (1.30)	<u> </u>
P18	0.30 (0.40)	0.34 (0.46)	_
P20	0.19 (0.25)	0.25 (0.34)	0.34 (0.46)
	$\gamma = a_0/\beta_0$		
P12	5.3	_	
P14	3.7		<u> </u>
P16	4.7	-	- i -
P18	4.6	-	_
P20	3.4	-	3.6
	I_s Wm ⁻² Pa ⁻¹ (Wcm ⁻² Torr ⁻¹)		
P12	255 (3.4)	-	_
P14	300 (4.0)		_
P16	285 (3.8)		_
P18	428 (5.7)		
P20	488 (6.5)	526 (7.0)	451 (6.0)

The obtained values of the absorption parameters $a_0 + \beta_0$, γ and I_s

5. Discussion of results

The intensity of laser beam radiation has been estimated by measuring the laser power (by means of a bolometer) and an output laser beam diameter. The laser beam diameter has been estimated assuming the transverse distribution of laser beam intensity as the Gaussian. That assumption is valid only at single-transverse-mode TEM_{00} laser operation. The upper range transverse modes have been suppressed by a diaphragm D placed inside the laser cavity (Fig. 2). The errors of measurement of the laser beam diameter and the lack of control of a laser beam mode spectrum [27] in the course of experiment could yield different results than those obtained by other authors.

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Насыщение инфракрасного поглощения в гексафториде серы вблизи 1.5 мкм на эмиссионных линия Р12, Р14, Р16, Р18 и Р2О СО₂ лазерах

Представлены результаты исследований поглощения в SF₆ для нескольких эмиссионных линий CO₂ лазера. Полученные результаты сравниваются с результатами, полученными другими лабораториями. Вычислены характеристические параметры поглощения SF₆ при помощи четырехуровневой модели поглотителя.