Optica Applicata, Vol. XV, No. 1, 1985

Nonlinear laser spectroscopy applied to SF_6

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The investigations on Lamb dip effect in sulfur hexafluoride as a resonance-absorbing medium using a low power cw CO_2 laser are presented in the paper. The locations of SF_6 absorption peaks are given in relation to the centres of the respective CO_2 emission lines, such as P12, P14, P16, P18 and P20 of 10.6 μm.

1. Introduction

Nonlinear laser spectroscopy enables the effective elimination of the Doppler broadening of spectral lines, i.e., the penetration within the Doppler profile, and permits spectroscopic measurements of unusual precision [1–3]. In experiment with nonlinear laser spectroscopy, where the investigated absorber is placed in a cell outside the laser resonator, the absorber molecules are excited simultaneously with two laser waves travelling oppositely, i.e., with a saturate wave and a probe wave. If the laser frequency Ω is detuned from the centre ω_0 of the absorption line, then the two oppositely directed waves, the saturate and the probe ones, will interact with two distinct groups of absorbing spectral profile, i.e., with molecules, the Doppler shift for which is equal to $\omega_D = \Omega(1 \pm v/c)$, where v – velocity of absorbing molecules, c – velocity of light. In this case, the probe wave is attenuated by another group of absorbing molecules and Lamb dip effect cannot be observed [4]. If the laser frequency Ω, i.e., the frequency of the two opposite waves, coincides with the frequency of the absorption line centre ω_0, the oppositely travelling probe wave will interact with the same group of absorbing molecules having no motion component along the direction of the laser beam. Thus, the probe wave will interact with the group of molecules saturated by the intense saturate wave, and therefore the absorption of the probe wave will decrease near the frequency of the absorption line centre ω_0. As a result sharp peak in probe wave transmission (so-called inverted Lamb dip) can be observed on a laser gain profile as the laser frequency is tuned.

In this paper the experiment with Lamb dip spectroscopy has been performed for sulfur haxafluoride as an examplary absorber and a tunable low power cw CO_2 laser as a spectrometer. The experiment has been carried out for P12, P14, P16, P18 and P20 laser emission lines of 10.6 μm band, i.e., for those ones coinciding with SF_6 absorption lines.

2. Experiment and its results

A homemade cw CO_2 laser described previously in paper [5], has been used in the experiment. In the laser used for the spectroscopic measurements a diffraction grating was placed instead of one of the mirrors. The laser produced an output power of approximately 2 W per one preselected emission line. The cavity on the opposite side of the grating was terminated by the concave ($R = 10$ m) gold-coated mirror with transmission hole 2.5 mm in diameter [6]. To adjust the position of the coupling-out hole mirror a piezoelectric transducer PZT was used [7]. The laser was tuned by means of saw-tooth signal from an oscilloscope (Fig. 1).

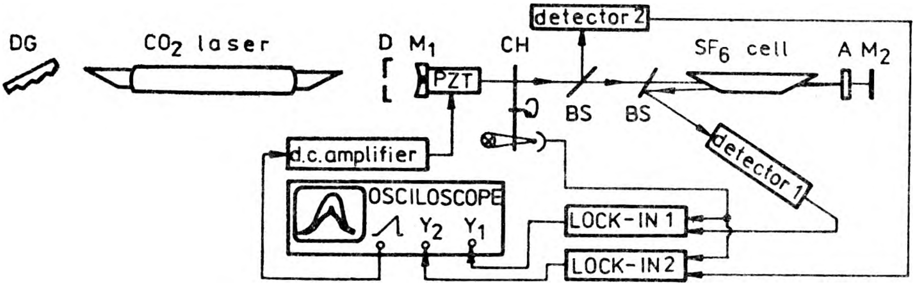

Fig. 1. Experimental setup: DG – diffraction grating, D – diaphragm, CH – chopper (283 Hz), BS – beam splitter, A – attenuator, M_1 – output laser mirror, M_2 – totally reflecting mirror

The setup of the laser spectrometer consisted of a 40 cm-long glass absorption cell of diameter of 2.5 cm filled with SF_6 and terminated with NaCl Brewster-angle windows. Intense output laser beam, as the saturate wave, fell onto the absorption cell. According to the theoretical investigations [8], in order to obtain maximal slop of the first derivative of absorption peak the intensity of the saturate wave should be approximately twice higher than that of absorption saturation. This is a necessary condition both for the achievement of higher resolution of the first-derivative method of spectroscopy (like in EPR spectrometers [9]) or for obtaining maximal frequency stability of output laser radiation during the laser stabilization to the zero of the first-derivative signal [10].

The measured absorption saturation intensity of SF_6 amounted approximately to 3.5–6.5 $\mathrm{W} \times \mathrm{cm}^{-2}\,\mathrm{Torr}^{-1}$ for P12–P20 emission lines of 10.6 μm band [11]. Spectroscopic measurements have been performed for intensity of the saturate wave of about 1 $\mathrm{W} \times \mathrm{cm}^{-2}$ under pressure of 80 mTorr for SF_6.

The saturate wave, having passed the absorption cell was reflected from totally reflecting mirror in the opposite direction and having been attenuated in a silicon attenuator, returned through the absorption cell as the weak probe wave. As it is known, the probe wave should be attenuated repeatedly with respect to the saturate wave in order to obtain contrastive peaks. In our experiment the probe wave was attenuated four times. The saturate wave and the probe one were monitored with non-cooled CdHgTe detectors and simultaneously observed with an oscilloscope, whereas the laser frequency was scanned in the $c/2L$ range by means of piezoceramic transducer. In our case the mode distance was $c/2L = 140$ MHz (for the resonator length $L = 1.07$ m), which allowed the voltage-frequency scaling of the piezoceramic transducer. This step was necessary for location of the absorption peaks in relation to the laser emission line centre.

The examples of such absorption peaks obtained at P16, P18 and P20 laser emission lines of 10.6 μm band can be seen in Figs. 2a, b, c. The observed peaks have been registered simultaneously along the CO_2 laser emission lines (upper

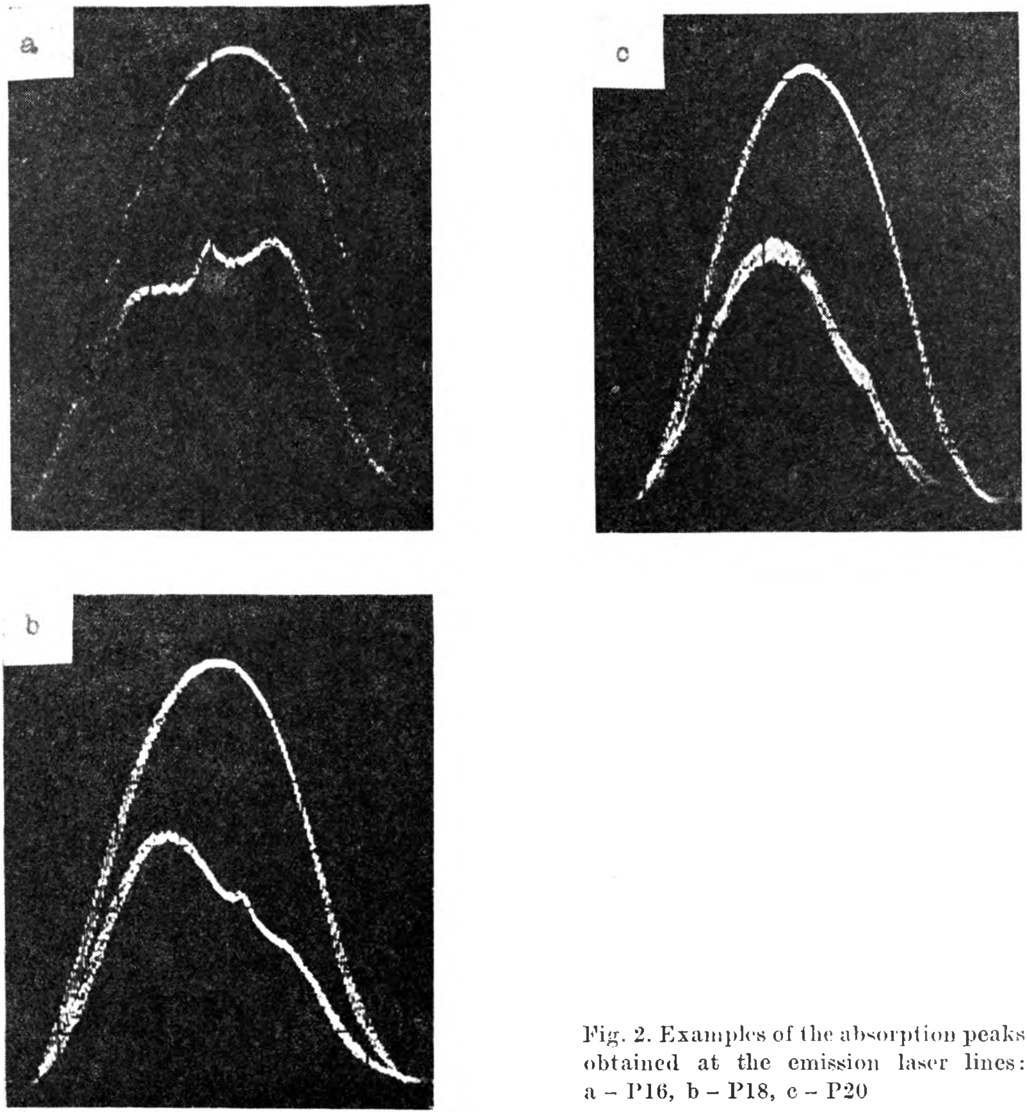

Fig. 2. Examples of the absorption peaks obtained at the emission laser lines: a – P16, b – P18, c – P20

curve) which permitted us to measure the pump offset in frequency scale (Table 1). The absorption peaks have been observed at five CO_2 laser lines. If, however, the first-derivative method was used, two absorption peaks at P12 emission line of CO_2 laser, a peak at P14 line and some additional peaks at P16 and P20 lines have been observed.

The SF_6 absorption lines coinciding with the respective CO_2 laser emission lines are shown in Table 2 [12–15].

Table 1. The locations of the SF_6 absorption peaks at the CO^2 laser lines

CO_2 emissive laser line	Frequency shift of an absorption peak in relation to the laser line center, [MHz]	
	Present work	Ref. [18]
P12	$+15(\pm 1)$*	—
	$-15(\pm 1)$*	—
P14	$+20(\pm 1)$*	—
P16	$+11(\pm 1)$*	$+10.5$
	$-8(\pm 1)$	-5.0
P18	$+12(\pm 1)$	$+9.5$
P20	$+26(\pm 1)$	$+10.0$
	$-35(\pm 1)$*	-13.0

* the peaks identified by means of the first-derivative method

Table 2. The SF_6 absorption lines coinciding with the respective CO_2 laser emission lines

CO_2 emissive laser line	SF_6 absorption line of band			
	[12]	[13]	[14]	[15]
P12	—	—	R70	—
P14	R25 ± 1	R30	R28	R28
P16	Q54 ± 10	—	—	—
P18	P40 ± 2	P34	P33	—
P20	P69 ± 3	—	P59	P59

3. Conclusions

The results of the above experiment indicate that a single-mode tunable cw CO_2 laser may be a useful radiation source in a laser spectrometer which allows the investigation of fine structure of molecular systems with high resolution. Unfortunately, the band width of such a laser spectrometer is limited to the tuning range of the laser frequency, i.e., to the Doppler broadening of a CO_2 laser line (about 40–70 MHz in this experiment). The band of the considered laser spectrometer may be increased by using either a tunable cw CO_2 laser with the mixture of a few isotopes of CO_2 molecule, or a high pressure waveguide CO_2 laser. In these cases, the tuning range of the laser frequency may be increased to a few hundred MHz [16] and to about 1 GHz [17], respectively.

Acknowledgements – The authors would like to thank Dr. K. M. Abramski and Dr. E. Matras who spent many hours discussing the subject and greatly contributed to the realization of the experiment.

References

[1] Brever R. G., Science **178** (1972), 247.
[2] Feld M. S., Letokhov V. S., Sc. Am. **229** (1973), 69.
[3] Letokhov V. S., Opt. Laser Techn. **2** (1979), 13.
[4] Lamb W. R., Jr, Phys. Rev. **134** (1964), 1429.
[5] Pliński E. F., Abramski K. M., Optica Applicata **14** (1984), 301.
[6] Pliński E. F., Optica Applicata **14** (1984), 333.
[7] Abramski K. M., Godziński Z., Pomiary, Automatyka, Kontrola (in Polish), **12** (1978), 452.
[8] Pliński E. F., Doctor's Thesis, Report No. *128*P–008/83, Institute of Telecommunication and Acoustics, Technical University of Wrocław, Wrocław 1983.
[9] Kęcki Z., *Podstawy spektroskopii molekularnej* (in Polish), PWN, Warszawa 1975.
[10] Wallard A. J., J. Phys. E: Sci. Instr. **6** (1973), 793.
[11] Brunet H., IEEE J. Quant. Electron, QE–**6** (1970), 678.
[12] Houston P. L., Steinfeld J. I., J. Mol. Spectr. **54** (1975), 335.
[13] Novak A. V., Lyman J. L., J. Quant. Spectr. Radiat. Transf. **15** (1975), 945.
[14] Brunet H., C. R. Acad. Sci. (Paris) **264** (1967), 1721.
[15] Borde Ch., Ouhayoun M., J. Mol. Spectr. **73** (1978), 344.
[16] Beterov I. M., Chebotayev V. P., Provorov A. S., Opt. Commun. **7** (1973), 410.
[17] Bazarov E. N., Gerasimov G. A., Sazanov A. I., Kvant. Elektron. (in Russian) **6** (1979), 582.
[18] Rabinowitz P., Keller R., LaTourrette J. T., Appl. Phys. Lett. **14** (1969), 376.

Received August 24, 1984

Нелинейная лазерная спектроскопия в применении для SF_6

В работе представлены исследования по провалу Ламба в поглощающей среде гексафлуорида серы при использовании CO_2 лазера малой мощности. Представлены расположения пиков поглощения относительно центра эмиссионных линий P12, P14, P16, P18 и P20 полосы 10,6 μм CO_2 лазера.